

Naturally Occurring Radioactive Material (NORM VII)

Proceedings
of an International Symposium
Beijing, China, 22–26 April 2013



IAEA

International Atomic Energy Agency

NATURALLY OCCURRING
RADIOACTIVE MATERIAL
(NORM VII)

The Agency's Statute was approved on 23 October 1956 by the Conference on the Statute of the IAEA held at United Nations Headquarters, New York; it entered into force on 29 July 1957. The Headquarters of the Agency are situated in Vienna. Its principal objective is "to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world".

PROCEEDINGS SERIES

NATURALLY OCCURRING
RADIOACTIVE MATERIAL
(NORM VII)

PROCEEDINGS OF THE
SEVENTH INTERNATIONAL SYMPOSIUM ON
NATURALLY OCCURRING RADIOACTIVE MATERIAL
ORGANIZED BY THE
CHINA INSTITUTE OF ATOMIC ENERGY AND THE
NUCLEAR AND RADIATION SAFETY CENTRE,
MINISTRY OF ENVIRONMENTAL PROTECTION, CHINA,
IN COOPERATION WITH THE
INTERNATIONAL ATOMIC ENERGY AGENCY
HOSTED BY THE
CHINA ATOMIC ENERGY AUTHORITY AND THE
NATIONAL NUCLEAR SAFETY ADMINISTRATION, CHINA
AND HELD IN BEIJING, CHINA,
22–26 APRIL 2013

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2015

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email: sales.publications@iaea.org
<http://www.iaea.org/books>

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Printed by the IAEA in Austria

January 2015

STI/PUB/1664

IAEA Library Cataloguing in Publication Data

Naturally occurring radioactive material (NORM VII) : proceedings of the seventh International Symposium on naturally occurring radioactive material / organized by the China Institute of Atomic Energy and the Nuclear and Radiation Safety Centre, Ministry of Environmental Protection, China in cooperation with the International Atomic Energy Agency, hosted by the China Atomic Energy Authority and the National Nuclear Safety Administration, China and held in Beijing, China, 22-26 April 2013. — Vienna : Vienna : International Atomic Energy Agency, 2015.

p. ; 24 cm. — (Proceedings series, ISSN 0074-1884)

STI/PUB/1664

ISBN 978-92-0-104014-5

Includes bibliographical references.

1. Radioactive substances — Congresses. 2. Ionizing radiation — Dosage — Congresses. 3. Radiation protection. I. International Atomic Energy Agency. II. Series: Proceedings series (International Atomic Energy Agency).

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14-00945

FOREWORD

All minerals and raw materials contain radionuclides of natural origin, of which the most important for the purposes of radiation protection are the radionuclides in the ^{238}U and ^{232}Th decay series. For most human activities involving minerals and raw materials, the levels of exposure to these radionuclides are not significantly greater than normal background levels. Such exposures, while having been the subject of much investigation, are not of concern for radiation protection. However, certain work activities can give rise to significantly enhanced exposures that may need to be controlled by regulation. Material giving rise to these enhanced exposures has become known as naturally occurring radioactive material (NORM).

Historically, most regulatory attention has been focused on the mining and processing of uranium ore, because such activities are a direct consequence of the radioactivity in the ore and form part of the nuclear fuel cycle. Since the 1990s, however, more and more States have introduced measures to regulate exposures arising from a wider range of natural sources, in particular minerals and raw materials other than those associated with the extraction of uranium. Two important developments in this regard are the establishment of IAEA Safety Standards No. GSR Part 3, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards (initiated in 1996 as IAEA Safety Series No. 115) and the European Council Directive 96/29/Euratom of 13 May 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation — both of which contain provisions for protective measures against significantly increased exposures of workers and members of the public to natural sources.

As a direct consequence of the European Council Directive 96/29/Euratom and its possible implications for non-nuclear industries in Europe, a symposium on NORM, the first in the current series, was held in Amsterdam, the Netherlands, in 1997. The second in the series (NORM II) was held in Krefeld, Germany, in 1998; the third (NORM III) in Brussels, Belgium, in 2001; the fourth (NORM IV) in Szczyrk, Poland, in 2004; the fifth (NORM V) in Seville, Spain, in 2007; and the sixth (NORM VI) in Marrakesh, Morocco, in 2010. In addition, the second International Symposium on Technologically Enhanced Natural Radiation was held in Rio de Janeiro, Brazil, in 1999, reflecting the growing interest within regions beyond Europe in the management of exposure to NORM.

The close involvement of the IAEA in most of these symposia is reflected in the fact that the proceedings of the Rio de Janeiro and Szczyrk symposia were published as IAEA-TECDOC-1271 and IAEA-TECDOC-1472, respectively, while the proceedings of the Seville and Marrakesh symposia were published in the IAEA Proceedings Series. In the case of NORM VII, the IAEA entered

into a formal cooperation arrangement with the organizers (as it had done for other recent symposia in the series), in terms of which the IAEA, in addition to publishing these Proceedings, served on the Steering Committee and Scientific Committee and provided financial support to several participants from Member States eligible to receive assistance under the IAEA technical cooperation programme.

The NORM VII symposium was attended by 120 participants from 28 countries and 25 observers from Chinese universities and institutions. It provided an important opportunity to review the developments that had taken place during the three year period since the Marrakesh symposium, in 2010. This period was characterized by ongoing activities to revise international standards on radiation protection and safety and the further implementation of these standards in many countries. These Proceedings contain 48 papers accepted for oral presentation and 4 rapporteur reports, as well as a summary, which concludes with the main findings of the symposium. Text versions of 19 poster presentations are provided on a CD-ROM which accompanies these Proceedings.

The IAEA, on behalf of the organizers, the China Institute of Atomic Energy and the Nuclear and Radiation Safety Centre, Ministry of Environmental Protection, China, gratefully acknowledges the cooperation and support of all the organizations and individuals that have contributed to the success of the symposium. In particular, the assistance provided by the following organizations is acknowledged: the China Society of Radiation Protection; the Commission of Nuclear Safety and Environmental Radiation Safety, Chinese Society for Environmental Sciences; the National Institute for Radiological Protection, Chinese Centre for Disease Control and Prevention; Tsinghua University; the China University of Geosciences; and the University of South China. The IAEA officer responsible for this publication was P.P. Haridasan of the Division of Radiation, Transport and Waste Safety.

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SUMMARY

1. BACKGROUND TO THE SYMPOSIUM

1.1. Objectives

This symposium, the seventh in a series of symposia on NORM, once again provided an important opportunity to review recent technical and regulatory developments concerning exposure to NORM. The symposium brought together experts from a wide range of countries to report on and discuss the progress made in identifying, quantifying and managing the radiological risks associated with industrial processes involving NORM. The revision of international standards, which was completed during the period since the last NORM symposium in 2010, provided an important backdrop to the presentations and discussion.

As with previous symposia in this series, the technical programme was well subscribed, with 48 contributions being accepted as oral presentations for publication in these Proceedings. There were also many contributions in the form of posters. To help realize the objectives of the symposium, arrangements were made in the programme for each day's presentations and discussions to be reviewed and summed up by a rapporteur. On the final day of the presentations, the rapporteur's report included a review of the entire symposium and of the extent to which the objectives of the symposium were met.

1.2. International aspects

The first NORM symposium, held in Amsterdam, the Netherlands, in 1997, had been organized in response to concerns within the non-nuclear industry in the European Union that the implementation of a new European Council Directive (Directive 96/29/Euratom of 13 May 1996 laying down basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation) would place unreasonable and unwarranted legal obligations on many industrial enterprises that handled and processed material containing low levels of radionuclides of natural origin. Subsequently, as new regulations for the control of exposure to NORM became established in EU Member States and as knowledge about levels of exposure improved, those concerns diminished to some extent, although the definition of the scope of regulation remained controversial. Furthermore, it became apparent that this was becoming more of a global issue because of the increasingly international profile of the mining and mineral processing industry, with large quantities of minerals being mined and beneficiated in countries far from Europe and shipped to other

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countries — often over vast distances — for further processing. In line with this trend, successive NORM symposia began to take on a more international flavour and the involvement of the IAEA became progressively greater.

Given this background, it was fitting that China — a major source, processor, user and supplier of industrial minerals, many of which contained elevated concentrations of radionuclides of natural origin — was chosen as the venue for the NORM VII symposium. The planning of the symposium followed an approach similar to that adopted for NORM V and NORM VI, in that steps were taken to encourage participation from all regions of the world. The steering committee arranged for broad international representation on the scientific committee of the symposium and encouraged the members of that committee to actively promote participation in the symposium from within their own geographic regions. Furthermore, the IAEA provided financial support to nine participants from Member States eligible to receive assistance under the IAEA technical cooperation programme. These efforts were evidently successful in that the symposium attracted 120 participants from 28 countries. While the high level of participation by individuals from regulatory authorities and scientific institutes was encouraging, it was noted that, once again, there were relatively few participants from NORM industries.

2. RADIATION PROTECTION STANDARDS AND REGULATORY APPROACHES

2.1. Revision of the international standards

In line with the approach adopted at NORM V and NORM VI, the keynote address delivered in the opening session was aimed at establishing the overall background for the symposium by describing the international standards published by the IAEA and their application to natural sources. At the time of the NORM VI symposium, in 2010, a revision of the IAEA Basic Safety Standards (BSS) was in the final stages of development. An updated version of the BSS — IAEA Safety Standards Series No. GSR Part 3 (Interim) — was published by the IAEA in

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the following year.¹ The revised BSS reflected the latest recommendations of the International Commission on Radiological Protection (ICRP). Participants were reminded of how the general approach to radiation protection had changed from one based on the concepts of ‘practices’ and ‘interventions’ to one based on three types of exposure situation — planned exposure situations, emergency exposure situations and existing exposure situations. Most exposures to natural sources were treated as existing exposure situations but some, including exposures associated with certain industrial activities involving minerals and raw materials, had to be treated as planned exposure situations.

The new radiation protection approach placed greater emphasis on the concept of optimization of protection, regardless of the type of exposure situation. For existing exposure situations, a new approach based on ‘reference levels’ had been adopted. These reference levels were conceptually different from the ‘action levels’ used previously and were relevant to natural sources. The requirement for optimization applied below the reference level as well as above it — this was a new development and concern was expressed during the symposium as to the practicality of implementing this in some real life situations, for instance in the remediation of contaminated sites where there was a tendency to set a very low reference level of 1 mSv/a. There was now a greater use of quantitative criteria for controlling exposure to natural sources, particularly in planned exposure situations when defining the scope of regulatory control in terms of exemption and clearance. In addition, greater emphasis was being placed on the graded approach to regulation, although it appeared that this concept was still not universally understood in a consistent manner.

New recommendations and requirements for exposure to radon were now in place, and these reflected the increased risk coefficient of 8×10^{-10} per $\text{Bq} \cdot \text{h} \cdot \text{m}^{-3}$ (nearly double the previous value) recently recommended by the ICRP. There were still some questions about the reliability of the new risk coefficient, and there was concern about the implications for certain underground mines in which radon levels were significantly elevated and may be difficult or impossible to reduce further. The new requirements placed greater emphasis on the establishment of a national radon policy, with a reference level of 10 mSv/a (presently equivalent to a radon concentration of 300 Bq/m^3 , although this could change). The ICRP

¹ [Editor’s note] The BSS has since been updated: EUROPEAN COMMISSION, FOOD AND AGRICULTURE ORGANIZATION OF THE UNITED NATIONS, INTERNATIONAL ATOMIC ENERGY AGENCY, INTERNATIONAL LABOUR ORGANIZATION, OECD NUCLEAR ENERGY AGENCY, PAN AMERICAN HEALTH ORGANIZATION, UNITED NATIONS ENVIRONMENT PROGRAMME, WORLD HEALTH ORGANIZATION, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, IAEA Safety Standards Series No. GSR Part 3, IAEA, Vienna (2014).

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recommend that, for workplaces, the radon level should be optimized to a value below 300 Bq/m³. If this was not feasible, then the dose criterion of 10 mSv/a should be applied. In workplaces for which the dose from radon continued to exceed 10 mSv/a, the workers should be considered as occupationally exposed and subject to the requirements for planned exposure situations, including a dose limit of 20 mSv/a. More emphasis was now being placed on the minimization of radon levels in new buildings through appropriate design and construction. This strategy needed to be integrated with strategies for general indoor air quality and energy savings.

2.2. Regulatory approach

During the course of the symposium, it became evident that the assignment of the correct type of exposure situation was often a source of confusion and this continued to lead to inconsistencies in the regulatory approach. Despite the greater use of quantitative criteria for defining the scope of regulatory control, as well as a greater emphasis on the graded approach to regulation, there were still many instances of activities involving minerals and raw materials being over-regulated or regulated unnecessarily. A consistent regulatory approach to natural sources was still lacking in many States, although several instances were reported of States now actively developing or updating their regulations, along with associated technical databases and guidelines. These included States such as China and Brazil, which have particularly extensive and diverse industrial activities involving minerals and raw materials. It was noted that many States do not yet have adequate regulations covering the disposal of NORM waste. With regard to the development of new standards and regulations, several presenters specifically referred to the graded approach and some described initiatives to encourage the use of NORM residues. The symposium highlighted the importance of having to consider all hazards from NORM — radiation exposure was only one element in the overall approach to health and safety in NORM industries and the control of other hazards, such as the presence of heavy metals, was often more important.

It was clear from the presentations and discussions that the control of exposure to natural sources was now generally accepted as being unwarranted for materials with radionuclide activity concentrations below 1 Bq/g or for practices giving rise to doses below 1 mSv/a. However, exemption mechanisms for NORM were often poorly defined in national regulations or absent altogether, posing a dilemma for the regulatory body. It was not always appreciated that exemption applied only to the control of practices in planned exposure situations and not to existing exposure situations (i.e. the use of building materials containing radionuclides of natural origin). Furthermore, when considering the exemption

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of practices involving NORM, there was a tendency for regulatory bodies to make the mistake of using the activity concentration criterion of 1 Bq/g as an exemption level (or even as a limit), rather than using a dose criterion of the order of 1 mSv/a as set out in the BSS. It was not fully appreciated that practices qualifying for exemption in accordance with the BSS (i.e. on the basis of dose) might well involve materials with activity concentrations significantly above 1 Bq/g (often up to 10 Bq/g and in some cases much higher). The problem appeared to have arisen for two reasons. Firstly, there was a tendency to disregard the 1 mSv/a criterion because it required a dose assessment to be carried out, which was not as straightforward as simply measuring the activity concentration. Secondly, the true purpose of the 1 Bq/g criterion — namely, for applying the requirements for planned exposure situations rather than those for existing exposure situations (and also for approving clearance of material from regulatory control) — was widely misunderstood. Problems such as this led to the general impression that the new international standards were too complicated, particularly in their application to natural sources, and were thus easily misinterpreted by regulatory bodies.

3. INDUSTRIAL ACTIVITIES INVOLVING NORM

Among the various presentations, there were many references to Safety Reports Series No. 49, Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials. That report identified 12 industrial activities that should be considered with respect to the control of exposure to natural sources:

- (1) Mining and processing of uranium ore;
- (2) Extraction of rare earth elements;
- (3) Production and use of thorium and its compounds;
- (4) Production of niobium and ferroniobium;
- (5) Mining of ores other than uranium ore;
- (6) Production of oil and gas;
- (7) Manufacture of titanium dioxide pigments;
- (8) The phosphate industry;
- (9) The zircon and zirconia industries;
- (10) Production of tin, copper, aluminium, zinc, lead, iron and steel;
- (11) Combustion of coal;
- (12) Water treatment.

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From the presentations and discussion, it was evident that this list, in one form or another, is now widely recognized internationally. It was encouraging to note that all 12 of the industrial activities on the list were specifically referred to during the course of the symposium. In addition, no new types of industrial activity were identified as needing to be added to the list, although from the Chinese experience it was evident that vanadium production needed to be included in item 10. These findings suggest that the list of industrial activities in Safety Report Series No. 49 remains both relevant and complete. The information presented during the symposium confirmed the need to adopt an industry specific approach and many references were made to the suite of five IAEA publications in the Safety Reports Series dealing with individual NORM industries.

Important new information was presented concerning mining and mineral processing activities in China as a result of a national survey of facilities producing ore, ore concentrates and waste, conducted during the period 2006–2009. These facilities were associated with the production of rare earths, niobium–tantalum, zircon, tin, lead–zinc, copper, vanadium, aluminium, phosphate, coal, iron and steel. The presence of NORM had been identified in each of these types of industry. The facilities included 876 mining companies (the most numerous of which were coal mines) with a total annual production of 267 million t of ore. The survey also included 587 facilities that annually processed a total of 191 million t of feedstock and generated a total of 171 million t of solid waste and 831 million t of wastewater. About 3% of that wastewater (24.6 million t) had a gross alpha activity concentration exceeding 1 Bq/L or a gross beta activity concentration exceeding 10 Bq/L.

Information presented at the symposium provided further evidence to confirm that most workers in operating facilities involving NORM receive doses less than 1 mSv/a excluding radon. This was also true of workers engaged in the remediation of NORM contaminated sites. Greater use was being made of on-site measurements for dose assessment purposes, although modelling was still heavily relied upon and would continue to play an important complementary role as long as it was conducted using realistic assumptions. Doses received by workers from the inhalation of radon continued to be low or insignificant except in underground mines where newly available measurement data showed large numbers of underground workers exposed to high levels of radon. Reports suggested that attention to ventilation in underground mines had been very effective in reducing radon concentrations. The presentations also provided confirmation that doses received by members of the public from industrial activities involving NORM remain very low and usually insignificant. The following is a summary of worldwide information presented at the symposium relating to the operation of various types of NORM industrial facility.

3.1. Mining and processing of uranium ore

This industry is undergoing a revival and many States with no previous experience of uranium production are now entering the scene. With uranium production having been in the doldrums for many years, this industry has now been beset by a shortage of professional skills, including radiation protection skills. In situ leaching is growing in popularity. Compared with traditional mining techniques, this technique has fewer implications for radiation exposure. For instance, it does not lead to the generation of large volumes of mine residues. Some new uranium mining ventures are characterized by the co-production of other minerals such as rare earths and phosphate. Many States continue to treat the mining and processing of uranium ore from a nuclear fuel cycle perspective rather than as a NORM activity. This tends to result in differences in regulatory approach that are both unwarranted and undesirable.

3.2. Rare earths

Worldwide production of rare earths is currently dominated by China, although several other sources of production, some of them in remote parts of the world such as Greenland, are now being investigated and established. The technology for the separation and purification of rare earths, including the removal of radionuclides, continues to be developed, and the recovery of uranium as a by-product is under consideration. More radiological data are becoming available from Chinese rare earths operations. Average worker doses associated with rare earths production reported at the symposium were less than 1 mSv/a for gamma exposure, while doses from the inhalation of radon progeny were far below the reference level of 10 mSv/a. Public doses associated with rare earths production were insignificant. Disposal solutions still needed to be established for NORM residues at Chinese rare earth facilities — these residues were currently kept in temporary storage. A recent national survey found evidence of significant radioactivity concentrations in wastewater at some rare earth facilities.

3.3. Production of niobium and ferroniobium

The average ^{238}U activity concentrations associated with columbite–tantalite artisanal mining and processing activities in Rwanda were 0.6 Bq/g for the ore and of the order of 1–2 Bq/g for processed material. The maximum worker dose was 0.5 mSv/a.

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3.4. Mining of ores other than uranium ore (China)

Mines in China employ some 10.5 million workers, of which 6.5 million work in coal mines, 1 million work in metal mines and 3 million work in other mines. A survey of 44 mines (40 of which were underground operations) indicated that radon concentrations in 15% of them were above 1000 Bq/m³. Radon levels in coal mines have been significantly reduced in recent years, with an average worker dose of currently less than 1 mSv/a, but radon levels in metal mines remain high because of inadequate ventilation, with an average worker dose of more than 5 mSv/a. The average worker dose due to radon in non-metal mines was much less than 1 mSv/a. Although uranium and thorium activity concentrations in coal were generally at the same levels as in normal soil (i.e. 0.04 Bq/g), it was reported that some coal deposits in China had activity concentrations of up to 5.6 Bq/g for ²³⁸U and 29 Bq/g for ²³²Th. There are concerns about environmental impacts. A survey of wastewater from Chinese coal mines is being conducted, but so far no radiological problems have been identified.

3.5. Mining of ores other than uranium ore (other States)

In the copper mining area of Zambia, mine water is discharged into surface water bodies which are used for the supply of drinking water. The quality of the water was investigated and no problems were identified. Underground radon levels in Zambian copper mines can exceed 1000 Bq/m³. At a copper mine in South Africa, the copper concentrate has an average ²³⁸U activity concentration of 1.4 Bq/g, while the average ²³⁸U and ²¹⁰Pb activity concentrations in the copper tailings are 3.5 and 8.8 Bq/g, respectively. A radiation protection programme has been in place for 20 years and now forms part of an integrated management system of which radiation is only one part. The doses received by workers are generally less than 1 mSv/a. Investigations are continuing at mines in Sierra Leone producing gold, tantalum, rutile and iron. The doses received by members of the public have been found to be less than 1 mSv/a.

3.6. Phosphate industry

Measurements at two ammonium phosphate plants in Spain showed that worker doses due to gamma radiation and dust inhalation were far below 1 mSv/a, with no significant exposure to radon. This confirms previous estimates for ammonium phosphate production. Measurements of the leachability of radionuclides from phosphogypsum and phosphate fertilizer showed that when such products were applied to the soil, there was no availability of these radionuclides to the environment.

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3.7. Production of zircon

Although Nigeria is not a major zircon producer, some zircon mining and beneficiation is practised in central Nigeria, including artisanal mining and panning conducted on a very small scale. Measurements showed unusually high activity concentrations in the zircon compared with normal commercial product — nearly 6 Bq/g for ^{238}U and nearly 29 Bq/g for ^{232}Th . High ^{238}U concentrations (27 Bq/g) have also been found in non-conductive material separated during the extraction of zircon and titanium dioxide from mineral sands in Madagascar. Controlled areas have been established and it appears that adequate radiation protection measures are in place.

3.8. Combustion of coal

The release of ^{210}Pb to the atmosphere and into the sea from a coastal coal fired power station in China was studied. The activity concentration in the wastewater was ten times greater than in normal seawater.

3.9. Water treatment

At a groundwater treatment plant in the Czech Republic, the activity concentration of ^{226}Ra in the filter sand was found to be 0.3–5.6 Bq/g, with an average of 1.8 Bq/g. It was demonstrated that periodic backwashing of the filter sand produces pronounced spikes in radon concentration in the air inside the filtration hall. The average radon concentration inside the hall was 141 Bq/m³, but the peak values were several times higher. In Morocco, the treatment of wastewater by infiltration through phosphate beds was investigated. It was found that radioactivity in the phosphate was transferred to the water, but the water quality remained acceptable.

4. NORM RESIDUES

The management of NORM residues continues to be a topic of intense interest. There was still a lack of harmonization of regulatory approaches to NORM residue management, including not only their disposal as waste but also their recycling and use as by-products. For the disposal of NORM residues as waste, there are indications that some progress is being made in identifying suitable disposal options and establishing the necessary facilities. Landfill sites, ranging from simple municipal waste sites to highly controlled facilities for hazardous waste, continued to be the preferred option for the disposal of most

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types of NORM waste. This was the case, for example, in Sweden, but only for NORM waste in which the radionuclide activity concentration did not exceed 10 Bq/g. More highly active waste, such as some types of scale and water treatment resin, had to be disposed of under licence at a special disposal facility. It was also mentioned that studies had been performed in Sweden on scrap metal contaminated with ^{226}Ra at an average activity concentration of 55 Bq/g, indicating low doses but a need to use volume reduction and to license scrap recycling facilities.

China, as the world's principle producer of rare earths, generated large volumes of NORM residues at its many rare earths production facilities. It was reported that residues from 11 such facilities had been held in temporary storage pending a disposal solution. The total amount stored was about 30 000 t, comprising 5000 t of acid dissolution waste and 25 000 t of neutralization waste. Evidence of environmental contamination was mentioned. There was a need for a suitable waste repository and improved regulations governing NORM residue disposal. It was also mentioned that waste treatment had the potential for large volume reduction and that recycling of material containing 1–2% thorium needed to be explored.

Another presentation from China addressed the disposal of waste from the front end of the uranium fuel cycle (referred to as UF waste) arising from the purification, conversion, enrichment and fuel fabrication steps. The waste comprised waste filter cores, resins, equipment and other residues containing mainly triuranium octaoxide, uranium oxide and uranium hexafluoride and was currently in temporary storage. Radioactive material associated with nuclear installations is excluded from the definition of NORM, but the question was posed as to whether UF waste should nevertheless be treated as a natural material (i.e. NORM waste). This was considered to be an important question in terms of the regulatory requirements governing the disposal of the material. The uranium activity concentrations were similar to those in many types of NORM waste (including waste from the mining and processing of uranium ore), while, in contrast to NORM waste, the decay products were absent (at least for the next thousand to million years). For these reasons, it was proposed that UF waste be treated as NORM waste, thus opening more options for disposal, such as in a uranium tailings dam.

There was evidence of a growing trend towards NORM residue recycling and use and this was now starting to be actively encouraged by some national authorities as opposed to being discouraged or even prohibited in the past. The recycling and by-product use of NORM residues was mentioned as being one of the key components of the newly emerging 'comprehensive extraction' concept described in the symposium. However, some national authorities had not yet recognized the acceptability (and desirability) of blending NORM residues with

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lower activity material to facilitate their recycling and use. It was also pointed out that the use of NORM residues as by-products could be hindered by the presence of non-radiological constituents such as heavy metals.

The use of phosphogypsum as a by-product continues to attract attention. Applications mentioned in the symposium included its use in agriculture and building construction, as a landfill cover and as a source of sulphur and rare earths. It was reported from Brazil that a standard for the use of phosphogypsum was being developed. Phosphogypsum is also the subject of ongoing studies in Brazil — one presentation described work to characterize the elemental distribution of uranium and other heavy metals in phosphogypsum, indicating that such elements tended to reside in the more mobile fine fraction of the material.

5. NORM IN BUILDING MATERIALS

There was growing support for the use of NORM residues such as fly ash, steel slag, phosphogypsum and bauxite processing residue (red mud) in building construction materials in order to conserve natural resources and reduce the amounts of NORM residues requiring disposal as waste. It was reported that the European Union would now be encouraging such use of NORM residues, subject to the development of suitable mechanisms for regulatory approval. There was general agreement on a dose criterion (reference level) of 1 mSv/a, based on external gamma exposure, but practical approaches to the use of NORM residues in building materials had not yet been harmonized. The ‘activity concentration index’ (ACI) approach, based on a combination of the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , had been adopted throughout Europe and, in one form or another, in many other countries (it should be noted that the ACI applies to the building material itself, not to individual constituents). However, it was not always appreciated that the role of the ACI in the decision making process was limited, since the index was merely a screening tool based on very conservative assumptions. Decisions on whether to allow the use of a particular building material could turn out to be wrong if they were based only on the value of the ACI. If the limiting ACI value was exceeded, a situation specific dose assessment needed to be carried out before making the final decision. The radiological implications for building occupants continued to be a highly sensitive issue when considering the use of NORM residues in building materials. The prospect of an increase in such use raised particular concerns for radon exposure, and there are still some questions about whether radon needed to be controlled separately using an additional criterion.

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In Brazil, guidelines were being developed for the use of phosphogypsum in building materials, based on the modelling of indoor radon exposure and a 1 mSv/a dose criterion. Radon exhalation from phosphogypsum bricks and panels was found to be two orders of magnitude lower than that from phosphogypsum stacks.

NORM residues are regarded as an important constituent of building materials in China. Smelting slag from the co-production of rare earths, iron and steel has been used in building materials for homes, causing a small increase in the average gamma dose received by the occupants (from 1.86 to 2.0 mSv/a). Activity concentration guidelines for the use of NORM residues in building construction have been developed using the ACI approach and material has been classified into three categories, depending on whether the dose is below 0.5 mSv/a (free use), between 0.5 and 1 mSv/a (use restricted to roads, bridges, dams or, with dilution, low occupancy buildings) or above 1 mSv/a (prohibited use). Coal ash, coal slag and smelting slag were among the materials investigated. It was found that 97.2% of the materials qualified for free use, 2.3% qualified for restricted use and 0.5% were prohibited.

6. TRANSPORT OF NORM

The doses received by workers in the transport of process materials and products from the mineral sands industry were assessed in a four year study. The dose assessment was based on measurements made in actual transport situations within Australia, between Australia and China and between Australia and Japan, involving transport by road, rail and sea. The mode of transport made little difference to the dose received. An exposure period of 1200–1400 h/a was considered to be a representative exposure period and the highest dose assessed from the measurement data was 0.739 mSv/a. It was concluded that the criterion for application of the IAEA Transport Regulations to NORM (ten times the activity concentration for exempt material) was consistent with a dose of about 1 mSv/a and was therefore appropriate.

Portal monitors for detecting radioactive material at ports in the Republic of Korea were giving many false alarms. When an alarm was triggered, there were often difficulties in tracing the owner of the consignment and the operating personnel were unsure of what action to take. Attempts were made to optimize the system by setting the trigger level as a function of the background standard deviation, which was not a fixed parameter. The main reason for the alarm being triggered was the presence of NORM in the form of refractories and processed stone products. Operator training was needed. A second, higher trigger level

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(an investigation level) was introduced in order to separately identify situations requiring immediate action. The system is still under review.

7. LEGACY SITES AND REMEDIATION

Several examples of legacy sites and remediation projects were described in the presentations. One such example related to a very large remediation project in Germany at a former uranium production facility operated by the Wismut company (one of many uranium legacy sites around the world, some of which have also been remediated or are in the process of remediation). Most of the material at the Wismut site had an activity concentration of less than 1 Bq/g, but the site was close to populated areas. The average dose to the local population from the unremediated site was 2 mSv/a, of which 1.7 mSv/a was due to radon. The remedial action was justified more on the basis of legal requirements, non-radiological hazards and stakeholder interests rather than on pure radiological considerations. The characteristics of the situation, including the existence of strong public concerns, led to the adoption of a very low reference level (1 mSv) which, in contrast with practice elsewhere, included exposure to radon. Consequently, the costs of remediation were high. Doses received by remediation workers were generally 0.5–2 mSv/a, although workers handling chemical processing materials with activity concentrations of up to 10 Bq/g received doses of up to 4 mSv/a. Underground workers preparing for mine flooding received the highest doses (up to 10 mSv/a in the absence of optimized ventilation). Doses received by the local population during the remediation were less than 1 mSv/a. Radon barriers were constructed over the remediated tailings dams but their effectiveness declined as a result of biointrusion (roots and burrowing animals) and the doses at the ‘toes’ of the tailings dams increased from the target value of less than 1 mSv/a to values of 2–3 mSv/a. The continued presence of mine voids is causing high radon concentrations in nearby homes. It was concluded that an optimized central ventilation system was the best long term solution to deal with this problem.

Another presentation described the remediation of a legacy site in the United Kingdom which had been used for the manufacture of thorium containing gas mantles and magnesium–thorium alloys. A site survey was conducted using gamma probes capable of detecting contamination buried as deep as 2 m, surface contamination measurements, trial excavations and analysis of samples. Thorium-232 concentrations were found to be 0.1–100 Bq/g and occasionally up to 500 Bq/g. Surface gamma measurements revealed dose rates of up to 20 μ Sv/h while whole body dose rates were up to 5 μ Sv/h. A very low reference level of 0.3 mSv/a is commonly applied in the United Kingdom, requiring a cleanup level

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of less than 0.1 Bq/g for future residential land (this was only just achievable) and less than 0.5 Bq/g for partial remediation of an operating site. The radiation protection programme ensured that the remediation workers received doses of less than 0.1 mSv/a from external gamma radiation (compared with predictions of 0.1–1 mSv/a) and less than 0.1 mSv/a from internal exposure (compared with predictions of 0.2–2 mSv/a). Waste with an activity concentration of more than 5 Bq/g was sent to a controlled disposal site. Waste in the range of 0.5–5 Bq/g was classified as exempt NORM waste and sent to a conventional landfill site. Waste with an activity concentration of less than 0.5 Bq/g was classified as ‘out of scope’ material and was left on site or recycled.

In the remediation of a former rare earths production site in China, a soil cleanup level of 0.5 Bq/g for ^{232}Th and for ^{228}Ra was chosen. The depth of contamination was 1.5 m on average, 3 m maximum. NORM waste with an activity concentration of less than 1 Bq/g was classified as exempt waste and backfilled on site. About 35 000 m³ of NORM waste in the range 1–20 Bq/g was sent for municipal landfill disposal. A further 3000 m³ of NORM waste in the range 20–1000 Bq/g was sent to an engineered surface disposal facility. Finally, 99 m³ of NORM waste with an activity concentration exceeding 100 Bq/g was sent for deep geological disposal.

A legacy site in Belgium had been used for the processing of pyrochlore to produce ferroniobium. Thorium containing slag and contaminated soil were removed. The slag was sent to a hazardous waste landfill site that was controlled with a 1 mSv/a limit. Much of the slag had a low activity concentration and was only sent to the hazardous waste site because of its chemical constituents. The soil was contaminated to a depth of 0.75–1.5 m and contained hotspots with activity concentrations of about 8 Bq/g (^{238}U decay series) and 32 Bq/g (^{232}Th decay series) due to buried slag. Groundwater contamination was found. There was no increase in radon levels on the site or in the buildings.

In Austria, an old radium facility in an urban environment was remediated to make way for a new school to be built nearby. Hotspots containing ^{226}Ra at 40 Bq/g and more were found on concrete and brick surfaces. High radon levels were measured, which persisted after demolition of the building because they were due to radium contamination buried in the soil to a depth of several metres. The remediation of the ground entailed the removal of 2000 t of soil with a ^{226}Ra activity concentration of 7 Bq/g and of 200 barrels of soil with a ^{226}Ra activity concentration of 8–200 Bq/g.

The remediation of a former phosphate processing facility at a coastal location in Tunisia was described. Although the material on site (essentially phosphogypsum) had a low activity concentration, the location was sensitive and there was great potential for use of the land for recreation purposes including

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the re-establishment of beaches. The remediation was therefore justified on this basis. The phosphogypsum was retained on site in an engineered containment.

Hotspots of contamination were identified during a pre-remediation site survey of a former coal fired power plant in Croatia. The heavy metals content was considered to be of greater concern than the radionuclide content. The goal was to make use of the residues rather than dispose of them.

In Spain, acid mine drainage associated with former mining activities in a pyritic area over the past 150 years had caused the mine pit lakes to become contaminated with uranium (possibly exceeding 1 Bq/L) and heavy metals. This was influencing the water quality in the estuary of a nearby river some 100 km downstream.

From the remediation examples described above, it could be concluded that reference levels tended to be set at (or below) the lower bound of the 1–20 mSv/a range recommended by the ICRP — generally for societal reasons — and this increased the remediation costs significantly. Prior to remediation, it was important to perform not only a thorough site survey but also a review of all available historical data. Gamma monitoring was a good, all round technique for site surveys, but buried contamination (sometimes several metres deep) could cause difficulties. Radon measurements had been found to be useful for detecting buried radium contamination. It was important to specify cleanup levels in advance and to agree on the degree of averaging to be used. During remediation, a dose constraint of 1 mSv/a was often appropriate for remediation workers. The waste management strategy should be agreed on in advance, using an approach based on waste segregation according to the concentrations of radionuclides and other hazardous constituents.

8. MEASUREMENT AND DOSE ASSESSMENT

The standardization of measurement methods and protocols was highlighted as being important for evaluating practices involving NORM. Developments in measurement techniques and equipment were reported, as well as new approaches to the statistical evaluation of measurement data. Interest was growing in the further development of mobile measurement instruments and techniques for in situ applications such as site remediation surveys (e.g. gamma dose rate, soil analysis, radon and dust) and screening of building materials. Developments in analytical techniques were reported for various NORM residues including contaminated soil, water, scale and residues from the smelting and separation of rare earths. There was also ongoing interest in techniques for measuring the decay progeny of radon and thoron (as well as associated dose assessment techniques) in enclosed environments such as homes, basements and caves. The

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alarm trigger levels of portal monitors tended to be set at some multiple of the standard deviation of background variations. The performance of such portal monitors with respect to shipments of NORM through ports was not without its problems and continued to be a topic of attention. Several presentations from China focused on surveys of the natural environment (and possible perturbations caused by human activities), using techniques such as geomapping, remote sensing and airborne gamma surveys.

9. STAKEHOLDER INVOLVEMENT AND RAISING OF AWARENESS

Stakeholder involvement was highlighted as a key component of a proposed new integrated approach to mineral resource management described in the symposium. Reference was made to the need for a state of equilibrium to be established between the interests of stockholders (i.e. operators) and concerned parties through a 'social licence'. Features of the proposed approach in relation to mining and mineral processing activities included the 'comprehensive extraction' concepts of disturbing the ground only once by ensuring that everything of value was extracted together and of managing residues in a manner that eliminated waste. During the course of the symposium, several references were made to developments in industrial activities involving NORM that were aligned with this approach. The regulatory process for uranium mining in ecologically and culturally sensitive areas of the Northern Territory of Australia provided a very good example of how stakeholder interests can be formally addressed in such situations. Some general trends in the mining and mineral processing industry can be regarded as being indicative of the comprehensive extraction approach. These include the trend towards co-production of minerals from a given resource, for instance the co-production of rare earths and phosphate in new uranium mining ventures, and the trend towards increased recycling of NORM residues and their use as by-products.

The need for concerned parties to be made aware of the risks associated with NORM residue management was highlighted as an area that needed ongoing attention. Awareness of radiation and its health effects needed to be addressed not only among interested and affected members of the public but also among workers in NORM industries.

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10. MAIN FINDINGS OF THE SYMPOSIUM

10.1. Regulatory aspects

- (a) During the period since the NORM VI symposium in 2010, much progress had been made in addressing exposure to natural sources in national laws and regulations, but a consistent regulatory approach had still not been fully achieved.
- (b) A revised BSS, published by the IAEA in 2011, was regarded as being too complicated and open to misinterpretation in their application to natural sources. This confirmed fears expressed previously at the NORM VI symposium when the revised BSS was still being finalized. More effort was therefore needed to improve the level of understanding among national authorities.
- (c) Inconsistencies in applying the requirements of the revised BSS to natural sources were particularly prevalent with respect to the application of the graded approach, including the application of the concept of exemption. There were still some States for which the regulatory framework did not provide a mechanism for exemption. However, there was now widespread acceptance in principle that regulatory control was unwarranted for materials with radionuclide activity concentrations below 1 Bq/g and for practices giving rise to doses less than 1 mSv/a.
- (d) Dose assessments based on measurements made during the transport of NORM had confirmed that the criterion for defining the scope of application of the IAEA Transport Regulations to NORM (ten times the activity concentration for exempt material) was appropriate.

10.2. Industrial activities involving NORM

- (a) Information presented at the symposium provided further confirmation that the list of 12 types of industrial activity that needed to be considered for possible control of exposure to natural sources was indeed valid and complete.
- (b) It was further confirmed that an industry specific approach to the regulation of NORM was the correct approach. The industry specific information provided in the suite of five IAEA publications in the Safety Reports Series was proving to be useful to national authorities.
- (c) A large amount of new information on exposures in NORM industries had become available. This new information verified previous findings that the majority of workers received doses less than 1 mSv/a and that doses received by members of the public were very low or insignificant.

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However, the new information highlighted the potential for high radon levels in underground mines if they were not adequately ventilated.

10.3. NORM residues

- (a) There is still a lack of harmonization of national approaches to the management of NORM residues. However, acceptance of the need to minimize NORM waste by recycling NORM residues or using them as by-products (with dilution if necessary) continues to grow. Some national authorities are now actively promoting this approach instead of discouraging or prohibiting it as in the past.
- (b) Although the use of NORM residues as components of building materials was a sensitive issue, national authorities are, to an increasing extent, actively pursuing this approach because of a growing recognition of the need to conserve raw materials and to reduce the amounts of NORM residues requiring disposal as waste. Exposures arising from the use of NORM residues as constituents of building materials are subject to the requirements for existing exposure situations and a reference level of 1 mSv/a for the control of such exposures appeared to be universally accepted. However, there are differences in practical approaches to the translation of this level of dose into an equivalent activity concentration of the material, with reluctance on the part of some authorities to carry out the necessary situation specific dose assessment.
- (c) Progress continues to be made in identifying suitable options for the disposal of NORM waste. Landfill disposal in one form or another is being favoured for a wide range of NORM wastes.

10.4. Remediation of legacy sites

- (a) Remediation projects continue to be carried out at sites contaminated by radionuclides of natural origin. The presentations made at this symposium demonstrated clearly that, although the scale of these projects varies widely, there are many common features that provide great potential for the sharing of experience.
- (b) There is a tendency for national authorities to set a very low reference level for site remediation, the value sometimes being at or below the lower bound of the 1–20 mSv/a range recommended by the ICRP. This is largely driven by societal factors, but the resulting costs can be very high. While such reference levels may have been considered to be justified and affordable in the countries concerned, this may not be the case in other countries, which may therefore need to consider a more pragmatic approach.

10.5. Involvement of interested parties

- (a) The importance of the involvement of all concerned parties is being recognized to an increasing extent in industrial activities involving NORM, as evidenced by various examples presented at the symposium.
- (b) A related issue is the raising of awareness of radiation exposure from NORM and the associated health effects. Ideally, this should involve not only members of the public — workers are also concerned and need to have an appropriate level of understanding of the risks associated with their work.

OPENING SESSION

Chairpersons

A. POFFIJN

Belgium

HUA LIU

China

OPENING ADDRESS

Hua Liu

Ministry of Environmental Protection,
Beijing, China

Ladies and gentlemen, dear participants, good morning!

I am very honoured to be invited to the Seventh International Symposium on Naturally Occurring Radioactive Material and to chair this opening session. Today, we have 120 experts and representatives from nearly 30 countries and international organizations attending the event. On behalf of the China Ministry of Environmental Protection (MEP), National Nuclear Safety Administration, I would like to extend my congratulations to the successful opening of the symposium and my warm welcome to all participants. I would also like to thank the International Atomic Energy Agency and the international community for the long term trust and support for China. My special appreciation goes to the hosts and organizers of the symposium.

NORM related issues on radiation protection and the radiation environment have attracted global attention. Under the leadership of the IAEA and the International Commission on Radiological Protection, many governments are undertaking research or implementing regulatory measures for NORM. This international symposium on NORM serves as a platform that promotes technical, academic, policy, regulation and standards exchange and cooperation in this field, which has proven to be essential. This NORM VII symposium has already identified several topics, including the management of exposure to natural sources, NORM in industries and regulatory aspects of NORM in meeting the new BSS requirements; environmental aspects of NORM; site specific measurement; dose assessment; application of the ALARA principle; development of NORM measurement methods and strategies; management of NORM residues and wastes; and NORM related transport issues. I believe these topics are conducive to providing insight to NORM and will greatly boost the regulation and management of NORM.

The Chinese Government has been attaching great attention to NORM related issues. China follows the standards and regimes of the IAEA and the European Union, and combines these with China's legal framework and actual conditions to regulate NORM. Between 2006 and 2009, the MEP completed a nationwide survey of pollutant sources, including NORM. To study NORM pollution, the MEP organized Chinese experts to carry out detailed monitoring and evaluation in such key areas as Baotou and Bai Yun Ebo of the Inner

Mongolia Autonomous Region, and recently in the Lincang area of Yunnan Province. Based on this achievement, the MEP released a regulatory catalogue of NORM for a partial list of industries this year to further enhance the regulation of NORM. With the monitoring and evaluation in place, the MEP will continue to release the NORM catalogue, push forward with regulations on NORM and deal with NORM legacy situations. The MEP and related radiation safety management agencies are willing to share their experience with international peer counterparts and carry out exchange and cooperation.

Ladies and gentlemen, the Fukushima accident had a great impact on global nuclear energy development. The accident further raised public awareness of radioactive material and its release to the environment. It also gave rise to new requirements for nuclear and radiation regulation and radioactive pollution control. The gaining of experience and feedback from the accident has progressed in phases, but the lesson learning process still has a long way to go. The international community will work together and take measures to avoid a recurrence of such an accident, so that people can enjoy the benefits of nuclear energy while avoiding the harm done by radioactivity. After the Fukushima accident, China conducted a comprehensive nuclear safety inspection, released improvement requirements and published a nuclear safety plan which provides key guidance for strengthening nuclear and radiation safety and radioactive pollution control. China is willing to share its experience and achievement in this regard.

Finally, I wish this symposium a great success. I hope everyone can make use of this great symposium to further discuss NORM regulation, promote experience sharing and contribute to protecting people and the environment. I hope you all enjoy your stay in Beijing.

Thank you!

OPENING ADDRESS

Senlin Liu

China Institute of Atomic Energy
Beijing, China

Honourable leaders, experts and participants,

It is a great honour for me to participate in this official opening ceremony. I welcome you here to attend the Seventh International Symposium on Naturally Occurring Radioactive Material and I wish you a happy and comfortable stay in Beijing.

This is a multidisciplinary subject, related to nuclear physics, environmental studies, chemistry, hydrogeology and health physics. The environmental impacts of mining have received considerable emphasis. An additional goal was to consolidate and strengthen knowledge and competence in radiation protection, which was seen as being very important in the evaluation and management of enhanced exposures to radiation from natural sources.

Historically, most regulatory attention has been focused on the mining and processing of uranium ore because such activities are a direct consequence of the radioactivity in the ore and form part of the nuclear fuel cycle. Over the past decades, however, more and more countries have introduced measures to regulate exposures arising from a wider range of natural sources, in particular minerals and raw materials other than those associated with the extraction of uranium. Long lived radioactive elements such as uranium, thorium and potassium and their decay products such as radium and radon are examples of radionuclides associated with NORM. These elements have always been present in the Earth's crust and atmosphere.

The term NORM exists also to distinguish 'natural radioactive material' from anthropogenic sources of radioactive material, such as those produced by nuclear power and used in nuclear medicine, where incidentally the radioactive properties of a material are maybe what make it useful. Exposure to naturally occurring radiation is responsible for the majority of an average person's yearly radiation dose and is therefore not usually considered to be of any special health or safety significance. However, certain industries handle significant quantities of NORM, which usually ends up in their waste streams. Over time, as potential NORM hazards have been identified, these industries have increasingly become subject to monitoring and regulation. However, there is as yet little consistency in NORM regulations among industries and countries. This means that material which is considered radioactive waste in one context may not be considered so

in another. Also, material which may constitute low level waste in the nuclear industry might go entirely unregulated in another industry.

This NORM VII symposium, which is being attended by 120 participants from 28 countries, has provided an important opportunity to review the many developments that have taken place over the past few years. It also coincides with various current initiatives to review and revise international recommendations and standards on radiation protection and safety. The proceedings will contain all written contributions, including invited papers, contributed papers and poster presentations covering seven different areas of NORM. On this occasion, I would like to thank all those who have helped us to achieve the organization of this major scientific event, especially the Steering Committee members who were closely involved in the organization process. They provided real support to us through their guidance and suggestions, so let me thank them once again.

Finally, I welcome you for your attendance and gratefully acknowledge the cooperation and support. And I wish for you to have a fantastic stay in Beijing. I wish the conference every success.

Thank you!

OPENING ADDRESS

Junxin Zhang

China Atomic Energy Authority

Beijing, China

Distinguished guests,

I am very pleased to open this Seventh International Symposium on Naturally Occurring Radioactive Material today. On behalf of the China Atomic Energy Authority (CAEA), I would like to take this opportunity to extend my warm welcome to all the officials and experts from government and industry research institutes from different countries.

The development of nuclear energy and technology is one of the most important achievements since the twentieth century. The application of nuclear technology in industry, agriculture, food and cancer treatment has brought great benefits to the public. On the other hand, radiation has been perceived as a big threat to the health of the public and the environment. The radiological impact of NORM on mining workers, radiation contamination by building materials, and the coordination between the development and safety of nuclear energy and technology has become a common problem faced by all countries.

As the competent authority for nuclear industry development in China, the CAEA is responsible for research and the drawing up of policies, regulations, planning, and programmes and industry standards related to the peaceful uses of nuclear energy and exchanges and cooperation with other foreign governments and international organizations in the nuclear field, and takes a leading role in national nuclear emergency response and management. After acceding to the IAEA in 1984, China has continued to make a contribution to the IAEA's activities in promoting the universal uses of nuclear energy and technology in industry, agriculture, health, environmental protection, and public security, and so on, including this symposium. We believe that international cooperation is one of the best ways of promoting the sustainable development of nuclear energy and technologies.

NORM is not a new topic. For a long time, the impact of NORM on the public environment and on working conditions has not been adequately addressed by regulatory bodies. For example, as I know, the concentration of radon in buildings is one big concern of the public. But the detection and mitigation technologies are not broadly used. Radiation measurement, dose assessment for mining or mineral processing sites, and the management of NORM waste are still falling behind the development of nuclear application technology.

JUNXIN ZHANG

This symposium is a very good opportunity for us to expose ideas and scientific results concerning new methods and technologies for NORM management. This will certainly be an opportunity to reflect on a framework of partnership and cooperation in order to work together for developing new projects and to ensure their implementation.

Thank you very much!

OPENING ADDRESS

P.P. Haridasan

International Atomic Energy Agency,
Vienna

Respected dignitaries on the dais, distinguished delegates, ladies and gentlemen,

It is indeed a great pleasure and honour for me to represent the International Atomic Energy Agency at this opening ceremony of the Seventh International Symposium on Naturally Occurring Radioactive Material here in Beijing. As you all might know, the NORM symposia series originated in Europe in 1997 as a result of a European Commission directive to control exposures to NORM, and the first symposium was held in Amsterdam, the Netherlands. Subsequently, the symposium moved to Germany, Belgium, Poland, Spain and then progressively out of Europe to Marrakesh, Morocco, in 2010 and now Beijing. The IAEA has been associated with the symposia from NORM III onwards and, from NORM IV onwards, has played a leading technical role and published the proceedings. Now we are adopting a global approach towards understanding and utilizing the best practices and regulations that are used in large countries such as China, India, Australia, Brazil and so on.

With that in mind, NORM VII is aiming to focus on the latest developments in the field of radiation protection associated with natural sources; in particular industries processing minerals and raw materials containing natural radionuclides. This is all the more important when considering the life cycle approach of industries and in particular the issues related to residues and waste management. The main objective of this conference is to create a forum for discussion on the recent advances made in the various topics, to understand the best practices that are useful for sustainable development and to improve on the radiation protection of workers and the public in relation to exposures to NORM.

On behalf of the International Atomic Energy Agency, I place on record our deep appreciation and thanks to the government of China and in particular to the China Atomic Energy Authority, the National Nuclear Safety Administration, the China Institute of Atomic Energy, the Ministry of Environmental Protection and all co-organizing institutions — including the three universities: Tsinghua University, the University of South China and the China University of Geosciences — for hosting and organizing this NORM VII symposium.

Our special thanks go to Dr Hua Liu; Professor Pan; Gang Wang, president of the China Institute of Atomic Energy; Professor Senlin Liu, China Institute of

Atomic Energy and the chancellors of the three universities. I would also like to thank one and all including any media representatives in attendance. My special thanks to Ms Xiaoyun Li and Mr Ruirui Liu for the excellent hard work and dedication put in for making this event a success.

Beijing is a beautiful city associated with several wonders including the Great Wall of China, one of the Seven Wonders of the World. The Forbidden City, Tiananmen Square, the Olympic Village, etc. are interesting and memorable places — make a point of visiting these locations of course after each day of our proceedings. China is a great country with a rich culture and technological excellence. Special thanks again to Dr Senlin Liu for such wonderful organization. Have a very successful symposium and a memorable stay in Beijing.

Thank you.

KEYNOTE ADDRESS

MANAGING EXPOSURE TO NATURAL SOURCES: INTERNATIONAL STANDARDS AND NEW CHALLENGES

P.P. Haridasan

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Vienna

Abstract

Radiation protection in exposure to natural sources has been evolving for decades. In the last two decades, developments concerning exposure to NORM have resulted in progress towards achieving broad international consensus on managing exposure to NORM. However, the standards and regulatory approaches being adopted at the national level still need to be harmonized, especially in developing countries with limited regulatory resources. A large effort is underway at the national and international level to assess exposure to NORM and to develop strategies to address existing situations that give rise to exposures. The United Nations Scientific Committee on the Effects of Atomic Radiation, in its 2008 report, encourages further development of inventories and methodologies for dose assessment in order to have a more comprehensive view over the topic. The revised International Basic Safety Standards (BSS) published by the IAEA in 2011 provides requirements reflecting the concepts of planned exposure situations, emergency exposure situations and existing exposure situations. Exposure to natural sources is generally subject to the requirements for existing exposure situations, with some exceptions to be considered as planned exposure situations. The BSS provides numerical criteria for exemption and clearance for regulatory purposes as well as reference levels for control of exposure to radon in workplaces. From a global perspective, the new radiation protection challenges for natural sources include the following: the harmonization of standards and regulatory approaches; the diverse nature of the industries and the need for an industry specific approach in determining radiation protection measures; the identification of situations that could be classified as either existing exposure situations or planned exposure situations; and the extent to which exposures should be optimized using, as appropriate, reference levels or dose constraints. The use of a graded approach in national regulations; the realistic estimation of individual doses and the need for an evidence based approach for regulatory decision making; radon in workplaces; transport issues; the recycling of NORM residues and their use as by-products; the management of NORM residues and wastes are related issues. A summary is provided of the international standards and guidance on the control of exposure to natural sources and of the work of the IAEA on industry specific safety reports.

1. INTRODUCTION

Natural radiation sources are ubiquitous in the living environment. All minerals and raw materials contain radionuclides of natural origin commonly referred to as primordial radionuclides. The ^{238}U and ^{232}Th decay series are the main radionuclides of interest for radiation protection purposes. Radionuclides in the ^{235}U decay series are less important for exposures, except for ^{227}Ac , which can contribute significantly to inhalation exposure. Since natural potassium contains 0.0117% ^{40}K , this radionuclide is also found in minerals and raw materials. The levels of other primordial radionuclides in minerals and raw materials (i.e. ^{87}Rb , ^{138}La , ^{147}Sm and ^{176}Lu) are not normally of concern for radiation protection. In normal rocks and soil, the activity concentrations of radionuclides in the ^{238}U and ^{232}Th decay series and of ^{40}K are variable but generally low. The average concentrations are 0.033 Bq/g for ^{238}U , 0.045 Bq/g for ^{232}Th and 0.412 Bq/g for ^{40}K [1]. However, certain minerals, including some that are commercially exploited, contain uranium and/or thorium series radionuclides at significantly elevated activity concentrations. Furthermore, during the extraction of minerals from the earth's crust and subsequent physical and/or chemical processing, the radionuclide concentrations in materials arising from the process may be significantly higher than those in the original mineral or raw material. During chemical processes, selective mobilization of radionuclides can disrupt the original decay chain equilibrium that existed in the ore. Such items of natural raw materials, ores, minerals, process residues and wastes containing elevated concentrations of natural radionuclides fall within the definition of NORM. Any mining operation or other industrial activity involving a mineral or raw material has the potential to increase the effective dose received by individuals from natural sources, as a result of exposure to radionuclides of natural origin contained in or released from such material. Where this increase in dose is significant, radiation protection measures may be needed to protect workers or members of the public. The presence of radionuclides at elevated activity concentrations is an important factor in determining which radiation protection requirements are applicable in terms of the IAEA International Basic Safety Standards¹ (BSS). This paper outlines the radiation protection challenges in applying the requirements of the BSS to exposure to NORM, including some of the emerging issues.

¹ INTERNATIONAL ATOMIC ENERGY AGENCY, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, IAEA Safety Standards Series No. GSR Part 3 (Interim), IAEA, Vienna (2011). [Editor's note] This has been superseded by Ref. [2].

2. EVOLUTION OF RADIATION PROTECTION STANDARDS AND REGULATORY CRITERIA

Radiation protection philosophy has been evolving since the discovery of radioactivity and the prominent recommendations in the recent past are the 1977, 1990 and 2007 recommendations of the International Commission on Radiological Protection (ICRP) [3–5]. The system of protection has changed from being a process led approach (practices and interventions) to a situation led approach (planned exposure situations, emergency exposure situations and existing exposure situations) with a greater emphasis on source related control. In its latest recommendations [5], the ICRP shifted from a mathematical approach to optimization to a more qualitative approach, always questioning whether the best possible has been done in the prevailing circumstances, and recommended a framework for **constraints** and **reference levels** to facilitate decision making by the national authorities. The use of reference levels is more relevant in the context of exposure to natural sources and the ICRP has recently revised downwards the reference level for radon in homes and at work. These recommendations and other developments led to the revision of the BSS, and in 2011 a revised version was published by the IAEA.

The BSS greatly influences the radiation protection regime, including natural sources. The BSS is consistent with the radiation protection recommendations of the ICRP, particularly in that it categorizes exposure situations into three types: planned exposure situations, emergency exposure situations and existing exposure situations. The stringency of protection in existing exposure situations (referred to in the previous version of the BSS as chronic exposure situations) has been significantly increased by strengthening the requirements for optimization of protection and widening their scope of application. In the case of radon, the stringency of control is further increased as a result of the Statement on Radon by the ICRP (see pp. 61–64 of Ref. [6]), in which it considers the health risk due to inhalation of radon to be significantly higher than previously assumed. Another important aspect on NORM in the revised BSS has been the greater use of quantitative criteria for deciding on the mechanism of control to be applied to exposures to natural sources and — in the case of exposures to be controlled as practices — for deciding on exemption and clearance.

Non-excluded exposures to natural sources are, in terms of the safety standards, normally subject to the requirements for **existing exposure situations**. In an existing exposure situation, the exposure is not regulated as a practice but, where necessary, may be controlled instead by remedial or protective actions, provided that such actions are undertaken only when they are justified and that their duration, nature and extent are determined by an optimization process to achieve the maximum net benefit. In some industrial activities involving minerals

and raw materials with elevated activity concentrations, however, it may be more appropriate to control exposures by regulating the industrial activity as a practice. In such situations, the exposures are subject to the requirements for **planned exposure situations**. A key question then arises — at what level of activity concentration does it become appropriate to regulate the industrial activity as a practice or planned exposure situations? It is important to appreciate that the selection of activity concentrations that are so low as to invoke widespread regulatory consideration, in circumstances where this is unlikely to achieve any worthwhile improvement in protection, would not be an optimum use of regulatory resources [7]. Consequently, the following activity concentration criteria are specified in the safety standards [2]:

- (a) If, in every process material, the activity concentrations of all radionuclides in the ^{238}U and ^{232}Th decay series are 1 Bq/g or less and the activity concentration of ^{40}K is 10 Bq/g or less, the material is not regarded as radioactive material (NORM), the industrial activity is not regarded as a practice and the requirements for **existing exposure situations** apply.
- (b) If, in any process material, the activity concentration of any radionuclide in the ^{238}U or ^{232}Th decay series exceeds 1 Bq/g, or if the activity concentration of ^{40}K exceeds 10 Bq/g, that material is regarded as radioactive material (NORM), the industrial activity is regarded as a practice and the requirements for **planned exposure situations** apply.

A summary of exposures to natural sources that are subject to the requirements for existing exposure situations, together with the exceptions to this normal approach which are considered instead as planned exposure situations, is provided in Table 1.

2.1. Radon in workplaces

Exposure to radon is normally subject to the requirements for existing exposure situations [2] and, in terms of the requirements for existing exposure situations, dose limits do not apply but the relevant national authority must establish a radon reference level, above which it is inappropriate to plan to allow exposures to occur and below which optimization should be implemented. The reference level for workplaces is not to exceed an activity concentration of 1000 Bq/m^3 . For homes, the maximum reference level is specified as 300 Bq/m^3 . Remedial and/or protective actions may be implemented to reduce exposure to radon provided that such actions are justified and optimized, with special attention being given to situations where the reference level is exceeded.

KEYNOTE ADDRESS

TABLE 1. EXPOSURES TO NATURAL SOURCES THAT ARE SUBJECT TO THE REQUIREMENTS FOR EXISTING EXPOSURE SITUATIONS

Exposures subject to the requirements for existing exposure situations	Exceptions (subject instead to the requirements for planned exposure situations)
Exposure to radionuclides from residual radioactive material from unregulated or inadequately regulated past activities	No exceptions
Exposure to radionuclides in everyday commodities (e.g. food, feed, drinking water, fertilizer, soil amendments, construction materials)	No exceptions
Exposure to radionuclides in material other than everyday commodities	<p>An exception applies if the radionuclide concentration exceeds 1 Bq/g (U, Th series) or 10 Bq/g (K-40)</p> <p>The exception also applies to public exposure to discharges and waste from the facilities concerned, irrespective of their activity concentrations</p>
Public exposure to Rn-222, Rn-220 and progeny indoors	No exceptions
Occupational exposure to Rn-222, Rn-220 and progeny	<p>An exception applies if:</p> <ul style="list-style-type: none"> (i) Exposure to other U, Th series radionuclides is controlled (as a planned exposure situation) or (ii) Rn-222 concentrations remain above the reference level after remedial action

There are two types of situation, however, where exposure to radon in the workplace becomes subject to the requirements for planned exposure situations (including the requirement for compliance with the dose limit for workers):

- (1) The first type of situation is when exposure to other radionuclides in the uranium and thorium decay series is already controlled as a planned exposure situation. An example of such a situation would be the underground mining of radioactive ore.

- (2) The second type of situation is where exposure to other radionuclides in the uranium and thorium decay series does not require to be controlled but where, after remedial action, the radon concentration remains above the reference level. An example of such a situation would be the underground mining of non-radioactive ore where increased ventilation is not possible and significant buildup of radon occurs in the workplaces.

2.2. Thoron in workplaces

Thoron (^{220}Rn) is not normally of concern in NORM industries, except where material with a high thorium content is processed, for example the processing of monazite to extract rare earths and thorium. The short lived progeny of thoron are likely to be severely out of equilibrium with the parent. Owing to the short half-life of thoron (55.6 s), the spatial distribution of thoron is much different from that of its progeny especially in enclosed workplaces. The assessment of an equilibrium factor is difficult and, for dose assessment purposes, an approach based on the measurement of thoron progeny concentration is easier and more appropriate than an approach based on measurement of the thoron concentration. Of the various thoron progeny nuclides, only ^{212}Pb and ^{212}Bi make significant contributions to the potential alpha energy. Since ^{212}Pb contributes almost 90% of the total potential alpha energy concentration (PAEC), its activity concentration in air can be used as a surrogate for PAEC, in which case a ^{212}Pb concentration of 1 Bq/m³ corresponds to a PAEC of 0.0691 $\mu\text{J}/\text{m}^3$.

2.3. The graded approach to regulation

If the relevant criterion for regulation is exceeded a graded approach to regulation has to be adopted, in accordance with para. 3.6 of the BSS. This means that the application of the requirements for planned exposure situations must be commensurate with the characteristics of the practice or the source within a practice, and with the magnitude and likelihood of the exposures. This is particularly important for industrial activities involving NORM owing to the economic importance of many industries, the larger volumes of residue generation with limited options for their management, and the potentially high cost of regulation in relation to the reductions in exposure that can be realistically achieved.

In order to determine the optimum regulatory approach, the regulatory body has to go beyond just establishing that the activity concentration criteria are exceeded. It must consider, in addition, particular types of operation, process and material in more detail, including an initial assessment of exposure or dose and consideration of the costs of regulation in relation to the benefits achievable.

Clearly a detailed understanding of the industrial activity concerned is essential for proper implementation of the graded approach. As part of the graded approach, the BSS makes provision for four levels of regulatory control. These levels are, in ascending order of stringency of control:

- (1) Exemption;
- (2) Notification;
- (3) Notification plus authorization in the form of registration;
- (4) Notification plus authorization in the form of licensing.

2.4. Radiation protection for planned exposure situations

For planned exposure situations or authorized practices, there are several radiation protection requirements specified in the standards. One of the fundamental requirements embodied in the BSS is that protection and safety should be optimized, that is, the magnitude of individual doses, the number of individuals exposed and the likelihood of exposure should be as low as reasonably achievable, economic and social factors being taken into account. In addition, the annual effective doses received by workers and members of the public should not exceed the applicable dose limits (20 mSv for workers, 1 mSv for members of the public and other equivalent dose limits to organs, e.g. the lens of the eye). Responsibilities include the performance of an appropriate safety assessment and the establishment and maintenance of a system of protection and safety to protect workers and members of the public against exposure. This includes an appropriate radiation protection programme (RPP) for occupational exposure consisting of organizational, procedural and technical arrangements for the designation of controlled areas and supervised areas, for local rules and for monitoring of the workplace, assessment and recording of occupational exposure, workers' health surveillance and provision of adequate information, instruction and training.

Ideally, the system should also ensure that members of the public are adequately protected against exposure, by means of the management of radioactive waste and discharges of radioactive material to the environment in accordance with the conditions of the authorization and proper environmental monitoring and surveillance.

As there exist various conventional hazards in industrial processes involving NORM, radiation protection must be recognized as being only one element in ensuring the overall health and safety of workers and the public and protection of the environment. In order to keep radiation protection in perspective with the need for protection against these other hazards, the RPP should ideally be established in close cooperation with those responsible for other areas of protection and

safety, including those responsible for industrial hygiene, industrial safety, fire safety and environmental protection.

3. RADIATION PROTECTION CHALLENGES

3.1. National regulatory approaches

National regulatory bodies either adopt the BSS in their regulation directly or incorporate relevant parts with modifications suited to national situations. Safety Reports Series No. 49, Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials [7], identifies 12 industry sectors that are likely to require some form of regulatory consideration and has now received widespread acceptance, thus providing national authorities with the means to focus their regulatory attention on those areas where it is most needed. There has been progress made towards the harmonization of standards and regulatory approaches for the control of exposure to NORM, however, still there are differences between countries or within individual countries. There is general acceptance of the 1 Bq/g criterion for uranium and thorium series radionuclides as a tool for determining which industrial process materials need to be considered for regulation. However, concerns exist over the need for an evidence based approach for regulatory decisions. Uncertainties in worker and public dose assessments, conservative modelling and prediction of exposure scenarios far from real situations (ultimately leading to inappropriate regulatory decisions), and the determination of the nature and extent of surveillance are some of the current regulatory and operational challenges.

3.2. Planned exposure situation or existing exposure situation?

It is recognized that the principles and concepts behind the ICRP recommendations on planned exposure situations and existing exposure situations have not been fully understood in terms of their practical application to exposures to NORM. The identification of situations that could be classified as either existing exposure situations or planned exposure situations and how such exposures should be optimized using, as appropriate, reference levels or dose constraints is one of the main challenges. Recognizing this issue and several others, the ICRP's Committee 4 formed a task group to develop a decision aiding framework for the practical implementation of the ICRP recommendations on radiation protection for NORM, as set out in ICRP Publication 103 [5]. The framework would cover the entire range of NORM activities, including shipment and waste management of bulk quantities, as well as the presence of NORM in

consumer products, especially construction materials, while taking into account the recent publications and documents of other international organizations such as the IAEA. The most appropriate regulatory considerations specific to a particular industry and type of NORM process have been provided in five industry specific IAEA publications in the Safety Reports Series [8–12].

3.3. Reference levels and dose constraints

Reference levels and dose constraints are important tools in the optimization of protection to restrict individual doses. Establishing an appropriate or single national reference level is an issue in several countries and a complex one when considering countries with federal and state level administrative systems. Reference levels and dose constraints have sometimes been used or considered as limits, defeating the purpose of optimization. Also, there exists some confusion between reference levels and the previously used ‘action level’ (at or below which remedial action and thus the need for optimization is not normally necessary) in terms of practical application in workplaces. The choice of an appropriate national reference level is complex and the determination of a particular value has large economic implications for industry and the country as a whole.

3.4. Exclusion, exemption and clearance

Numerical criteria for exemption and clearance of NORM have been included in the revised BSS. Exemption is determined on the basis of dose, commensurate with natural background levels (about 1 mSv per year). Clearance criteria for NORM are 1 Bq/g for radionuclides in the uranium and thorium decay series and 10 Bq/g for ⁴⁰K. These criteria are now gaining increased acceptance among industry, regulatory bodies and the public at large. However, differences in interpretation of the concepts of exclusion, exemption and clearance were reported at the NORM VI symposium in 2010 [13]. For example, the 1 Bq/g criterion for subjecting material to regulatory consideration as a source within a practice was variously referred to as an exclusion level, an exemption level, a clearance level or even a limit. There was a tendency to apply the concept of exemption not only to planned exposure situations but to existing exposure situations as well. The use of the terms exclusion and exemption interchangeably without appropriate qualification was observed. For instance, reference to the 1 Bq/g criterion as an exclusion level could mean either that the material satisfying this criterion was excluded from regulation as a planned exposure situation (but not necessarily from control as an existing exposure situation), or that the material was excluded from the standards entirely. Similarly, NORM in transport that fell outside the scope of IAEA Safety Standards Series No. SRR-6, Regulations for the Safe

Transport of Radioactive Material (Transport Regulations) [14], was sometimes referred to as being excluded, but such material was not necessarily excluded from the requirements of the BSS. Differences in interpretation of the concept of clearance were also reported.

3.5. Industry specific approach

It is recognized that a uniform approach in determining radiation protection measures in all industries involving NORM is sometimes impractical. This is true in the regulatory context, since the nature and level of the radiological risk varies considerably from one industrial process to another. Similarly, it was noted on several occasions that actions taken to comply with regulation were situation specific and could not be defined on a generic basis as being applicable to all industrial processes. Good progress had been made in developing good practices tailored to most of the industry's own particular set of circumstances. This highlights the particular need for an industry specific approach when applying radiation protection measures in challenging operating environments.

3.6. Exposure of workers

As part of the RPP and the graded approach to the control of exposure of workers in planned exposure situations, there is a need to assess the external and internal exposures to workers. Several methodologies are in place. However, the acquisition of exposure data for workers and the assessment of dose still suffer from a non-standardized approach and incomplete information in several countries, making a reliable assessment of the need for, and extent of, regulatory control difficult. It is becoming increasingly clear that in most of the industrial workplaces the doses are of the order of less than 1 mSv with a few exceptions such as uranium mining and processing and rare earths extraction from thorium containing minerals. However, possible scenarios with higher exposure potential are observed if adequate control measures are not implemented.

The radon concentrations in most of the workplaces concerned except uranium and thorium ore processing were generally less than about 100 Bq/m³. Some of the uranium mines and underground workplaces are reported to have higher radon concentrations which pose an additional challenge for the protection of workers in the industry. More practical guidance for the protection of workers is called for in cases of existing exposure situations, where the radon concentration in the workplaces continues to be above the national reference level after taking all reasonable remedial actions.

Recently, the ICRP has stated that the risk associated with exposure to radon is now considered to be significantly greater than previously estimated [6]. This, in conjunction with the proposed new dosimetric approach to the derivation of the dose conversion coefficient for intake of radon, may pose a great challenge to those responsible for the control of exposures to radon in workplaces especially in underground uranium mines. Considering the expected doubling of the dose for the same concentration previously, protection against radon in uranium mines and other workplaces with significant radon concentrations may require increased attention and measures to control exposures.

3.7. Exposure of the public

The results of various estimates of doses received by members of the public from mining and minerals processing facilities, as well as from the use of residues from such facilities, are summarized in the proceedings of the NORM VI symposium [13]. As is inevitably the case when assessing doses to members of the public, the estimates are likely to be subject to considerable uncertainty and tend to be conservative, owing to the necessity for modelling of the relevant exposure scenarios. The estimated doses are in general significantly below 1 mSv per year. However, instances of concern with respect to groundwater contamination have been reported, but in most cases the toxicity of other contaminants such as heavy metals, acidity and/or alkalinity were prominent rather than radionuclide concentrations.

There is a lack of uniformity in the approach to the use of NORM as a component of building materials, although it is generally accepted that any situation giving rise to a dose of more than 1 mSv per year would need special consideration and in certain cases some form of restriction. National approaches to the use of NORM in building materials need to be more pragmatic and realistic. There is a strong need for an evidence based approach in assessing exposure of the public to NORM. Many national regulatory bodies in the European Union are either considering or using a methodology based on the 'activity concentration index' to evaluate building materials for radiation protection purposes.

3.8. Management of NORM residues and wastes

One of the important issues in industries involving NORM is the management of residues and wastes. NORM residues are generated in a variety of forms and usually in very large quantities, for example, mine tailings and waste rock piles. Their sheer volume and high visibility in the environment make them a subject of radiological interest, even though the radionuclide activity concentrations may be low. Some types of solid residues are in the form of pipe

scale, sediments or sludge, usually in moderate volumes but having rather higher activity concentrations — sometimes up to three orders of magnitude higher than in the process feedstock. Other solid NORM residues are generated from high temperature processes, either in large quantities such as furnace slag or in small quantities such as furnace dust particles captured from stack emissions. Residues containing NORM may also be generated as liquids in the form of spent process water or spent solvents. There is also potential for off-gases generated from high temperature processes containing NORM residues or releases through ventilation air containing elevated radon concentrations.

The need for minimizing radioactive waste is one of the basic principles embodied in the IAEA safety standards. Since NORM residues contain radionuclides of very long half-lives and in very large quantities, the principle of recycling them or using them as by-products is an important consideration in NORM residue management. An increased acceptance of NORM residues as a resource rather than as waste has been observed globally. The disposal of NORM residues as waste should be considered only as a last resort. The opportunities for recycling residues or using them as by-products depend on a variety of factors, including the type of residue, the rate at which it is generated, the location of the facility and, in the case of by-product use, local market conditions. Consequently, the approach to the management of NORM residues, especially the degree to which NORM residues are recycled or used as by-products, needs to be tailored to the particular industrial activity and its location. Nevertheless, there is an overall trend worldwide towards greater recycling of residues and their use as by-products, sometimes being driven by sustainability issues and economic and liability considerations. This is particularly relevant to bulk NORM residues, for which the full extent of the problems associated with their storage and eventual disposal is only now being recognized. These problems arise from the large volumes of material involved, the large land areas needed for storage and disposal, structural safety considerations, environmental protection issues such as groundwater contamination and the possibility of financial liabilities that are sufficiently large to threaten the viability of the industrial activity concerned.

With regard to the establishment of good practices for the management of NORM waste, a risk based and situation specific approach is essential. Some of the wastes in this category usually considered for disposal are obviously the tailings and other waste from the processing of uranium ore, but also the tailings, slag and chemical processing wastes associated with the production of thorium and rare earths, the radium rich scale from the oil and gas industry and sludge from water treatment facilities, among others.

3.9. Transport issues

The IAEA Transport Regulations were recently updated, and the new version was published in 2012 [14]. The criterion for application of the IAEA Transport Regulations to NORM (namely, ten times the activity concentration for exempt material) remains essentially the same. Flexibility for the application of the requirements on a case by case basis does not exist in the regulations. NORM related transport issues were raised mainly at ports around the world (portal monitoring) because of the triggering of alarms designed to detect radioactive sources in scrap metal or to combat the trafficking of illicit nuclear material. Methodologies to determine activity and activity concentration of each radionuclide of natural origin in such portal systems have been developed; however, there is still a lack of a uniform protocol for measurement and interpretation. Continued improvements will be needed in the design and operation of such monitoring systems and in the training of operators.

3.10. Uranium mining and exploration activities

Uranium mining and exploration activities are carried out on a large scale in several countries around the world. Many abandoned mines from a previous era are being re-examined for their potential to be reopened or to have their residues reprocessed. Furthermore, planning for the exploitation of previously known but undeveloped uranium deposits is proceeding in many countries new to uranium mining. All of these activities pose significant challenges for the radiation protection profession particularly in meeting the operating and regulatory requirements at every stage of the uranium production cycle, from exploration through mining and processing to remediation. In addition, the assurance of a supply of qualified experts to address the radiation protection and other conventional safety issues is a challenge to the industry.

3.11. Legacy situations

There exist a number of legacy situations worldwide from former industrial activities where industrial production stopped and sites were simply abandoned with little or no attention paid to remediation. Most of these sites are associated with the mining and processing of uranium and, in a few cases, with monazite/thorium processing. The situation in central Asia regarding former uranium production sites (nearly 48 sites in Kazakhstan, Kyrgyzstan, Tajikistan and Uzbekistan) was often highlighted as a major challenge in this regard, requiring coordinated international effort to assist the countries concerned in planning and carrying out the necessary remediation work. The sites were

generally uncontrolled and the NORM residues from the mining and processing were a source of environmental contamination. Much of the efforts have been undertaken by the IAEA in developing remediation strategies and providing support to Member States.

4. EMERGING ISSUES

4.1. Involvement of interested parties and communication

Social licensing and public communication are important challenges when considering worker and public protection in NORM industries. The importance of stakeholder involvement as a critical component of communication has been recognized in the past. Good communications with all concerned parties are essential for sustainable industrial development but challenging in the case of issues involving radioactive material. The licensing and decision making process should involve all concerned parties from the very beginning to enhance the understanding, transparency and visibility of regulatory actions. Public communication is also vital in the establishment of large scale industrial operations involving NORM. At least a few countries are currently facing such issues relevant to radiation protection in relation to the processing of minerals and raw materials containing radionuclides of natural origin.

4.2. Hydraulic fracturing in oil and gas industry

Hydraulic fracturing is a well stimulation technique used to maximize the extraction of oil and natural gas in unconventional reservoirs, such as shale, coal beds and tight sands. During hydraulic fracturing, specially engineered fluids containing chemical additives are pumped under high pressure into the well to create and hold open fractures in the formation. These fractures increase the exposed surface area of the rock in the formation and, in turn, stimulate the flow of natural gas or oil to the wellbore. There are increased concerns about the potential environmental and human health impacts with the increased use of hydraulic fracturing, at least in a few countries. Many concerns about hydraulic fracturing centre on potential risks to drinking water resources including risks from radionuclides of natural origin, although other issues have been raised. The release of radon to the atmosphere as a result of hydraulic fracturing is also of growing public concern.

4.3. Recycling/use of NORM residues

Many countries are exploring possibilities on the recycling and use of NORM residues, primarily for economic reasons. The recycling of residues, as well as their direct use as by-products, are becoming increasingly attractive. Some countries are now specifically providing for NORM residue recycling and use in their regulatory systems. However, there are concerns about public acceptability. With regard to the use of NORM residues in building materials, there is no uniform approach to the translation of the 1 mSv dose criterion into some directly measurable parameter such as activity concentration and there are concerns about radon exposure, especially in European countries.

4.4. Measurement methods

Many industries involving NORM have neither an adequate infrastructure for analysing and interpreting radionuclide concentrations in their process materials nor qualified experts for radiation protection. There is a need for the development and standardization of field methods to analyse radionuclides in industrial samples. This is particularly important for industries such as the oil and gas industry when considering the storage of NORM residues and their disposal as waste.

5. CONCLUSIONS

The technological and regulatory developments in the last two decades have resulted in considerable progress towards harmonization of standards and regulatory approaches for the control of exposure to NORM, but still there are differences in national approaches. The requirements embodied in the recently revised version of the BSS provide greater clarity on the control of exposure to natural sources. Further progress in the harmonization of national standards and regulations on NORM can be achieved through the incorporation of the relevant BSS requirements. Exposure to natural sources is normally subject to the requirements for **existing exposure situations**. However, if the activity concentrations are significantly elevated above background levels, regulatory control in accordance with the requirements for **planned exposure situations** may need to be considered. The 1 Bq/g criterion for uranium and thorium series radionuclides as a tool for determining which industrial process materials need to be considered for regulation has been widely accepted. There is growing recognition that an industry or process specific approach is needed for ensuring that exposures to NORM are controlled optimally and effectively. Industrial

activities involving NORM, and legacy situations from such activities, are very diverse and each has to be addressed by developing 'good practice' according to the particular set of circumstances. The IAEA strongly promotes the graded approach to regulation to control exposures to NORM.

Several new radiation protection challenges have been identified. These include: differences in standards and regulatory approaches between countries; the need for an industry specific approach; confusion in classifying existing exposure situations or planned exposure situations; setting of appropriate reference levels in existing exposure situations; the implications of conservative modelling for estimating doses, in particular public exposures; the need for an evidence based approach for regulatory decision making; and differences in interpretation of the concepts of exclusion, exemption and clearance. The non-standard approach to the assessment of worker doses and issues emerging with the new ICRP risk estimate for exposure to radon pose a great challenge in workplaces. The management of NORM residues and wastes, issues associated with the recycling and use of NORM residues and legacy situations are still major issues around the world. The five industry specific IAEA publications in the Safety Reports Series have been widely recognized by regulatory bodies, industry, workers and the public at large. Emerging issues include stakeholder involvement and communication, growing public concern about radon and drinking water risks from the increased use of the hydraulic fracturing technique for oil and gas production, radiation protection issues from the recycling and use of NORM residues, and the need for the development and standardization of field methods to analyse radionuclides in industrial samples.

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Invited Paper

NORM EXPOSURE IN CHINA

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Abstract

The paper gives a brief introduction to exposure to NORM and other natural sources in China. The number of workers monitored for occupational exposure to natural sources exceeds 10.5 million, of whom 6.5 million work in underground coal mines and 3 million work in other non-metal mines. The average annual dose received by monitored workers, mainly due to inhalation of radon, is 2.1 mSv overall. For workers in metal mines, the average dose is more than 5.5 mSv. The paper also reviews the exposure of workers in the production of thorium containing gas mantles and welding electrodes, the exposure of aircrew to cosmic radiation and exposure to radon in underground workplaces other than mines, in hot spring facilities and in homes.

1. INTRODUCTION

It is well known that humans have been exposed to natural sources of radiation since they first inhabited the Earth. However, increases in such exposures as a result of the rapid expansion of industrial activities have not yet received sufficient attention. Table 1 summarizes the exposures to natural sources in China compared with worldwide exposures. One important natural source of radiation is NORM, defined as radioactive material containing no significant amounts of radionuclides other than naturally occurring radionuclides (where the exact definition of ‘significant amounts’ would be a regulatory decision). Although human activities do not change the origin of natural radiation, they can change the distribution and exposure pathways, an example being exposure to indoor radon.

TABLE 1. EXPOSURE TO NATURAL SOURCES

Source	Annual effective dose (mSv)		
	China		Worldwide
	1990 ^a	2000 ^b	
External			
Cosmic radiation ^c			
Ionizing component	260	260	280
Neutrons	57	100	100
Terrestrial gamma radiation	540	540	480
Internal			
Radon and its progeny	916	1560	1150
Thoron and its progeny	185	185	100
K-40	170	170	170
Other radionuclides	170	315	120
Total (rounded)	2300	3100	2400

Source: Data for China are from Ref. [1], for worldwide from Ref. [2].

^a Estimated from surveys up to the year 1990.

^b Estimated from surveys up to the year 2000.

^c Exposures to cosmogenic radionuclides are not included, as they are relatively small.

China has some of the largest coal resources in the world and also has abundant resources of rare earths, with proven reserves accounting for 80% of the world's total. There are more than 400 proven rare earth deposits, distributed widely across 19 provinces and municipalities, including Inner Mongolia, Sichuan, Shandong, Jiangxi, Guangdong and Hunan. These rare earth resources are often found to be associated with deposits of other ores. Among the proven deposits are a considerable number of mines with high levels of natural radioactivity contained in minerals such as monazite, zircon, xenotime and petscheckite. The Baiyunebo Iron Mine, in Baotou City, Inner Mongolia, is the country's largest commercially exploited deposit, consisting mainly of iron, rare earths and thorium. The content of thorium in the ore is about 0.04%. Mining and milling operations are carried out at the site. There are altogether 12 900 workers involved in the mining of rare earth ores throughout the country.

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Methods for the classification of mineral resources differ between countries. In China, there are 168 types of mineral resource classified into four categories: energy minerals; metal minerals; non-metal minerals; and water and gas minerals. The underground water resources are both mineral resources and water resources. China has the largest number of workers engaged in mining and mineral processing — nearly 10 million workers overall, of which more than 6 million are involved in coal mining.

Since the early 2000s, exposure to natural sources, particularly indoor radon exposure, has given rise to some social concerns in China. Nearly 100 million people live in cave dwellings, in which the radon concentrations are relatively high. In the 1980s and 1990s, the Ministry of Public Health and the Environmental Protection Agency organized a nationwide survey on radon. By the early 2000s, an indoor radon survey in 26 cities had been concluded. The results indicated that indoor radon activity concentrations had increased by 70% compared with those in the 1990s.

Activities related to NORM are mainly concerned with the extraction and processing of ores which can lead to enhanced levels of radionuclides of natural origin in products, by-products and wastes. These activities were identified internationally as follows: mining and processing of uranium ore; metal mining and smelting; the phosphate industry; coal mines and coal fired power generation; oil and gas extraction; the rare earths and titanium oxide industries; and the zircon and zirconia ceramics industries.

In order to estimate the exposure of the Chinese population to natural sources, a national survey was carried out. Exposures to natural sources were categorized as follows:

- (a) Exposure of workers in underground coal mines;
- (b) Exposure of workers in non-ferrous metal mines;
- (c) Exposure of workers in rare earths mines;
- (d) Exposure of workers to other minerals;
- (e) Exposure of aircrew to cosmic radiation;
- (f) Exposure of the public to indoor radon.

The results of that survey are described in this paper.

2. EXPOSURE OF WORKERS IN UNDERGROUND COAL MINES

2.1. Mine categories and workforce size

At present, there is no consistent method that can be applied to the categorization of coal mines in the country. Various categorization schemes can be found in the literature. In this paper, the coal mines countrywide are, for purposes of exposure assessment, grouped into the following three broad categories based on annual production and ventilation conditions:

- (1) Key state owned large sized coal mines with a high annual production and good ventilation;
- (2) Local state owned medium sized coal mines with a high annual production and good ventilation, run at provincial, municipal and county level;
- (3) Small sized, single shaft coal mines with poor ventilation, run by townships or private owners. Although, individually, these mines have a small annual production, their combined production is high. According to data from the China coal administration, there were 72 900 of these local mines across the country in 1995 and 82 000 in 1997 [3]. Although efforts to reform and reorganize these mines were made by the national and provincial governments in 1999, there were still 25 000 mines remaining in 2002, representing about 90% of the total number of coal mines in China [4].

A preliminary estimate indicates that the total number of workers in underground coal mines is about 6 million, comprising about 1 million in large sized mines, 1 million in medium sized mines and 4 million in small sized mines [5]. Of the workers in small sized mines, about 50 000 are involved in underground mining of bone coal.¹

2.2. Radon concentrations in underground coal mines

The arithmetic mean radon activity concentrations in the three types of underground coal mines, estimated according to monitoring results and published data, are shown in Table 2. Average radon concentrations weighted according to annual output and number of workers are shown in Table 3. In general, the average concentrations weighted according to the number of workers can be considered as having good representivity. Consequently, typical radon concentrations can be

¹ Bone coal is impure coal that contains much clay or other fine grained detrital mineral matter.

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taken to be 50 Bq/m³ for large sized mines, 100 Bq/m³ for medium sized mines and 500 Bq/m³ for small sized mines. In addition, the typical radon concentration in bone coal mines can be taken as 1500 Bq/m³. On the basis of data from more than 30 underground uranium mines over the past four decades, a radon progeny equilibrium factor of 0.35 can be assumed [6], giving typical equilibrium equivalent concentrations (EECs) of 17.5, 35, 175 and 525 Bq/m³ for large sized, medium sized, small sized and bone coal mines, respectively.

TABLE 2. RADON CONCENTRATIONS IN UNDERGROUND COAL MINES

	Activity concentration (Bq/m ³)			
	Measured values		Estimated values	
	Range	Arithmetic mean	Range	Arithmetic mean
Large sized coal mines	18–65	49.0	18–202	78.5
Medium sized coal mines	22–1 963	173	22–1 963	195
Small sized coal mines	14–3 115	631	14–3 115	536
Bone coal mines	136–4 183	1 244	136–23 976	5 997

TABLE 3. WEIGHTED AVERAGE RADON CONCENTRATIONS IN UNDERGROUND COAL MINES

	Average activity concentration (Bq/m ³)	
	Weighted according to annual production	Weighted according to number of underground workers
Large sized coal mines	49.0	52.9
Medium sized coal mines	173	142
Small sized coal mines	592	526
Bone coal mines	1133	1148

2.3. Dose assessment

Measurements made in underground coal mines [6] indicated that gamma radiation levels were generally equivalent to those at the Earth’s surface, or even lower in several large sized mines. Higher dose rates were found in a few mines. In the case of one bone coal mine, the gamma radiation levels were 142–932 nGy/h, with an average of 490 nGy/h. Assuming a natural background dose rate of 100 nGy/h, the incremental dose received by a worker at this mine would be about 0.9 mSv/a [6], which cannot be ignored. In general, however, the dose contribution from gamma radiation did not need to be considered when estimating worker exposures since the dose was principally due to radon exposure.

The doses received by underground workers due to radon inhalation were assessed by assuming annual exposure periods of 2000 h for large sized and medium sized mines and 2400 h for small sized mines including bone coal mines [6]. The results for ²²²Rn are shown in Table 4.

For ²²⁰Rn, the EECs were derived from the ²²⁰Rn activity concentrations reported in Refs [7–9] and were in the range of 0.4–0.9 Bq/m³. The typical EEC value was taken to be 0.5 Bq/m³ which, using the dose conversion coefficient reported in Ref. [2], gave an annual effective dose of about 0.04 mSv. Because of the limited data available for ²²⁰Rn in underground coal mines, the contribution to doses from ²²⁰Rn was not considered further.

TABLE 4. AVERAGE DOSES FROM ²²²Rn INHALATION

	Representative activity concentration (Bq/m ³)		Annual exposure period (h)	Radon progeny exposure (mJ·h·m ⁻³)	Annual effective dose (mSv)
	Rn-222	EEC			
Large sized coal mines	50	17.5	2000	0.198	0.28
Medium sized coal mines	100	35	2000	0.389	0.55
Small sized coal mines	500	175	2400	2.372	3.3
Bone coal mines	1500	525	2400	7.080	10.9

Note: EEC — equilibrium equivalent concentration.

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Table 5 shows the individual doses received by underground workers in the various categories of mine, together with the corresponding collective doses [6, 10]. The collective doses normalized to the dose per 10 000 t production were calculated from 2003–2004 production data — 880 million t for large sized mines, 290 million t for medium sized mines and 630 million t from small sized mines (with 6.5 million t from bone coal mines [11]).

TABLE 5. INDIVIDUAL AND COLLECTIVE DOSES

	Underground workers	Individual annual effective dose (mSv)	Collective annual effective dose	
			Total (man Sv)	Normalized (man Sv per 10 000 t)
Large sized coal mines	1 000 000	0.28	280	0.003 2
Medium sized coal mines	1 000 000	0.55	550	0.019
Small sized coal mines	4 000 000	3.3	13 200	0.21
Bone coal mines	50 000	10.9	545	0.84
Overall	6 000 000 ^a	2.4	14 600	0.081

^a The number of workers in bone coal mines is part of the number for small sized coal mines is therefore not included separately in the total for underground workers.

The results in Table 5 show that the average dose received by underground workers in coal mines is 2.4 mSv/a. The highest average dose is for bone coal mines, at more than 10 mSv/a, followed by an average dose of 3.3 mSv/a for small sized coal mines. The average annual collective effective dose for all underground workers is 14 600 man Sv, 91% of which (13 200 man Sv) is for small sized coal mines. The normalized annual collective effective dose is 0.081 man Sv per 10 000 t, to which the main contribution is from bone coal mines and the next largest is from other small sized coal mines.

In general, attention needs to be drawn to two categories of mine worker with respect to occupational exposure. The first category relates to workers in small sized coal mines, with a large exposed workforce receiving a relatively high dose, and thus representing the majority of the total collective dose. The

second category relates to workers in bone coal mines, who receive the highest dose, although the size of the workforce is relatively small. If attention is not given to the control of radon exposure in underground mining, a few workers could incur doses in excess of 50 mSv/a.

3. EXPOSURE OF WORKERS IN NON-FERROUS METAL MINES

3.1. Mine categories and workforce size

According to the Chinese Mining Industry Yearbook, non-ferrous metal mines typically mean metal mines other than ferrous metals and noble metals. Generally, non-ferrous metal mines include mines for light metals with densities less than 4.5 g/cm^3 and mines for heavy metals with densities higher than 4.5 g/cm^3 , as well as rare earth metal mines [12–14]. Ferrous metal mines comprise iron mines, manganese mines, chromium mines and associated alloy mines. Iron, chromium and manganese as well as their alloys are known as ferrous metals because of their surface being often covered with Fe_3O_4 . Noble metal mines mainly include gold mines, silver mines and platinum mines [14].

In response to a rapid expansion in market demand, the output of non-ferrous metals has shown a rapid upward trend in China. According to the Chinese Mining Industry Yearbook, published separately in 2005 and 2006, the number of workers involved in non-ferrous metal mining was 370 000 in 2004 and 399 000 in 2005 [12–14]. Based on statistical data compiled in 2006 by the Ministry of Land Resources of China, the total number of workers was 427 900. Consequently, it seems to be a reasonable to assume 400 000 as the typical value of the size of workforce when estimating the collective dose to the workers involved.

3.2. Worker exposures

The radiological impacts of ^{222}Rn , ^{220}Rn and their decay products in underground non-ferrous metal mines are larger than those in open pit mines. Because of poor local ventilation or the lack of any ventilation system, radon released from the fractured rock or from mine fissure water is not readily removed from the workplaces and thus accumulates to high concentrations. This should merit attention in the radiation protection of underground workers in non-ferrous mines.

A systematic analysis of radon concentrations and EECs in non-ferrous metal mines undertaken in 2002 indicated a typical EEC value of 1000 Bq/m^3 [15]. In a recent report, it was indicated that the average ^{222}Rn and ^{220}Rn activity

concentrations in non-ferrous mines, derived from the monitoring results by using a cumulative measuring method, were 1470 and 269 Bq/m³, respectively [16]. The average gamma dose rate was 463 µGy/h. In view of the relatively sparse data on exposures in non-ferrous metal mines, a ²²²Rn EEC of 1000 Bq/m³, a ²²⁰Rn activity concentration of 270 Bq/m³ and a gamma dose rate of 460 µGy/h could be considered as representative values for the purpose of dose estimation.

3.3. Dose assessment

Measurements indicate that ²²²Rn, ²²⁰Rn and their decay progeny dominate the internal exposure of underground workers in non-ferrous metal mining. In Ref. [2], dose conversion factors of 9×10^{-6} and 4×10^{-5} mSv per Bq·h·m⁻³ were reported for the progeny of ²²²Rn and ²²⁰Rn, respectively. The measured equilibrium factor for ²²⁰Rn was reported to be 0.004 [9]. Using the method described in Ref. [2] to estimate exposure and assuming an annual working period of 2000 h, the annual effective dose received by workers would be 18 and 0.08 mSv, respectively.

The external radiation dose to underground workers in non-ferrous metal mines arises mainly from external gamma exposure [17]. By subtracting the outdoor gamma dose rate, averaged across the country, the annual average effective dose was estimated to be 0.46 mSv.

As a result, for underground workers in non-ferrous metal mines, the total average annual effective dose was preliminarily estimated to be 18.5 mSv. Assuming the number of workers to be 400 000, the annual collective effective dose for non-ferrous underground mines would be 7420 man Sv.

Measurements in several non-ferrous metal mines show that ²²²Rn activity concentrations can be as high as 19 600 Bq/m³ [16]. More attention needs to be given to radiation risks to which the miners in this sector would be exposed when carrying out underground non-ferrous metal mining. Meanwhile, with the rapid expansion of the national economy, the demand for non-ferrous metal continues to increase. Therefore, the reinforcement of regulatory requirements should be considered for radiation protection to be improved in underground mines and for labour protection measures to be taken in engineering practice in such a way as to reduce the exposure levels of non-ferrous underground mines.

4. EXPOSURE OF WORKERS IN THE EXPLOITATION OF OTHER MINERALS

4.1. Rare earths

4.1.1. Mine categories and workforce size

China is a country abundant in rare earth resources, with proven reserves accounting for 80% of the world's total. There are more than 400 proven deposits, widely distributed over 19 provinces [18], including Inner Mongolia, Sichuan, Shandong, Jiangxi, Guangdong and Hunan. Rare earth resources across the country are often associated with deposits of other minerals. Among these proven deposits, a considerable number of mines are found to have high levels of natural radioactivity in minerals such as monazite, zircon, xenotime and petscheckite.

As revealed by the 2006 circular of the Ministry of National Land Resources on development and utilization of mineral or ore resources, there are 12 900 workers involved in the rare earths industry.

4.1.2. Exposures

As is well known, Baiyunebo Mine in Baotou City, Inner Mongolia is the largest commercial deposit of iron, rare earths and thorium in China. The content of thorium in the crude ore is about 0.04% [19].

The rare earths production activity consists mainly of mining and milling operations. The development and utilization of rare earths could lead to enhanced radiation levels, including environmental gamma radiation dose rate in air, radioactive aerosol concentrations in air, radon concentrations and radon progeny potential alpha energy in air, and natural radionuclide concentrations in water.

The gamma exposure levels measured in several rare earth mines are shown in Table 6 along with the annual effective doses received by the workers concerned [20–22].

4.2. Phosphates

The phosphate industry comprises a wide range of industrial activities associated with the processing of phosphate ore, consisting of mining, extraction and processing among others. The mining of phosphate ore could result in environmental contamination and potential doses to the public caused by radionuclides of natural origin, especially ^{238}U and its decay progeny (see Ref. [23] and pp. 129–142 of Ref. [24]).

TABLE 6. EXPOSURES OF WORKERS IN RARE EARTH MINES

	Gamma dose rate ($\mu\text{Gy/h}$)		Annual effective dose (mSv)
	Range	Mean	
Sichuan	110–380	2.4 ± 1.18	3.0
Guangdong	—	—	3.9
Shandong	9.9–135	0.325 ± 0.219	—
Inner Mongolia	15.9–42.3	0.298 ± 0.012	1.42–6.13
Shandong Weishan	14.4–21.5	0.180 ± 0.05	—

The activity concentrations of radionuclides of natural origin in 77 samples collected in the provinces of Yunnan, Guizhou, Hunan, Hubei, Sichuan, Jiangsu, Shaanxi, Jiangxi and Anhui are shown in Table 7. Of the total number of samples, 35% had a ^{226}Ra activity concentration exceeding 0.4 Bq/g. A few samples of phosphate ore were associated with high activity concentrations [18]:

- (a) Kaiyang phosphate mine, Guizhou Province: 2.35 Bq/g ^{226}Ra ; 14 Bq/g gross alpha.
- (b) Shuangfeng phosphate mine, Hunan Province: 5.22 Bq/g ^{226}Ra ; 57 Bq/g gross alpha.

4.3. Manufacture of gas mantles containing thorium nitrate

Radiation exposure to thorium nitrate is mainly from its use for the production of incandescent gas mantles [25]. During the manufacture of gas mantles, exposure occurs mainly during the process of dipping the mantle into thorium nitrate solution [25, 26]. The radionuclide of concern is ^{232}Th , with a half-life of 1.4×10^{10} years. The exposure levels in gas mantle manufacturing plants are shown in Table 8.

TABLE 7. ACTIVITY CONCENTRATIONS IN PHOSPHATE ROCK

Province in which sampled	No. of samples	Activity concentration (rounded) (Bq/g)											
		U-238			Ra-226			Th-232			K-40		
		Mean	Range	Range	Mean	Range	Range	Mean	Range	Range	Mean	Range	Range
Yunnan	16	0.3	0.2-0.5	0.3	0.1-0.4	0.02	0-0.06	0.1	0-0.4	0.1	0-0.4	0.1	0-0.4
Guizhou	19	0.6	0.2-3	0.5	0.2-3	0.01	0.01-0.04	0.1	0.06-0.3	0.1	0.06-0.3	0.1	0.06-0.3
Hunan	19	0.4	0.15-2	0.4	0.05-0.7	0.02	0-0.03	0.02	0-0.4	0.02	0-0.4	0.02	0-0.4
Hubei	12	0.1	0.02-0.3	0.1	0.02-0.3	0.01	0-0.05	0.2	0.06-0.6	0.2	0.06-0.6	0.2	0.06-0.6
Sichuan	1	0.3		0.3		0.03		0.1		0.1		0.1	
Jiangsu	5	0.4	0.03-0.9	0.3	0.03-0.93	0.03	0-0.1	0.1	0-0.2	0.1	0-0.2	0.1	0-0.2
Shaanxi	2	0.1	0.08-0.1	0.1	0.07-0.1	0.004	0.001-0.006	0.1	0-0.2	0.1	0-0.2	0.1	0-0.2
Jiangxi	2	0.1	0.09-0.3	0.2	0.09-0.3	0.02	0.009-0.03	0.1	0.1-0.1	0.1	0.1-0.1	0.1	0.1-0.1
Anhui	1	0.02		0.01		0.02		0.5		0.5		0.5	
Overall	77		0.3	0.3	0.3	0.2	0.2	0.1		0.1		0.1	

^a Zero values indicate a result below the lower limit on detection.

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TABLE 8. OCCUPATIONAL EXPOSURE AT GAS MANTLE MANUFACTURING PLANTS

	Gamma dose rate	Activity concentration in air (Bq/m ³)	
		Th-232	Rn-220
Nantong plant A	0.011 7 mGy/h		
Nantong plant B	0.016 6 mGy/h		
Jiangsu plant A	0.92 mSv/min	0.157	39
Jiangsu plant B	1.82 mSv/min	0.043	10.3
Zhenjiang plant	0.004 12 mGy/h	0.070	3.2
Danyang and four other plants	0.009 8 mGy/h	0.031	147
Zhejiang plant	0.003 8 mGy/h		
Mean value	0.009 2 mGy/h	0.085	98.4

Since 1993, monitoring has been conducted at a gas mantle manufacturing facility in southern Zhejiang. Doses were estimated for all workplaces based on 300 working days per year, each with 8 working hours per day, and an average breathing rate of 1.0 m³/h. The results are shown in Table 9. The monitoring results indicated high radiation exposure levels occurring both in the plant area and in its surrounding environment. The plant buildings are now undergoing cleanup after being monitored by the local environmental protection agency. Workers engaged in gas mantle manufacturing, especially for thorium dipping, may receive high doses and measures should be taken to reduce the exposure levels.

4.4. Manufacture of thoriated tungsten welding electrodes

Thoriated tungsten welding electrodes are widely used around the world because of their easier arc initiation, higher current carrying capacity and long life. They are highly suitable for argon arc welding and plasma welding. Thorium nitrate and ammonium paratungstate are the main materials for production of thoriated tungsten electrodes. The ²³²Th content gives rise to occupational exposure during the production process and also during the use

TABLE 9. OCCUPATIONAL EXPOSURE AT A GAS MANTLE MANUFACTURING PLANT

	Annual effective dose (mSv)											
	Nov. 1993			May 1994			Oct. 1996			July 1997		
	Ext.	Int.	Sum	Ext.	Int.	Sum	Ext.	Int.	Sum	Ext.	Int.	Sum
Thorium dipping	11.5	18.6	0.69	15.6	352.8	368.4	5.6	184.8	190.4			
Drying		5.9	5.21	7.5	241	248.0	7.8	126.0	133.8			
Incrustation	1.6	4.1	7.84	9.0	13.4	22.4	37.5	2.5	40.0			
Bottom sealing	9.5	8.8	5.54	7.4	13.4	20.7	1.7	2.5	4.2			
Mouth sealing		3.9	11.9	3.5	3.02	6.5	13.3	2.5	15.8			
Ironing	3.3	4.5	7.84	4.1	13.4	17.5	5.3	2.5	7.8			
Check	3.2	16.8	7.84	11.0	13.4	24.3	5.5	2.5	8.0			
Packaging	11.4	18.0	6.05	7.0	23.5	30.5	16.7	2.5	19.2			

Note: Ext. — external exposure; Int. — internal exposure.

of these electrodes. Exposure levels could also be enhanced in the environment surrounding the production facilities, such as around the manufacturing shop, raw material storage, finished parts storage, and waste storage, giving rise to exposure of the public in the vicinity of the sites. Table 10 shows the radiation doses that may be received by a worker at a thoriated tungsten electrode factory in a year. The workplaces where thoriated tungsten electrodes are handled are worthy of attention because of potentially additional radiation exposure to the workers.

TABLE 10. OCCUPATIONAL EXPOSURE AT A THORIATED TUNGSTEN ELECTRODE PRODUCTION FACILITY

	Annual exposure period (h)	Annual effective dose (mSv)
Adulteration	1186	0.18
Reduction	1825	0.08
Profiling	1825	0.17
Pre-burning	1825	0.13
Tube melting	1156	0.28
Pneumatic (rotary) hammer	912	1.05
Mean value	1454	0.32

4.5. Construction materials

In recent years, radioactivity in construction materials has been attracting more and more attention. The radioactivity can be due to natural minerals in the construction materials and to added constituents in the form of NORM residues, an example being coal slag with elevated activity concentrations which has been extensively used for the production of bricks [27]. Rock, as a type of construction material, has been intensively studied, with focus on the measurement of activity concentrations in marble, granitic rock and slabstone among others. Marble is mainly taken from magnesium carbonate rocks and metamorphic carbonate rocks; granitic rock is from metamorphic granite and slabstone is from slate and sedimentary rocks. Marble is found in such provinces as Henan, Zhejiang, Yunnan and Sichuan, among others. Granitic rock, with a red, incarnadine, black

or gray colour, is mainly distributed in Fujian, Shandong, Guangxi, Guangdong and Sichuan and other provinces. Slabstone is mainly from the city or provinces of Beijing, Shaanxi and Guizhou.

The activity concentrations in these three types of construction materials are shown in Table 11 [28–33]. Analytical results indicated that activity concentrations are particularly elevated in granitic rock, with the highest values, and thus the highest exposure potential, occurring in red granitic rock [32, 33]. Some imported granitic rock has an especially high radioactivity content, as shown in Table 12.

TABLE 11. ACTIVITY CONCENTRATIONS IN THE THREE MAIN TYPES OF CONSTRUCTION STONE

	Activity concentration (Bq/g)					
	Ra-226		Th-232		K-40	
	Mean	Range	Mean	Range	Mean	Range
Marble	0.021	0.000 34–0.097	0.019	0.000 65–0.193	0.059	0.009–1.003
Granitic rock	0.089	0.000 6–0.374	0.095	0.000 5–0.255	1.102	0.01–3.357
Slabstone	0.105		0.004 2		0.024	

TABLE 12. ACTIVITY CONCENTRATIONS IN IMPORTED GRANITIC ROCK

	Activity concentration (Bq/g)		
	Ra-226	Th-232	K-40
Italy (thin)	0.404	0.130	1.390
Italy (thick)	0.140	0.390	1.876
Finland (red)	0.620	0.161	1.347
South Africa (red)	0.256	0.057	1.283
India (red)	0.158	0.305	1.247

4.6. Geothermal springs

An assessment indicated an average radon EEC of 149 Bq/m³ at the sites of hot springs [15]. For this reason, investigations were made of Jiangzha hot spring, an artesian spring located in Jiergai, Sichuan Province [16, 34]. The ²³⁸U and ²²⁶Ra concentrations in the soils through which the spring water flows are 1.6 and 20 Bq/g, respectively, higher by a factor of nearly 500 than those measured at a control location. In the vicinity of the site, the gamma dose rates were 3.94 μGy/h for the fields, 5.89 μGy/h for the road and 1.47μGy/h for buildings. High concentrations of ²²²Rn and its progeny were found at the Jiangzha hot spring. The spring water is now being used for indoor baths, probably resulting in the inhalation of radon and its progeny at high activity concentrations by members of the public.

Table 13 shows the radon concentrations measured at several hot spring sanatoria, indicating that the concentration at Jiangzha is an order of magnitude higher than at the other sites. Although the radon concentration in the hot spring water at Jiangzha is high, the area is sparsely populated with only 200–300 residents, and is not therefore representative of the situation on a wider scale. Therefore, for the purpose of dose assessment, it would be reasonable to assume a radon EEC of 150 Bq/m³. Using the method for estimating dose given in Ref. [2], the annual average effective dose from workers' exposure to radon in hot spring sanatoria was estimated to be 2.7 mSv.

TABLE 13. RADON ACTIVITY CONCENTRATIONS AT HOT SPRING FACILITIES

	Activity concentration		
	Rn-222 in water (Bq/L)	Rn-222 in air (Bq/m ³)	
		Rn-222	Rn-222 EEC
Weihai sanatorium	110		48
Qingdao sanatorium	4.6		63
Zhaoyuan sanatorium	3.52		266
YW Zhuang sanatorium	27.2		249
Liaoning sanatorium	3270		330

TABLE 13. RADON ACTIVITY CONCENTRATIONS AT HOT SPRING FACILITIES (cont.)

	Activity concentration			
	Rn-222 in water (Bq/L)	Rn-222 in air (Bq/m ³)		
		Rn-222	Rn-222 EEC	Rn-220
Xianyang hot spring	231	190	85.2	
Puxzhe hot spring	230	126	56.9	
Xiongmi hot spring	1.41	11.6		
Lingyuan hot spring	3.91	15.6		
Tanggangzi hot spring	14.1	37.1		
Xincheng hot spring (I)	105.5	134.5		
Benxi hot spring	87.7	739.8		
Tanghe hot spring	2720	1240		
Xincheng hot spring (II)	52.7	303		
Jiangzha (Sichuan)		7098		35
Mean		989	149	

4.7. Underground workplaces other than mines

4.7.1. Occupational category and workforce size

Underground workplaces other than mines mainly include civil air defence works, underground stores, limestone caves and other underground venues for leisure and tourism. According to incomplete statistics, there are more than 3700 hotels with 220 000 beds, more than 1200 stores and/or eating houses, 1500 workshops, 13 300 store rooms and 2600 cultural and sports centres developed underground and a total of 234 civil air defence works in 23 large cities across the country [35], providing more than 1 million employment

opportunities [36]. There are 161 famous caves available to tourists and numerous underground breeding farms in operation.

4.7.2. *Exposures*

Large numbers of underground buildings were constructed in China, mostly in the years 1960–1970, as civil air defence works for the purpose of preparing for war. Now, large portions of these buildings are being converted into stores and hotels. Moreover, a large number of high rise buildings have been constructed with deep basements consisting of multifloor rooms, partly for use as stores. With the rapid expansion of urban subways and river-crossing tunnels, the number of underground structures is steadily growing. In recent years, awareness of the risk of lung cancer due to radon has led to growing concerns about underground structures, with numerous studies having been undertaken [37–39].

Radon activity concentrations measured in underground workplaces are shown in Table 14. The average concentrations are generally low in underground hotels, stores, eating houses and recreation facilities, owing to good ventilation equipment, but are high in underground workshops and store rooms due to poor ventilation. In caves, radon concentrations are high as a result of the deep underground locations and the low rates of ventilation, despite the installation of good ventilation equipment. A few underground breeding farms are equipped with ventilation equipment.

4.7.3. *Dose assessment*

Following the method for estimating exposure given in Ref. [2], the average annual effective dose received by workers is estimated to be 4.54 mSv in terms of an average radon EEC of 252.3 Bq/m³ and the corresponding collective effective dose, assuming a total workforce of 1 million, is 4540 man Sv.

Due to the lack of effective ventilation, radon activity concentrations are very high in this category of underground workplace, especially in caves and underground chicken farms, which could lead to workers receiving doses greater than 20 mSv/a. With the increasing urban population density, more and more underground workplaces such as subways and underground caverns will result in an increase in the average effective dose. On the other hand, the increased awareness of radon in underground workplaces and the imposition of regulatory control have already resulted in a downward trend in occupational exposure.

TABLE 14. RADON CONCENTRATIONS IN UNDERGROUND WORKPLACES OTHER THAN MINES

	No. of sites	No. of samples	Activity concentration (Bq/m ³)		
			Range	Mean	EEC
Civil air defence works	23 cities	500	14.9–2480	247	123
Underground hotel	1	20		173.8	86.9
Underground store	41	41	46.8–538	227	114
Underground workshop	18 cities	1024	3.1–4710	479	234
Underground store room	18 cities	539	27–5250	312	156
Limestone cave	39 caves	312	20–8660	1200	494
Underground breeding farm	32	32		2070	1034
Underground cultural and sports centre	20	20	136–256	172	86
Half-underground greenhouse	6	6	67.5–128	92.7	46.3
Underground café	7	7	417–864	612	306
Underground dance hall	6	6	136–256	190	94.9
Mean				522	252

Note: EEC — equilibrium equivalent concentration.

5. OCCUPATIONAL EXPOSURE TO COSMIC RADIATION

Cosmic radiation, consisting mainly of protons and heavy charged particles, originates from outer space and gives rise to occupational exposure during air travel and space travel. The particles interact with nuclei in atmospheric constituents, producing secondary radiation products, including particularly μ -mesons, electrons, positrons, photons (gamma radiation) and neutrons.

5.1. Altitude and latitude dependence of cosmic radiation dose rates

The radiation doses received by aircrew in the course of their work vary with flight altitude and duration, latitude, solar activity cycle and solar flare events. Cosmic radiation exposures to aircrew arise mainly from the ionizing and neutron components of cosmic rays, with their fraction varying with altitude. Generally, the neutron component accounts for 60–80% at altitudes of more than 6000 m [40]. Investigations have shown that the neutron dose contribution is about three to five times higher than the directly ionizing contribution at altitudes of 915–1215 m and latitudes of 50–80°N [41]. Information on the altitude dependence of the ionizing component of the dose rates from cosmic radiation at habitable locations can be found in Ref. [42]. In recent years, the variations of neutron dose with altitude have been estimated mostly from neutron spectrometry measurements with Bonner spheres, which have a good response to neutrons, especially high energy neutrons. Measurement results need to be corrected by using the depth in the atmosphere. The influence of latitude on the neutron fluence is estimated using the Florek equation.

Relevant measurements have also been conducted in China and indicate that dose rates from the ionizing component of cosmic radiation show an exponential increase with flight altitudes and a linear increase with latitude [43, 44]. Measurements of dose rates from cosmic radiation obtained for 34 domestic flight routes during the period 1992–1993 were reported in 1997 [45]. These flight routes were between longitudes 76–127°E and latitudes 18–50°N. The measurements were made using a Model HGL-1 environmental gamma continuous monitoring meter. An exponential relationship between dose rate and altitude was established from 424 data points. Investigations indicated that dose rates varied little with longitude but increased linearly with geomagnetic latitudes within the range investigated.

Measurements made within a spacecraft flying at low earth orbit indicated that when crossing the area known as the South Atlantic Anomaly, the dose rate within the spacecraft was higher by a factor of about four compared with flying across other areas [46].

5.2. Doses received by aircrew

5.2.1. Preliminary data

Statistics for over 34 flight routes within China [45] indicate an average take-off time of 19.2 ± 5.2 min and an average landing time of 22.4 ± 4.4 min, from which an average time of 20 min can be assumed. Cruising altitudes are mostly about 10 km. Based on measurement results, a relationship between altitude and dose rate was derived by the authors, giving an average dose rate of $0.0649 \mu\text{Gy/h}$ at sea level and $1.61 \mu\text{Gy/h}$ at 10 km. It was assumed that there are domestically 3200 scheduled flights per week, with a total flying time of about 6600 h and an average flying time of 2.06 h (approximately 120 min). The take-off and landing times account for about one third of the total flying time and contribute only about 13% to the total flight dose. Aircrew on domestic flights may be assumed to be airborne for 1000 h/a, comprising 670 h/a at 10 km altitude and 330 h/a in take-off and landing, and thus receive an estimated dose of 1.4 mSv/a from the ionizing component. According to estimates for northern European countries, the dose received by aircrew from the neutron component is three to five times higher than that from the direct ionizing component. Thus, China's aircrew on domestic flights receive an estimated dose of 4.5–7.0 mSv/a. This value is regarded as an overestimate because of the assumptions made with respect to the flying time and the estimation of the neutron dose.

BF3 neutron monitors are used to determine the neutron dose for international routes. Accordingly, the neutron dose rate for the Beijing to New York flight route was estimated to be 2.94 mSv per 1000 h, representing a neutron contribution of 37.5% to the total dose rate. For the Beijing to Stockholm route, the neutron dose rate is 3.50 mSv per 1000 h, about 42.3% of the total. Cumulative dose measurements were conducted over a three month period during 1997 for Shandong Airline aircrew using thermoluminescent dosimeters (TLDs) and CR-39 neutron track detectors [47]. An annual average flying time of 1300 h was estimated for aircrew in this company on the basis of the average three month flying time of 326 h. Taking account of the CR-39 response to neutron energy, the measured neutron dose was multiplied by three to derive the estimated neutron dose. The annual dose received by the aircrew was reported to be 2.40 mSv at altitudes of 8–10 km and 1.12 mSv at an altitude of 4 km.

For 30 domestic flight routes, the ionizing component of the dose rate was estimated from measurements made with an RS-111 high pressure ionization chamber and an FD-3013 scintillation survey meter, while the neutron component was calculated in accordance with the method described in Ref. [42]. The total dose rate was in the range of $0.120\text{--}9.170 \mu\text{Sv/h}$ [48]. The flying times for the 30 flight routes were in the range of 0.8–3.75 h. The annual effective

dose received by the aircrew, determined on the basis of an annual flying time of 1000 h, was 0.12–9.17 mSv.

5.2.2. Calculation of dose using computer programs

There are several methods to determine aircrew doses. A spherical tissue equivalent proportional counter, which is one of the reference instruments, can be used to carry out direct measurements, while TLDs with a good photon response and CR-39 detectors with a good neutron response may be used to conduct passive measurements of the individual cumulative dose. However, owing to the complexities of cosmic radiation and the very wide spectra, especially the neutron spectrum of dosimetric significance, generally available instruments cannot meet the measurement requirements. Therefore, computer programs, developed on the basis of particle fluence computations, are used in conjunction with measured data to carry out the dose calculations. The computer programs should ideally be either endorsed by the relevant international organizations or approved by the national civil aviation administration [49–50]. The most commonly used computer programs are CARI-6, developed by the United States Federal Aviation Authority [51], EPCARD, developed by the German Centre for Environmental Health [52], PC Air (Canada) and FREE (Austria). These codes have data on 3-D isodose profiles at different flight altitudes accessible to the database. Aircrew doses for a particular flight can be calculated on the basis of input data such as the departure airfield, the take-off and landing times, the cruising time, the cruising height, the flight profile and the flying date. The results calculated by the various computer codes are in close agreement with each other, with a few exceptions relating to updated data on the solar activity cycle, solar flare events and the magnetosphere. In China, the CAUCARD code was developed to estimate the aircrew doses [53]. It gives results within $\pm 15\%$ of the results from CARI-6. At present, there is no domestically authorized computer program in this regard.

In 2004, the CARI-6 code was used to calculate doses for 234 aircrew of Xinjiang Airline [54] over the three year period 1997–1999. The average dose for all types of aircrew was found to be 2.19 mSv/a, with a range of 0.89–4.42 mSv/a. The following average doses were also calculated for aircrew carrying out specific types of work:

- Security personnel: 3.28 mSv/a;
- Flight attendants: 2.84 mSv/a;
- Flight engineers: 2.04 mSv/a;
- Air navigators: 1.77 mSv/a;
- Pilots: 1.72 mSv/a.

Doses for specific routes, both domestic and international, were calculated in 2006 [55] and gave the results shown in Table 15. On the basis of these results, it can be concluded that domestic routes are generally associated with an annual dose of about 2 mSv per 1000 h flying time, while the corresponding figure for international routes is about 6 mSv per 1000 h flying time.

TABLE 15. AIRCREW DOSES FOR VARIOUS FLIGHT ROUTES

	Flight duration (min)	Effective dose (mSv)	
		Single flight	1000 h of flying
Domestic routes			
Beijing—Guangzhou	180	0.006 8	2.3
Beijing—Shanghai	115	0.004 1	2.2
Guangzhou—Shanghai	120	0.003 8	1.9
Shanghai—Guangzhou	120	0.003 7	1.8
Shanghai—Chengdu	140	0.004 8	2.1
Shanghai—Kunming	185	0.006 5	2.1
International routes			
Beijing—San Francisco	460	0.043	5.6
Beijing—Tokyo	205	0.008 9	2.6
Beijing—Copenhagen	535	0.040	6.0
Beijing—Brussels	665	0.068	6.2
Beijing—Paris	650	0.066	6.2
Shanghai—Vancouver	650	0.066	6.2
Shanghai—Brussels	845	0.082	5.8
Guangzhou—Melbourne	550	0.027	3.0
Guangzhou—Amsterdam	860	0.076	5.3

NORM EXPOSURE IN CHINA

Because specific data for every flight route are not available from airlines, doses can be estimated only in terms of average individual doses and collective doses. According to the 2006 Bulletin of the Civil Aviation Administration [56], China had 350 000 civil aviation workers in January 2006, including 12 840 flight crew. There are 4023 flights per day, including 3602 domestic flights and 421 international flights. For domestic flights, owing to the lack of reported data, the number of cabin attendants was estimated from the ratio of flight crew to cabin attendants, which was 1 to 1.5 according to Ref. [57]. This estimate was supported by data provided by Air China and Hainan Airline, giving the ratios of flight crew to cabin attendants as 2875 to 6166 and 1050 to 1530, respectively, representing an average ratio of 1 to 1.8. In Ref. [2], the ratio in the United States of America is given as 1 to 1.48. So here the ratio is taken to be 1 to 1.5. On the basis of 12 840 flight crew, it can be concluded that China has 19 260 cabin attendants, giving a total of 32 100 aircrew. Based on the domestic to international ratio of 3602 to 421 quoted in Ref. [57], domestic routes account for 89.5% of these 32 100 workers. Assuming an annual average flying time of 1000 h, the annual collective dose received by aircrew (pilots, air navigators, flight attendants, security personnel, flight engineers and communications officers) was estimated to be 60.5 man Sv during 1986–1990 and 77.7 man Sv during 1991–2006, as shown in Table 16. These collective doses are approximately one tenth of the annual collective dose of nearly 800 man Sv received by aircrew globally (see Table 17).

TABLE 16. COLLECTIVE DOSES RECEIVED BY AIRCREW, CHINA

	No. of aircrew	Annual average effective dose (mSv)	Annual average collective effective dose (man Sv)
1986–1990	25 000	2 ^a 6 ^b	60.5
1991–2006	38 400	2 ^a 6 ^b	77.7

^a Domestic.

^b International.

TABLE 17. DOSES RECEIVED BY AIRCREW, BY COUNTRY

	Year	No. of aircrew			Average effective dose (mSv)	Collective dose (man Sv)
		Flight crew	Cabin attendants	Total		
USA	1985	46 000	68 000	114 000	3.51	400
UK	1991			24 000	2.09	50.0
Finland	1990–1994			1 930	1.96	3.78
Bulgaria	1990–1994			1 400	2	2.80
Worldwide	2000			250 000	3	750
China	2006	12 840	19 260	32 100	2.42	77.7

It should be noted that the annual collective doses for China were not derived using internationally agreed methods, in terms of which the dose is estimated using all flight data provided by all airlines and computer programs. Instead, the individual doses to typical aircrew members were first estimated using computer programs and based on typical domestic and international flight data with an assumed annual flying time of 1000 h, and then the annual collective dose to aircrew in China was derived by applying the individual doses to the total numbers of aircrew. Domestic flight cruising altitudes, although generally lower than international altitudes, are dependent on the type of aircraft. On the one hand, owing to the large distances between cities in China, the type of aircraft used is often the same as that used for international flights, at a high cruising altitude. On the other hand, while there are 32 100 aircrew in total, only a portion of them are actually involved in flight tasks. At present, there are only 1000 commercial aircraft in the country, with 4100 flights per day. Assuming an aircraft is staffed with 2 flight crew and, at most, 20 cabin attendants, the total aircrew who are flying can be estimated to be fewer than 22 000 individuals.

Reference [58] reports on the measurement of flight crew and cabin attendant exposures on polar and non-polar flight routes during the period 1 October 2006 to 30 September 2007. Individual external exposure dosimeters, including four-element, optically stimulated luminescence dosimeters and CR-39 track detectors, were used to determine the quarterly cumulative doses due to penetrating radiation, $H_p(10)$, from photons and neutrons. The Beijing to New York flight route was selected as the polar route, for which 6 flight crew and

12 cabin crew were monitored. On non-polar flights, 20 flight crew and 20 cabin attendants were monitored, together with 20 ground crew as a control group. The doses over the course of a year were 5.79 mSv for aircrew on polar flights, 2.14 mSv for aircrew on non-polar flights and 0.62 mSv for ground crew. The corresponding doses per 1000 h were 3.10 mSv for aircrew on polar flights and 2.21 mSv for aircrew on non-polar flights.

It can be concluded from both the measured data and the computer code calculation data that aircrew doses exceed the public dose limit of 1 mSv/a and are possibly comparable with those received by radiation workers in other occupational categories. Without route rotation, aircrew on international routes would receive higher doses than workers at nuclear power plants.

5.3. Exposure of space crew

Finally, a brief reference will be made to the cosmic radiation dose received by astronauts and cosmonauts in China. By 2008, a total of 6 cosmonauts had entered space in the Shenzhou series spaceships, with a cumulative flying time of 462 h (21 h \times 1 person, 115.5 h \times 2 persons and 70 h \times 3 persons). The dose rate within the spacecraft cabin at low earth orbit from cosmic radiation of galactic origin is about 0.18 mSv/d [59]. Accordingly, the collective dose to cosmonauts in China may be 0.003 47 man Sv.

6. EXPOSURE TO INDOOR RADON

The earliest measurement of radon activity concentration in air was made at Tsinghua University in 1928 by Ganchang Wang, Academician of the China Academy of Science (CAS). The aim was to study the relationship between radon activity concentration and atmospheric phenomena. In 1958, a formal radon survey of a local area was made by Deping Li (Academician of CAS), Ziqiang Pan (Academician of the China Academy of Engineering) and their group in the China Institute of Atomic Energy (CIAE), for the first loading of nuclear fuel of the first nuclear research reactor in China, a multipurpose heavy water research reactor. In the 1980s and 1990s, the Public Health Ministry and the Environmental Protection Ministry carried out a national survey on radon activity concentrations. A more recent indoor radon survey was made during the period 1995–2005 by the National Institute of Radiation Protection (NIRP). This survey covered 26 typical cities and areas and 3098 rooms were measured. The indoor radon activity concentrations were found to be about 70% higher than those measured in the early 1990s. In order to confirm the NIRP results, two further national surveys were carried out by the CIAE, universities and institutes

during the period 2000–2010, with a coverage of up to 4166 rooms in up to 38 cities. The radon concentrations ranged from 5.3 to 398.1 Bq/m³, with mean values of 32.6 and 38.2 Bq/m³ (sampling weighted) and 30.7 and 35.8 Bq/m³ (population weighted). These new results showed a 90% increase in the mean radon concentration compared with the value determined in the early 1990s.

7. CONCLUSIONS

Occupational exposures to natural sources, including NORM, have been assessed throughout China. During the period 1996–2000, more than 10 million workers were being monitored and the annual average individual and collective doses were 2.1 mSv and 22 300 man Sv, respectively (see Table 18). Workers in coal mines represented the largest group of workers (61.4% of the total) and received the largest fraction of the collective dose (65.3% of the total). Workers in other non-metal mines were the next largest group (28.3%), while workers in metal mines received the highest annual average individual dose (5.53 mSv) and the second largest fraction of the collective dose (24.8% of the total).

TABLE 18. SUMMARY OF OCCUPATIONAL EXPOSURES TO NATURAL SOURCES, 1996–2000

	No. of monitored workers	Annual av. individual dose (mSv)	Annual av. collective dose (man Sv)
Coal mines	6 500 000	2.40	14 600
Metal mines	1 000 000	5.53	5 530
Other mines	3 000 000	0.688	2 060
Underground workplaces other than mines	50 000	1.56	78.0
Aircrews	38 400	2 ^a 6 ^b	77.7
Overall	10 588 400	2.1	22 300

^a Domestic routes.

^b International routes.

The average dose arising from exposure to natural sources has increased by more than 30% since the 1990s. Workers in the coal mines of China are the largest group of coal mine workers in the world (6 million) and receive an annual average effective dose of 2.4 mSv. In all, more than 10 million workers are employed in occupations involving enhanced exposure to natural sources and receive an annual average effective dose of 2.1 mSv. Relatively high radon concentrations are found in some other workplaces such as hot spring resorts and underground workplaces other than mines. There is a need for further investigation of exposures to NORM in the oil and gas industry.

In 2010, the population weighted, indoor radon concentration in typical cities throughout China was 35.8 ± 9.7 Bq/m³, an increase of 90% over the value determined in the 1990s.

In past years, insufficient attention has been given to the investigation of exposures to natural sources in a systematic manner. In particular, little consideration has been given to such exposures in the management approach to occupational exposure. Further efforts are needed in this direction in view of gaps in the available data and some uncertainties in the existing data.

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Invited Paper

THE NORTHERN TERRITORY'S URANIUM INDUSTRY: PAST, PRESENT AND FUTURE

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Abstract

Uranium and the Northern Territory (NT) of Australia have a long historical association. Currently, all phases of the uranium production cycle are active throughout the NT. More than 65 exploration authorizations under the Mining Management Act refer to uranium as the mineral of interest. Several of these prospects could be developed within the next few years. Also, the world's fifth largest operating uranium mine (ERA's Ranger Mine) is currently undertaking a significant exploration programme. In addition, there are remediated sites from earlier cycles of uranium production as well as some legacy sites that are the subject of active characterization and planning for remediation. The paper provides an overview of the NT's uranium mining history and the current situation in the industry locally, before commenting on the future for the industry and briefly explaining the current regulatory process operated by the NT Government's Department of Mines and Energy.

1. INTRODUCTION

The first recorded history of uranium in the Northern Territory (NT) of Australia was in 1869, when Wood, a member of Goyder's survey party, noted that, while working in the area known as Rum Jungle, they had found a green coloured mineral "that was not copper". It was recorded by Barrie (see p. 142 of Ref. [1]) that in 1912, the Government Geologist, H.L. Jensen, had reported the existence of uranium in the Rum Jungle area. There had been some mining of copper from small, shallow deposits between 1905 and 1907 and some drilling for copper in 1913 [2] but essentially the uranium story stayed quiet until 1948, when the Commonwealth Government announced a maximum reward of £25 000 would be paid for a significant uranium discovery located in Australia or its territories. On 11 August 1949, J.M. White reported the discovery of uranium mineralization at Rum Jungle and later received the reward. Other discoveries followed in the area of the Pine Creek geosyncline at Adelaide River and Fleur

de Lys and, in 1952–1953, the South Alligator Valley. The stories of these earlier discoveries have been well documented by Annabel [3]. Other discoveries were found to the east near the Queensland border and, in the 1960s, major discoveries were made in the Alligator Rivers Region (ARR) at Ranger, Nabarlek and Koongarra. Thus, since 1953, uranium mining has been more or less continuous in the NT.

2. EARLY MINING

Of the early discoveries the two most significant operations were those at Rum Jungle and the South Alligator Valley.

2.1. Rum Jungle

The mining at Rum Jungle was undertaken by the company CRA (now Rio Tinto) as an agent for the Commonwealth Government from 1953 until 1958 at the White's and Dyson's ore bodies, and by Davis Contractors Ltd from 1961 to 1963 at Rum Jungle Creek South, a satellite deposit about 10 km away from the main site. The processing plant at Rum Jungle was opened in 1954 and originally recovered uranium from the leaching circuit using ion exchange technology. In 1962, this changed when the plant became the first in Australia to adopt solvent extraction methods. The Rum Jungle mine produced 3530 t of uranium ore concentrate (UOC) and 20 222 t of copper between 1954 and 1971 [4]. The site was abandoned after some remediation in 1976; however, the nearby East Finnis River became polluted as a result of acid mine drainage arising from the waste rock dumps at the site. Between 1983 and 1986, the site was remediated to what were considered to be the highest standards of the day at a then cost of \$A18.6 million. In more recent times it has been accepted that the remediation strategies had not been sustainable and the site had become a source of pollution again. In 2009, a National Partnership Agreement (NPA) was entered into by the Commonwealth and NT Governments whereby it was agreed that a series of studies would be undertaken to better characterize the site and these data would be used to produce an updated remediation plan. The NPA was signed in 2009 and is due to end on 30 June 2013 at a cost of about \$A7 million. The Mining Environmental Compliance Group of the NT Department of Resources is responsible for the overall management and coordination of the programme.

2.2. South Alligator Valley

In the early 1950s, several explorers were searching for uranium in the highly prospective areas of the Pine Creek geosyncline [3]. One of the most successful locations was the upper South Alligator Valley, where some 54 radiological anomalies had been identified through crude airborne surveys using Geiger tubes hanging under a biplane. After follow-up ground work and some exploration drilling, 13 mines were developed, albeit all on a rather small scale. The total production from these mines, which operated between 1959 and 1965, was about 850 t UOC [5]. The mines were simply abandoned when the supply contracts had been filled, as was the usual practice at the time. In the late 1980s, the area was designated to become Stage 3 of the World Heritage listed Kakadu National Park, and so a programme of hazard reduction works was undertaken to reduce physical and radiological risks before the area was opened up to tourism. The works were undertaken at the sites in the dry seasons of 1991 and 1992 [6] and involved, among other things, closing off adits, putting up warning signs, burying scrap metal and contaminated materials and restricting public access to open cuts with fencing.

As a result of a successful land claim by the Aboriginal Traditional Owners (ATO), the area was returned to them in 1996. However, there was an agreement that the land would be immediately leased back to remain as part of Kakadu National Park. One clause of the lease required that all evidence of former mining activity be remediated by the Commonwealth Government. Work on planning began with a series of consultations in 1997. By 1999, a system of stakeholder consultation had been well established and the process of data gathering had begun in preparation for the development of the remediation plan [7]. In April 2003, the plan for dealing with non-radiologically contaminated sites had been agreed in principle (Part A) and some interim work was undertaken to deal with localized issues of contamination. Further studies were needed to provide data for use in the design and standards and funding had to be agreed with the Commonwealth Government. In the dry seasons of 2007 and 2008, the Part A sites were remediated. Also, in 2008 the Part B plan was agreed and the containment site selected. In 2009, the new containment was built, residues were collected from the various sites, material from the hazard reduction containments was relocated and all placed in the new containment. The work was completed on time and on budget and significantly the ATO had been involved at every stage of the process, including driving earthworks machinery and revegetation work as well as continuous consultations and providing on-site cultural advice [8].

3. MODERN MINING

After the rush in uranium activity immediately after World War II and then an apparent slump, a flush of exploration followed in the late 1960s and 1970s that resulted in the discovery of four significant deposits at Ranger, Koongarra, Jabiluka and Nabarlek, all located in what was soon called the ARR. In 1977, the Government announced that uranium mining would begin at all deposits but only Ranger and Nabarlek were established before a change of Commonwealth Government in 1983. The new Government introduced a policy in 1984 which stopped the development of new uranium mines until 1996, when another change of Commonwealth Government allowed new developments to be considered again.

3.1. Nabarlek mine

The Nabarlek mine was the first new uranium mine in Australia to be developed under modern environmental impact assessment procedures [9]. The deposit was discovered in 1970. The site operated from 1979 until 1988 and had a number of unusual features. The remote operation was an early fly-in/fly-out site with no associated mining town, this was because the site was in Aboriginal country and the plan was to keep social impacts to a minimum. At 2% triuranium octaoxide (U_3O_8), the ore body was considered high grade by the standards of the day, although this figure pales by comparison with some mines in the Athabasca basin of Canada. The most significant feature of the operation was possibly the way in which it was mined in one 143 day campaign in the dry season of 1979 and the ore stored on a pre-built stockpile pad. The milling campaign ran until 1988 with the tailings being disposed of directly to the mined-out pit, possibly a unique situation. Even the processing was different. The initial process used pyrolusite as an oxidant, but within a year the company adopted Caro's Acid as the lixiviant for the remainder of the mine life. Over the life of the mine the total production was 10 858 t U_3O_8 . There was hope that a new ore body might be found which would have extended the life of the mill but this did not happen. The site was remediated in 1995 with the final earthworks and seeding completed as the wet season began. The revegetation of the site has been disrupted by fires and a destructive cyclone in 2006. Also, exploration at the mine site and the surrounding area began again in the early 2000s, when Uranium Equities Limited acquired the site. Exploration is ongoing, although to date no significant new resources sufficient for development have been announced.

3.2. Jabiluka and Koongarra projects

Both these ore bodies were discovered in the region and were considered in the work of the Ranger Uranium Environmental Inquiry [10], also known as the Fox Report after the chairman, Mr Justice Fox. Neither project was far enough advanced to avoid being stopped by the change of Government in 1983.

The Jabiluka deposit was discovered in 1971–1973. It is located 20 km north of the Ranger mine and the mining lease abuts the Ranger Project area. Jabiluka was owned by Pancontinental Mining and has a resource in excess of 130 000 t U_3O_8 . In 1991, Energy Resources of Australia (ERA) acquired the Jabiluka site and, following the change of Government in 1996, began a development programme. The ERA completed approximately 2000 m of decline and underground workings and carried out some underground exploration as well as building some support infrastructure at the surface. However, although the environmental impact statement (EIS) was close to final approval the project was halted and the site placed on care and maintenance following discussions with the Aboriginal traditional owners of the land. The underground workings were backfilled and the site remediated apart from a large water management pond. It was agreed with the traditional owners that further development would only take place with their explicit and specific consent. The removal of the pond structure is currently under discussion.

The Koongarra deposit was discovered in 1970 by Noranda Australia Ltd and then passed to Denison Australia Pty Ltd. A final EIS was approved in 1981 but the development was unable to start before the change of Government in 1983. This prevented any further progress on the project. The resources of the two ore bodies total about 14 540 t U_3O_8 . Since 1995, the deposit has been owned by a local subsidiary of the French company Areva. The ATO has expressed his wish that there be no development and the area was placed on the World Heritage Register in 2011, a status it shares with the surrounding Kakadu National Park. It seems unlikely that development will proceed there.

3.3. Ranger uranium mine

The Ranger uranium deposit was discovered by Geo-Peko in 1964 as a result of a radiological anomaly spotted during airborne surveys being undertaken on adjacent tenements by Noranda. The site was finally brought into production in 1981 with an initial production capacity of 3300 t/a U_3O_8 . The first open pit (Pit 1) was mined from 1981 until December 1994. In May 1997, mining operations recovered the first ore from Pit 3, which was due to be mined out by the end of 2012. In August 1997, the potential maximum production capacity was upgraded to around 6000 t/a. The average annual production from 2002–2003 to

2009–2010 was 5147 t U₃O₈. In 2010–2011, the mine was obliged to shut down for six months as an exceptional wet season deposited over 2000 mm of rain in six months, which meant there was insufficient storage capacity for process water on site and discharge is not permitted. By early 2012, the mine was getting back into full production. However, the mining of primary ore from Pit 3 ended in November 2012. The backfilling of Pit 3 with low grade mineralized material began in December 2012 as an integral part of the long term remediation strategy which requires all tailings to be disposed of below ground level.

The mine has produced over 100 000 t U₃O₈ since it commenced operations and was generally regarded as the second most productive uranium mine in the world until the events of 2010–2011. Although production is getting back to historic levels, there will probably be a decrease after a year or two as primary ore stocks are depleted and milling moves to lesser grades from stockpiles. On the present schedule, stockpile milling could continue until 2020, assuming no further viable ore bodies are located on the project area.

The ERA recently began an intensive exploration programme across the whole of the Ranger Project Area which is aimed at locating new resources to extend the mine's life. While some of this programme is conventional drilling from the surface, the company launched a major underground exploration programme in May 2012 with the commencement of an exploration decline to examine the potential of an ore body known as Ranger 3 Deeps. The early expectation is that the resource could amount to at least 34 000 t U₃O₈. The programme has a budget of \$A120 million, and the portal was completed in October 2012, when hard rock tunnelling began. The decline is expected to be approximately 2000 m in length and will descend to a depth of 350 m. In addition to the decline itself there is planned to be up to 20 000 m of underground exploration drilling and possibly some other trial excavation work.

Ranger currently is constructing a \$A220 million brine concentrator as part of the strategy to reduce the inventory of process water on site. The project is due to be commissioned in the second half of 2103 and will reduce the process water volumes inventory by more than 60%. The residual brines will be stored in the lower portions of Pit 3, encapsulated below the tailings. By law, all uranium mill tailings in the ARR are required to be disposed of below ground and, at Ranger, Pit 1 was filled with tailings from 1997 until the fourth quarter of 2008, at which time the above ground tailings storage facility was brought back into service. This structure has functioned since the initial mining at Ranger and comprises a dam approximately 1 km² designed and built as a water retaining structure. It has been lifted several times since 1981, and the last lift was completed in late 2012. The dam will be the temporary tailings storage facility until the mining operations cease, at which time the contained tailings will be transferred to Pit 3

for disposal. The dam will eventually be decommissioned and demolished and the site remediated.

4. OTHER NORTHERN TERRITORY URANIUM RESOURCES

4.1. Deposits

The NT has a number of other identified, but as yet unexploited, uranium deposits which have been explored to varying degrees over the years. The main deposits of interest are shown in Table 1.

TABLE 1. SIGNIFICANT OTHER URANIUM DEPOSITS IN THE NT

Deposit	Grade (% U ₃ O ₈)	Contained U ₃ O ₈ (t)	Category
Mount Fitch	0.046	6 600	Resources
Angela	0.1	10 250	Resources
Bigrlyi	0.082	12 200	Indicated and inferred
Nolans Bore	0.02	3 977	Resources
Napperby	0.036	3 350	Inferred

With one exception the deposits are either under care and maintenance due to the current state of the uranium market or still being explored and proved up. The exception is the deposit at Nolans Bore which is primarily a rare earth oxide project which has associated uranium and the proponent is considering recovery of the uranium as a co-product, although a final investment decision has still to be made. There are also a number of other rare earth prospects under investigation, some of which could have associated uranium deposits. There are also other mineral deposits such as phosphates where uranium could also be present at grades warranting further investigation for exploitation as a co-product or by-product

4.2. Exploration

At present, over 370 authorizations are administered by the Mining Environmental Compliance Division (MECD) of the NT Department of Mines and Energy. Of these, 69 are related to the mining of uranium (one being Ranger) or exploration of uranium. Many of the exploration operations were started up by junior explorers in the rush of 2004–2007 when the spot market price of U_3O_8 surged. Sadly, many of these operators have found it hard to obtain further funding since the fall in price, and the global financial crisis has made it even harder. While uranium exploration may be found in locations all over the NT, the main areas of interest are unsurprisingly associated with former uranium mining areas. The Pine Creek geosyncline (which runs from Rum Jungle to the Coberg Peninsula), West Arnhem Land, the areas around Alice Springs and to the north east across the Tanami Desert and in the central east area of the Barkly Tablelands towards the Queensland border are all locations where uranium exploration is active. Some stories are circulating about the possibility of significant new finds, but little has been reported to the Australian Stock Exchange in the way of reserves and resources as yet.

5. REGULATION OF URANIUM MINING

The regulation of uranium mining in the NT is primarily undertaken by the Department of Mines and Energy. Two main units are involved, the Titles Division and the MECD. There are also several other NT Government agencies involved, as well as some Commonwealth Government agencies, particularly in the special case of uranium mining activity in the ARR. The main organizations involved, other than the MECD, are: Worksafe NT, which administers laws relating to occupational health and safety on mine sites; NT Health, which is responsible for administering the National Radiation Dose Rate Register for uranium miners in the NT; and the independent NT Environment Protection Agency, which has responsibility for discharge licences from mine sites. The MECD works in close cooperation with all these agencies. In addition, for uranium mines in the ARR there are special arrangements which are described in a later section.

5.1. General uranium mining

In the first instance, mine operators have to obtain title to the land before they can undertake any form of mining activity, especially if it involves environmental disturbance. The administration of access to land and granting of mining tenements is handled by the Titles Division, which administers the Mineral Titles

Act, which was last updated in 2011. Once title has been established, the operator has to submit a detailed Mining Management Plan (MMP) for assessment by the MECD. This group administers the Mining Management Act (MMA), which was last amended in 2012. The MMP has to include not only the work plan for the next 12 months but also a report on activity from the previous 12 months and a fully costed remediation plan for the works described in the MMP. Once the MMP has been assessed and approved by the MECG, which may require a number of iterations to bring the MMP up to standard, the operator is required to deposit a 100% security bond with the MECD either as cash or an unconditional bank guarantee. This bond will only be handed back by the MECD once remediation of the project has been completed to the satisfaction of the inspecting officers. At the moment, the NT is the only mining jurisdiction in Australia which requires a 100% security bond for all mining operations, including exploration and the extractive mineral industry. The bond is revised annually within the MMP at the time of renewal. The amount of money held as mining remediation security has risen from \$A38.2 million in 2005 when the legislation was introduced to about \$A744 million in mid-2012; and this does not include money for the Ranger Uranium Mine which is held in a separate trust account administered by the Commonwealth Government. Once the security deposit is paid, the operator is issued with an authorization and mining activity can commence.

During the life of a mining project, the MECD maintains a watching brief through a programme of field inspections and audits as well regular assessment of periodic monitoring data and the annual review and assessment of the updated MMP. Every effort is made to ensure that each uranium exploration project is inspected at least once a year, while mines generally are under greater scrutiny. Other changes to the MMA which came into force in 2012 include the introduction of an infringement notice system for minor regulatory transgressions and the requirement for mines to submit an annual Environmental Mining Report (EMR), which will be an integral part of the MMP. Once the MMP has been approved, this EMR will become a public document published to the internet. The minister can also require a mining company to make some incident or other reports available to the public.

5.2. Uranium mining in the Alligator Rivers Region

Notwithstanding the need to meet all the requirements of the MMA, the uranium mines of the ARR have a somewhat more intense regulatory regime which involves a significant number of additional concerned parties and some very specific processes. This arises mainly through the ownership of the uranium resting with the Commonwealth Government and the need for a number of other agencies and departments to be involved as well as a much more active

involvement of the ATO and their representatives on a day to day basis. The location of the Ranger Mine, surrounded completely by the World Heritage listed Kakadu National Park, requires that the environmental management of the operation is of the highest order at all times in order to protect the park and the RAMSAR listed wetlands downstream of the operation. As Kakadu is one of the few sites listed for both natural and cultural heritage, this presence of the ATO is integral to the process of efficient regulation.

The day to day regulation of the ARR uranium mines rests with the MECD, but it is carried out in a consultative manner through a series of Minesite Technical Committees (MTC). Each active uranium mining lease in the ARR has an MTC (Ranger, Jabiluka and Nabarlek). The membership of the MTC for a mine comprises: the relevant mining company; the MECD (chair of the MTC); the Supervising Scientist Division (SSD) of the Commonwealth Government Environment Department; the Northern Land Council, which acts on behalf of the ATO in the northern half of the NT; and, in the case of Ranger and Jabiluka, the Gundjhemti Corporation which represents the specific Traditional Owners of those two mine sites. The Commonwealth Department of Resources, Energy and Tourism, which controls export licences through the Atomic Energy Act, sits as an observer at MTC meetings. The MTC meets every two months for routine discussions of monitoring data and activities on the mines as well as providing a forum for the mining company to introduce project proposals, which can then be discussed by all the major concerned parties at once to establish initial ideas about the proposal. Much discussion also takes place outside the formal sessions.

The same organizations each send a representative as a member of the monthly inspection team at Ranger Mine. This ensures that any issues are identified and understood immediately by the members of the team without the need for protracted discussions and explanations. The inspection programme has a number of regular components as well as a structure which ensures that every significant aspect of the operation is inspected at least once in the annual cycle. For example, the tailings dam is inspected at least twice annually: once at the end of the dry season and once at the end of the wet season. The Jabiluka site is inspected between two and four times per year and Nabarlek usually twice — again related to seasonal issues such as erosion and weed control and maintenance. Monitoring is undertaken at all three sites on specific schedules with SSD and MECD both undertaking check monitoring to verify the appropriate programme undertaken by the relevant mining companies.

There is also an inspection and audit programme for exploration operations which is undertaken cooperatively by the core MTC member organizations. Each operation will be visited at least once per year and the decision on audit or inspection relates to the age, size and scope of each operation. The logistical issues involved in visiting some sites restricts visits to once per year in some cases.

Additional inspections and investigations may be carried out at any time or in relation to a specific incident or accident. Workplace safety issues are regulated by NT Worksafe, which also regulates the transport of UOC (yellowcake). Matters of radiation safety for workers are also dealt with by NT Worksafe, although the NT Health Department is the agency responsible for coordination in the NT of the Uranium Miners National Radiation Dose Register, a national safety programme for all uranium mine workers that has been in place nationally since 2012.

In addition, the ARR has two standing committees set up under Commonwealth Government legislation and administered by the SSD. The ARR Advisory Committee which meets twice per year is an information exchange forum for a very broad range of concerned parties including all the MTC members, other Government agencies (both NT and Commonwealth), organizations such as non-governmental organizations (NGOs), the Jabiru Town Council and a worker representative from the Ranger mine, and representatives of the various exploration companies looking for uranium in the ARR. The committee has an independent chairperson and the secretariat is provided by SSD.

The ARR Technical Committee (ARRTC) is made up of the MTC members and a selection of independent scientific experts, one of whom is the chairperson, in various fields relevant to the research being undertaken in the ARR that is relevant to the possible environmental impacts of uranium mining. There is also an independent scientist representing the NGOs. This committee usually meets twice a year and reviews scientific research programmes being carried out in the ARR by all the relevant organizations from the MTC membership as well as Parks Australia and other scientific organizations having relevant research programmes within the region. The ARRTC thus assists in identifying key knowledge needs and setting priorities for research programmes, particularly in relation to operational aspects of monitoring and impact assessment as well as the remediation and long term monitoring of uranium mine sites.

6. THE FUTURE OF NORTHERN TERRITORY URANIUM MINING

Notwithstanding the impacts of the Fukushima disaster on the nuclear power industry in the short term, the demand for uranium has continued to grow and electricity demand is still increasing globally as many States push ahead with nuclear power plant design and construction. The fuel for these developments will require ongoing supplies of the raw material, uranium. Demand at present exceeds supply, such that only 85% of the world's annual requirements are met from annual mine production [11]. Secondary sources and stockpile supply the balance. However, stockpiles are dwindling and some secondary sources, such

as the downgrading of Russian weapons grade material, are likely to close down over the short term and not be replaced. However, the current market prices and conditions for uranium favours those already established in the uranium production cycle as development costs for new 'greenfield' mines are likely to be more than some new projects could bear.

In this situation, the NT is well placed to supply some of this increasing demand. The main player will obviously be the ERA. The company is already well established in the industry, and if the Ranger 3 Deeps project should prove to be at least as good as the present estimates, then production to the end of the planned the mine life will be maintained at historical levels. Any extension beyond the present plan would be subject to all the necessary approvals and agreements being completed in good time. The smaller nature of some of the NT's uranium deposits may militate against their development in the present market, but expectations are that prices may rise, as a production/demand squeeze is likely to develop within five years or so. Also, by- or co-product production may be very much an option for some projects where the primary product will be fetching good prices and the present uranium production costs is marginally too high. Again, the larger uranium deposits of the NT, say 10 000 t and above, may well be attractive for development where buyers are looking for greater diversity of supply over the longer term as a means of improving energy security.

Further exploration is quite likely to result in more significant deposits being found; some of which could be worthy of immediate development, subject to all the approvals and agreements being in place, given the possible market scenarios set out above. The issue will be timing. Some States have been able to reduce the time for nuclear power plant development, so that from first concrete pour to the reactor reaching criticality is little more than five years. Few uranium mines have been developed within ten years of their initial discovery. With such long lead times market analysis and prediction are very important activities which need to be undertaken by operators and regulators alike to ensure that both parties will be ready to play their parts at the appropriate time.

7. CONCLUSION

Australia's NT has a long and cyclic history of uranium mining which goes back more than 50 years and is still very active today. The early days saw many small deposits worked and little remediation put in place as companies and governments rushed to supply material to post-war nuclear programmes. The next round of exploration in the 1960s was for uranium that would fuel nuclear power plants and provide the world with clean, cheap electricity. However, by the 1970s environmental issues had come to the public's attention and environmental

impact assessment laws were coming into place to control development and ensure that remediation and sustainability became part of the equation when plans for new uranium mining projects were being assessed. The market price surge of the early 2000s saw a new round of exploration begin with more than 60 companies involved and new standards of regulation and remediation were applied and enforced.

Today, the NT is home to one major currently producing uranium mine, a series of successfully remediating former uranium mines and an extensive series of uranium exploration campaigns including some identified resources that are presently awaiting development decisions. Changes in market conditions that are anticipated over the next few years will probably see some of these prospects come closer to development as the supply and demand situation alters again in favour of the suppliers.

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NORM IN INDUSTRIAL ACTIVITIES

(Topical Session 1)

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Invited Paper

OVERVIEW OF INDUSTRIAL NORM IN THE UNITED STATES OF AMERICA WITH EMPHASIS ON THE PHOSPHATE INDUSTRY

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Abstract

The IAEA identified 12 industries affected by NORM that had facilities and activities needing to be considered under the revised IAEA Basic Safety Standards (BSS): mining and processing of uranium ore; rare earths extraction; thorium extraction and use; niobium extraction; non-uranium mines; oil and gas; phosphate industry; zircon and zirconia; titanium dioxide pigment production; metals production (aluminium, copper, iron, lead, tin and zinc); coal combustion; and water treatment. Of those 12 industries, all have been practised in the United States of America, some are still active and some idle industries could be reactivated. The phosphate industry is unique in that it not only represents one of the 12 primary industries, but is also under consideration as a source for co-extraction of 3 other primary products on the list: uranium, rare earths and thorium. As an industry of significant global importance in food security as well as energy security, a consistent and coherent regulatory approach to NORM applied to the comprehensive extraction of resources is vital to the sustainability of mining and processing of phosphate rock. Moreover, the risk to workers and the public exposed to NORM as a consequence of processing and the use or disposal of co-products and residues must be weighed against the risk of creating critical materials shortages through the inappropriate application of regulatory controls. In such cases, an evidence based approach to risk management should be incorporated into a long term strategic resource management plan.

1. INTRODUCTION

Many mining and processing industries in the United States of America involve NORM as a consequence of extracting or using raw materials to make a variety of products. In most cases, the NORM is incidental to the process, but in a subset of industries one or more NORM constituents may be the products. Consequently, these industries are subject to federal, state and local regulations dealing with NORM. As we learned at the Sixth International Symposium on

NORM, in Marrakesh, NORM regulations in the United States of America were described by a legal expert as “uneven and fragmented” [1] and are therefore inconsistent and in some cases arguably incoherent. Jurisdictional issues arise at the federal level between the Environmental Protection Agency (EPA) and the United States Nuclear Regulatory Commission (NRC), which disagree about where and when to apply regulatory controls. There is vast inconsistency at the state level, with many states having no specific NORM regulations whatsoever, and significant differences between the ones that have such regulations. To add to the complexity, there are even local regulations (usually at the county level) that affect some companies but not others in the same industry.

The IAEA has identified 12 industries affected by NORM, 5 of which are the subject of IAEA publications in the Safety Reports Series [2]. There is an implied degree of radiological urgency in the presented order from the most urgent at position (1) to the least at position (12), but only preliminarily so considering that an in-depth review of most of the industries had not been started at the time the list was compiled. The IAEA list of NORM industries is as follows:

- (1) Mining and processing of uranium ore;
- (2) Extraction of rare earth elements;
- (3) Production and use of thorium and its compounds;
- (4) Production of niobium and ferroniobium;
- (5) Mining of ores other than uranium ore;
- (6) Production of oil and gas;
- (7) Manufacture of titanium dioxide pigments;
- (8) The phosphate industry;
- (9) The zircon and zirconia industries;
- (10) Production of tin, copper, aluminium, zinc, lead, and iron and steel;
- (11) Combustion of coal;
- (12) Water treatment.

Only 2 of these 12 industries directly target NORM as a product. For the other ten industries, NORM is incidental. However, one industry on the list has the ability to not only supply its own products, but also the products of two other important industries on the list. The phosphate industry has produced triuranium octaoxide (U_3O_8) in the past and may do so again in the United States of America. Furthermore, the phosphate matrix includes rare earth elements (REEs) and a small amount of thorium. Economic factors have encouraged new research into the extraction of REEs from phosphate mining and processing products and tailings including phosphoric acid and phosphogypsum. This potential for comprehensive extraction of multiple resources from one mining practice will be discussed later in this paper. The more specific categories in the IAEA list will

be discussed herein. The broader categories of ‘non-uranium mines’ and ‘metals production’ are omitted due to their non-specificity and in the interest of brevity.

2. URANIUM

The uranium information provided in this paper is very general and focused on total masses of products and wastes. Detailed information can be found in Ref. [3].

2.1. Uranium deposits

Economically exploitable uranium deposits occur mainly in western United States of America: primarily Utah, Colorado, Wyoming, Arizona, South Dakota, New Mexico and Texas. Although regionally variable, the mineralized overburden averages about 0.92 Bq/g for ^{238}U through to ^{226}Ra in equilibrium; ~0.66 Bq/g for radon progeny; 0.037 Bq/g for the ^{232}Th series to thoron; and 0.048 Bq/g for the ^{235}U series to actinium [4]. Historically, the total mass of excavated overburden varied but was of the order of 40 billion t/a, while the total mass of other mine waste was about 4 billion t/a.

2.2. Current U_3O_8 production

Five underground mines and five in-situ leach (ISL) sites are operating in Utah, Nebraska, Texas and Wyoming. These sites produced a total of 1.86 million kg of U_3O_8 in 2011 [5]. Production in the United States of America has been cyclical, with roughly 1 million kg of U_3O_8 produced in 1993, increasing to over 2 million kg in 1998, dropping to 1 million kg in 2003 and climbing again to 2 million kg in 2006 before slowing to the current rate. New ISL mines are planned for Colorado, New Mexico, South Dakota, Texas and Wyoming.

ISL technology is more economical and environmentally sound than traditional mining for permeable ore bodies. No tailings or waste rock are generated in the ISL process, so large volumes of NORM residues are avoided. Most of the ore body’s radioactivity remains well underground with minimal increase in radon release and no dust generation. After the uranium is recovered, the barren solution is treated and returned to the well field, but about 0.5% of the water, which contains radium, is placed in disposal wells.

3. RARE EARTH ELEMENTS

Detailed information about the radioactivity in REE processing and waste streams can be found in Ref. [6] for mineral sources containing thorium.

3.1. Rare earth deposits

Historical mining sites can be found in California, Idaho, Montana, Wyoming, Colorado, New Mexico, Nebraska, Missouri, Illinois and New York. Large placers exist in Idaho, North Carolina, South Carolina, Georgia and Florida.

3.2. Mineralogy

Bastnäsite (a carbonate) contains 70–74% rare earth oxides (REOs), up to 0.3% thorium oxide (ThO_2) and up to 0.9% uranium dioxide (UO_2). Monazite (a phosphate) contains 35–71% REOs, up to 20% ThO_2 and up to 16% UO_2 . Xenotime (a phosphate) contains 52–67% REOs, little ThO_2 and up to 5% UO_2 [7].¹

3.3. Production in the United States of America

All production in the United States of America in 2012 took place at Mountain Pass, California. In that year, 7000 t of REOs were produced and no thorium was co-recovered [8]. Naturally, thorium is an issue in processing and waste streams and there is the potential to exceed the NRC's 0.05% criterion for source material.

3.4. Dose rates

Dose rates can range from near background levels to over 100 $\mu\text{Sv/h}$ for monazite wastes, which are not currently exploited for REEs in the United States of America. Exposure rates vary because many of the decay products may no longer be in secular equilibrium with uranium and thorium.

¹ Percentages by mass.

4. THORIUM

Reserves are well characterized, defined and economically proven deposits. The United States of America has the largest reserves in the world, although larger resources (unproven) occur elsewhere. Monazite was recovered as a by-product of processing heavy mineral sands for titanium and zirconium minerals in the past, and thorium was a by-product of refining monazite for its REE content. Domestic mine production of thorium bearing monazite ceased at the end of 1994. Furthermore, regulatory and disposal costs limited imports to 43 t of monazite with 3 t of ThO₂ in 2012 [9]. Nevertheless, the potential for future production still remains should a compelling new use of thorium emerge.

5. NIOBIUM

Significant US niobium mine production has not been reported since 1959. Domestic niobium resources (150 000 t) are of low grade, some are mineralogically complex, and most are not commercially recoverable. Consequently, all needs were met by imports. The United States of America imported 9200 t of niobium in 2012 and there were no known plans to mine niobium domestically [10].

6. OIL AND GAS

The oil and gas industry is well established in the United States of America. This industry has been well described in Ref. [11].

6.1. NORM accumulation and waste

NORM accumulates in oil and gas and associated brine. Accumulation locations include pumps, pipes, storage tanks and as radium (²²⁶Ra and ²²⁸Ra) scale in oil field tubulars, casings, separators and valves. Barite is introduced from either the seawater or injected drilling mud. Radium co-precipitates with silicates, sulphates, and/or carbonates in sludge and scale and becomes a larger problem as brine production increases and oil/gas production declines.

6.2. NORM concentrations and dose rates

Sludge concentrations are found up to 10 Bq/g ²²⁶Ra. Typically, ²²⁶Ra is about 78% of the total radium and the rest is ²²⁸Ra. Scale concentrations

appearing as thick, dense precipitates in tubing and separators, among other things, can exceed 100 Bq/g ^{226}Ra . Air kerma rates from pipe scale range up to 175 000 nGy/h. Drums packed with scale can exceed 130 000 nGy/h [12].

NORM waste ^{226}Ra concentrations can reach high levels but are more typically 5–6 Bq/g. The petroleum industry generates hundreds of thousands of tonnes of NORM contaminated waste each year.

6.3. Radon and radon progeny

Radon emanation from scale and sludge is 0.1–0.2%. Radon presents another problem as it moves through tubing and production equipment with methane and leaves thin deposits of ^{210}Pb over time. Radon is dissolved in brines and the organic petroleum fractions in gas plants and partitions mainly in propane and ethane fractions in surface equipment.

Radon progeny deposition occurs in surface equipment at gas plants and compressor stations at very low mass compared to oil production. An invisible layer of ^{210}Pb and progeny is left on pipes, valves, and pumps, which can be of the order of thousands of becquerels per 100 cm².

7. ZIRCON AND ZIRCONIA

The zirconium silicate mineral zircon is produced as a co-product from the mining and processing of heavy minerals. The United States of America holds 14 million t of the 60 million t of zircon resources recorded worldwide [13]. Two firms produced zircon in 2012 from surface mining operations in Florida and Virginia (a new mine in Georgia was recently granted a permit). Production information is withheld by the United States Geological Survey to protect the interests of the two production companies. Zirconium metal and hafnium metal were produced from zirconium chemical intermediates by two domestic producers — one in Oregon and the other in Utah. Once again, this industry has already been described in detail in Safety Reports Series No. 51, Radiation Protection and NORM Residue Management in the Zircon and Zirconia Industries [14].

8. TITANIUM DIOXIDE

Production in the United States of America was performed by four companies at six facilities in five states. They combined to produce 1.3 million t of titanium dioxide. Additionally, 216 000 t were imported and only 674 000 t of the domestic production was exported [15]. Titanium dioxide is also the subject

of a report in Safety Reports Series No. 76, Radiation Protection and NORM Residue Management in the Titanium Dioxide and Related Industries [16].

9. COAL COMBUSTION

Coal is a significant energy source in the United States of America, with 17 434 quadrillion BTU generated in 2012 [17]. NORM concentrations are variable but generally lower in Appalachian coal. NORM accumulates in coal ash, which is classified as fly ash, bottom ash and boiler slag. Total coal ash production is tens of millions of tonnes per year. Coal ash contains up to 0.75 Bq/g ^{226}Ra with an average of less than 0.15 Bq/g. Fly ash is used in concrete production, but at a small fraction of the overall concrete mass.

10. WATER TREATMENT

Radionuclide concentrations in groundwater are highly variable. Treatment sludge contains accumulated and concentrated radium and uranium in pore spaces of charcoal beds and admixed with spent ion exchange resins. The waste generated each year is hundreds of thousands of tonnes, in which this NORM is accumulated. In addition, radon gas is also carried with the water supply into homes.

11. PHOSPHATE INDUSTRY

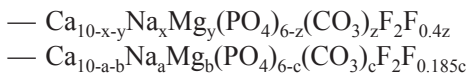
Safety Reports Series No. 78, Radiation Protection and Management of NORM Residues in the Phosphate Industry [18], is the most recent publication in the Safety Reports Series on industrial activities involving NORM. The production figures in this paper represent updates to that publication for the United States of America. Phosphate rock production in the United States of America in 2011 was 27 619 300 t with no exports [19]. This was 14.46% of world production. The United States of America also imported some beneficiated phosphate rock: 2 411 000 t from Morocco, 860 600 t from Peru and 400 t from China. Beneficiated phosphate rock is shown in Fig. 1.



FIG. 1. Beneficiated phosphate rock.

11.1. Sedimentary phosphate rock

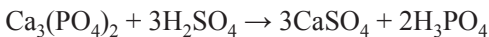
Most of the US rock was mined in Florida. It is a common sedimentary apatite known as francolite (carbonate–fluorapatite). Francolites have been described in literature in two similar ways:



Primary substitutions are from the uranium series followed by the thorium series to a much lesser extent.

11.2. Process chemistry

The process is represented by the formula:



At the chemical processing plant, beneficiated, concentrated phosphate rock (or high quality pebble) is reacted as slurry with sulphuric acid to produce phosphoric acid and calcium sulphate. The calcium sulphate may be in the hemi or dihydrate form.

Calcium sulphate is gypsum, and gypsum produced as a co-product with phosphoric acid is known as phosphogypsum (see Figs 2 and 3), which is stacked on land. The phosphoric acid is mainly used to make dry, ammoniated fertilizers. It has been known for decades that most of the uranium goes with the phosphoric



FIG. 2. Phosphoric acid production facility and phosphogypsum stacks.



FIG. 3. Process water retained in a phosphogypsum stack.

acid as soluble salts, and most of the radium goes with the phosphogypsum (primarily as radium sulphate).

11.3. Production of processed phosphates

Companies operating in the United States of America produce phosphoric acid (commonly called phos acid) and dry ammoniated fertilizers: monoammonium phosphate (MAP) and diammonium phosphate (DAP), for distribution both domestically and worldwide (see Table 1) [20]. Currently, there is no production of triple superphosphate (TSP) in the United States of America.

Worldwide production and distribution of the primary industry products is shown in Table 2 [20]. By comparing Tables 1 and 2, it is evident that the United States of America provides about 25% of the world's MAP and DAP fertilizers.

TABLE 1. PROCESSED PHOSPHATES IN THE UNITED STATES OF AMERICA, 2011

	US production (kt P ₂ O ₅)			
	Phos acid	MAP	DAP	TSP
Production	8423.6	2524.9	3050.7	0
Home deliveries		1471.9	1266.6	0
Exports	492.3	968.1	1694.2	0
Total deliveries		2440	2960.8	0

TABLE 2. PROCESSED PHOSPHATES WORLDWIDE, 2011

	World production (kt P ₂ O ₅)			
	Phos acid	MAP	DAP	TSP
Production	42 055.7	12 541.4	15 316	3 076
Domestic deliveries		8 778.8	8 453.8	1 179
Exports	4 405.4	3 639.7	6 604.8	1 806
Total deliveries		12 418.5	15 058.6	2 985

OVERVIEW OF INDUSTRIAL NORM IN THE UNITED STATES OF AMERICA

The United States of America imports TSP averaging 46% phosphorous pentoxide (P_2O_5), mostly from Israel, Morocco, Mexico, Tunisia and China. In 2011, 110.7 kt of TSP were imported.

Although phosphoric acid is primarily used to make dry, granulated fertilizer products for shipment, there is also a smaller trade of phosphoric acid (see Table 3). The United States of America has 21% of the world's production capacity [21] and has 11.2% of the world export market. Florida has 60.14% of US production capacity [21].

The United States of America accounts for 26.6% of the world export market for MAP (see Table 4) and 25.65% of the world export market for DAP (see Table 5) [20]. Florida has 66.8% of US production capacity of MAP and 66.8% of US production capacity for DAP [22].

TABLE 3. PHOSPHORIC ACID IMPORTS AND EXPORTS, 2011

	Imports (kt P_2O_5)	Exports (kt P_2O_5)
China	16	
Mexico	14.3	58.6
India		311.9
Canada		61.4
Brazil		58.8
Portugal		0.2
All others		1.4
Total	30.3	492.3

TABLE 4. EXPORTS OF MONOAMMONIUM PHOSPHATE FROM THE UNITED STATES OF AMERICA, 2011

	Amount exported (t)
Canada	290.6
Argentina	99.3
Brazil	266.9
Colombia	77.4
Mexico	23.7
Indonesia	52.8
Australia	122.2
All others	35.2
Total	968.1

Note: Average P₂O₅ content is 52%.

TABLE 5. EXPORTS OF DIAMMONIUM PHOSPHATE FROM THE UNITED STATES OF AMERICA, 2011

	Amount exported (t)
India	968.9
Japan	77.8
South Africa	20.1
Ecuador	27.4
Guatemala	24.2
Honduras	26.1
Paraguay	33.7
All others	516
Total	1694.2

Note: Average P₂O₅ content is 46%.

So what is the point of all these production numbers? They show that a lot of NORM is traversing the globe in phosphoric acid and ammoniated fertilizers. The United States of America alone loses over 1 million kg of U_3O_8 per year to farm fields: about half domestically and half abroad. This uranium represents an opportunity lost. This valuable energy source is dispersed on farm fields and lost to the energy sector. The environmental and public health consequences of uranium deposition on farm fields are debatable, but the loss of uranium as an energy source is undeniable.

Alternatively, phosphogypsum represents an opportunity being discovered and realized worldwide. Phosphogypsum has many uses that have been proven to be safe and beneficial. However, the NORM and metals content of phosphogypsum depend on the source of rock used and the processing conditions under which it formed. Consequently, care needs to be taken to match a given source of phosphogypsum to the appropriate end use so that exposures and risks can be managed. Most of the US phosphogypsum supply is represented by the 'stacks' in Florida. In fact, over 1.3 billion t of phosphogypsum are stacked there. Little use is permitted by the EPA because phosphogypsum made using central Florida 'Bone Valley' rock averages nearly 1 Bq/g ^{226}Ra and storage in stacks is mandatory. This stacked phosphogypsum represents an extremely large amount of material with low concentrations of NORM that should be used constructively rather than regarded as 'waste' left to future generations to resolve.

11.4. Phosphogypsum is a 'green' co-product of phosphoric acid production

Phosphogypsum has been called a waste or even a toxic waste, and it does contain trace amounts of heavy metals and radionuclides of natural origin. However, the concentrations of these constituents are very low in even the most extreme cases, and all phosphogypsum, from wherever sourced, has a safe and beneficial application.

Phosphogypsum contains naturally occurring trace elements from the rock source, such as arsenic, cadmium, chromium, cobalt, copper, lead, mercury, radium, selenium, uranium and zinc. However, these same elements are all found naturally in soils throughout the world in very similar amounts. Consequently, phosphogypsum is typically no more 'toxic' than soil and in fact may be classified as a soil. Depending upon the source rock, some phosphogypsum may contain slightly elevated concentrations of cadmium, radium and selenium. In toxicological terms, however, such concentrations are very low. The material does not merit being defined as hazardous with the concomitant effect of making it a waste.

11.4.1. Phosphogypsum characteristics

The physical/chemical characteristics (pH, density and P_2O_5 content, among other things) help to determine the usefulness of a particular source of phosphogypsum for a particular application. For example, the P_2O_5 content is important when considering use as a cement retarder. Other characteristics determine whether the phosphogypsum is appropriate for a use in which the public or environment may be exposed to its constituents via one or more pathways. These constituents can be broadly lumped into categories of metals and radioactivity.

- Metals: arsenic, cadmium, chromium, cobalt, copper, lead, mercury, radium, selenium, uranium and zinc (others to a lesser extent such as molybdenum and nickel);
- Radioactivity: radium, radon (specifically the fraction released from materials), uranium, other daughters of the ^{238}U series (^{232}Th series and ^{235}U series to a much lesser extent) and direct gamma radiation exposure from the radioactivity in source materials.

11.4.2. Pathways

Pathways are the routes taken by radionuclides or compounds of concern from source to human exposure or incorporation. Several inhalation, ingestion and environmental pathways are relevant to the uses of phosphogypsum:

- (a) Inhalation. The only constituent of significant concern is ^{222}Rn (the immediate progeny of ^{226}Ra decay). Inhalation scenarios are typically:
 - Phosphogypsum incorporated into construction products such as wallboard → home application → radon emanation into residence or workplace → buildup of radon progeny → inhalation of radon progeny;
 - Phosphogypsum stacked or in non-residential use → radon emanation into ambient air → inhalation of radon progeny (EPA determined that this is low dose/low risk);
 - Phosphogypsum stacked or in non-residential use → subsequent construction on abandoned stack or site of phosphogypsum use → radon emanation into residence or workplace → buildup of radon progeny → inhalation of radon progeny.
- (b) Ingestion. Heavy metals and metalloids such as arsenic are of concern but vary in significance by element. Uranium is more of a hazard as an ingested metal (kidney damage) than as a radionuclide. Ingestion scenarios are typically:

- Fertilizer or soil amendment → soil → crop or forage vegetation (uptake or deposition) → livestock or direct human ingestion;
 - Fertilizer or soil amendment → soil → groundwater or surface water body (runoff) → livestock or direct human ingestion;
 - Fertilizer or soil amendment → soil → direct human ingestion (scenario of a child eating dirt/soil).
- (c) Environmental:
- Fertilizer or soil amendment → soil → groundwater or surface water body (runoff) → environmental food chain;
 - Phosphogypsum stacked or in non-residential use → radon emanation into ambient air → atmospheric dispersion of dust and radon progeny → inhalation by local wildlife or ground deposition to environmental food chain (likely to be of little impact).

11.4.3. Potential uses of phosphogypsum

Any proposed use must be matched to the phosphogypsum properties, including trace elements. For example, the radium content of some phosphogypsum may prohibit its use in housing construction, but it would be acceptable as a road base, soil conditioner, fertilizer, daily landfill cover, or in construction of marine barriers. Phosphogypsum with higher levels of trace metals may be appropriate for construction applications.

- (a) Agricultural. Fertilizer, soil amendment to repair soils with too much sodium (near saltwater), repair magnesium affected soils and reduce water use in farming.
- (b) Construction. Road construction, cement, housing construction, marine barriers, sand replacement and embankments.
- (c) Landfill. To replace the daily landfill cover (usually soil) and enhance the breakdown of waste, which vastly improves the useful life (duration/volume) of the landfill.
- (d) Resource recovery. Sulphur recovery, calcium products, REEs and others. The residual uranium reports to the stack water, but at concentrations roughly 20 times less than phosphoric acid, making it much less attractive for recovery. Phosphogypsum in Florida contains about 79% of the REEs from the feed rock and extraction methods are currently being tested. If a suitable extraction method is found, the stacks become exploitable reserves. But again, producers are wary of the small amount of thorium that could become concentrated in the REE extraction process.

One note of caution must be mentioned. Since phosphogypsum has been designated as a waste material for so long, it has often been treated as such. Consequently, stacks have been used as dumps in some locations and may contain some areas contaminated with discarded objects, scale and sludge, together with the heavy metals or radioactivity they contain.

11.4.4. Matching sources to applications

In order to use phosphogypsum safely, it is prudent to match the phosphogypsum to a proposed use with a goal to limit exposure via target pathways. The choice of the appropriate application is based on the phosphogypsum characteristics and pathway considerations (note that there is **no combination** of metals and radioactivity that **excludes all uses**). The outcome can be summarized as follows and graphically in Fig. 4:

- (a) High concentrations of one or more of the critical metals — target ingestion pathway:
 - (i) Limit agricultural uses (graded approach to regulation);
 - (ii) Potential uses include construction applications, landfill, and resource recovery.
- (b) Radioactivity concentration $> 1 \text{ Bq/g } ^{226}\text{Ra}$ — target inhalation pathway:
 - (i) Limit residential construction uses (graded approach to regulation);
 - (ii) Potential uses include non-residential construction, agriculture, landfill, and resource recovery.
- (c) High concentrations of one or more of the critical metals and $> 1 \text{ Bq/g } ^{226}\text{Ra}$ — target ingestion and inhalation pathways:
 - (i) Limit residential construction and agricultural uses (graded approach to regulation);
 - (ii) Potential uses include non-residential construction, landfill, and resource recovery.

12. CONCLUSIONS

- (a) The United States of America currently operates or has operated all 12 types of facilities and activities identified in Safety Reports Series No. 49 [2].
- (b) Large quantities of NORM are continuously transferred both domestically and internationally in products and deposited locally as residues or unused by-products.
- (c) Utilization of phosphogypsum can convert an environmental burden into a profitable, safe and beneficial product.



FIG. 4. Uses of phosphogypsum.

- (d) ‘Comprehensive extraction’ — the phosphate industry can serve as a source of fertilizers, uranium oxide, and REOs with only a single disturbance to the environment.
- (e) ‘Triple bottom line return’ — comprehensive extraction of materials that are critical to both food security and energy security yields social, environmental, and economic dividends as a ‘win-win-win’ situation.

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DOSES FROM INDUSTRIAL PROCESSING OF NORM

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Abstract

Interest in the impacts of NORM in the processing of bulk raw materials from mining operations has grown considerably over the last 10 to 20 years, and much work has occurred at an international level to develop a consistent approach to the management and control of such materials. In this regard, the IAEA has established a framework for the control of exposure, based on the system of protection for planned exposure situations, which has been almost universally adopted in national regulations. The 'entry level' above which this framework becomes applicable is an activity concentration criterion of 1 Bq/g for radionuclides in the naturally occurring uranium and thorium decay chains. Dose assessments have shown that, even at activity concentrations significantly above this entry level of 1 Bq/g, the radiological risk from the material may be very small. However, the undertaking of dose assessments for such materials is complex and difficult and what usually happens in practice, therefore, is that the simpler, concentration based entry level of 1 Bq/g becomes the regulated control level. The paper provides the results of dose modelling for a number of scenarios and shows that such use of a simplified activity concentration level of 1 Bq/g is unnecessarily restrictive. The paper examines some practical aspects of the impact of the implementation of the blanket rule in relation to bulk raw materials containing NORM and proposes that more emphasis should be placed on the actual risks from a material, rather than using, as a default value, the entry level criterion of 1 Bq/g and that further education of all proponents needs to occur.

1. INTRODUCTION

Every year, large quantities of raw materials including metal concentrates are traded between States as feedstock for metal production facilities. Some materials contain elevated levels of radionuclides of natural origin and, when concentrations are high enough, controls are necessary to ensure human safety and environmental protection. Controls are manifested in the form of national legislation and regulations, and advice on such controls is provided at an international level by the IAEA.

This paper provides some practical perspectives on the application of the IAEA standards to raw materials containing low levels of radionuclides of natural origin and shows that the IAEA standards are generally complex, resulting in national regulatory systems being based on a simplified (and erroneous)

interpretation of the standards. When potential doses from exposures to such materials are calculated, it can be shown that this simplified interpretation of the standards results in the imposition of regulatory controls that are unnecessarily restrictive and conservative. The focus of this paper is on the potential doses arising from exposure to bulk quantities of metal concentrates containing natural uranium. However, the arguments contained herein are equally valid for bulk materials containing natural thorium.

It should be noted that, in this paper, reference is made in general terms to the IAEA activity concentration criterion of 1 Bq/g for application of the system of protection for planned exposure situations. In this regard, it is to be understood that this criterion applies specifically to each radionuclide in the ^{238}U and ^{232}Th decay chains.

2. OVERVIEW

Processing facilities such as metallurgical plants and smelters obtain their feed materials as metal concentrates directly from mining or beneficiation (concentration) facilities. Arrangements are established for the supply of metal concentrates that meet a certain specification in order to optimize the performance of operations and to minimize costs. When the materials do not meet the required specification, materials can be obtained from a number of suppliers and blended to the optimum levels. In some cases, blending is undertaken by third parties who buy, trade, blend and resell metal concentrates originating from various producers. There are also some cases where there are dedicated facilities that process a single metal concentrate. An important product specification consideration is the concentration of radionuclides of natural origin that may be present in a feed material. Generally, this is measured in units of radioactivity per unit mass (Bq/g) or mass concentrations of contained uranium or thorium (in mg/kg). Compared with other product contaminants, the presence of radionuclides in materials tends to carry a higher level of scrutiny and regulatory concern. In some cases, the presence of radionuclides, in whatever quantities, also attracts potentially unwarranted significant public concern and adverse media publicity. This is despite the fact that almost every substance contains naturally occurring quantities of radionuclides.

International standards for the safe use of materials containing radionuclides of natural origin have been established by the IAEA and apply to raw materials such as metal concentrates. The standards provide the basis for national authorities to establish regulatory systems for the control of the radiological impacts of such materials. Implicit in the standards is the determination of whether the material is regarded as radioactive and, if it is, such material and any

practices involving it would be subject to the requirements for planned exposure situations. Such material then becomes subject to some form of regulatory consideration, which can result in the granting of an exemption from control or the imposition of certain regulatory requirements to ensure that exposures are controlled. Depending on the level of risk, such requirements can range from a requirement to simply notify the appropriate regulatory body through to a requirement to apply for an authorization and implement a comprehensive radiation management programme. The IAEA standards provide guidance on what constitutes a material as being radioactive, this being, in effect, when the activity concentration of any radionuclide in the ^{238}U and ^{232}Th decay chains is such as to make it subject to the system of protection for planned exposure situations (i.e. when the activity concentration exceeds 1 Bq/g). The IAEA standards also provide for the assessment of radiological risk for materials above 1 Bq/g as a means of grading the regulatory response.

In practice, metal concentrates with a radionuclide activity concentration exceeding 1 Bq/g (and which are therefore defined as radioactive) are considered to be undesirable, with many processing facilities simply not accepting metal concentrates that exceed 1 Bq/g, regardless of whether there is any demonstrated radiological risk or not.

3. RADIOLOGICAL CONSIDERATIONS OF METAL CONCENTRATES

The majority of metal bearing ores contain low levels of naturally occurring uranium (and thorium). The beneficiation of the ore to produce a metal concentrate sometimes results in enhanced concentrations of radionuclides of natural origin. For undisturbed materials containing naturally occurring uranium, the radionuclides in the uranium decay chains usually exist in a state of equilibrium (i.e. the activity concentration of each radionuclide is approximately the same). This is known as secular equilibrium. Usually a metal concentrate produced from the beneficiation of the ore also contains the radionuclides in secular equilibrium. The concentration of the individual radionuclides in a material can be measured using standard analytical techniques. However, when a material has radionuclides in secular equilibrium, the radionuclide concentrations can be calculated instead from the grade of the uranium. This is done through the empirical relationship where 1 Bq/g of ^{238}U is equivalent to 81 mg/kg of uranium. When a material contains radionuclides which are out of equilibrium, care must be taken to consider the decay and ingrowth of other radionuclides in the decay chain. Individual radionuclides may need to be analysed to determine their concentration. A knowledge of the radionuclide composition of the material

is important when considering the IAEA standards and national regulations for radioactive materials and how they apply to the material.

4. THE IAEA STANDARDS

The system of protection for planned exposure situations, as embodied in the IAEA standards, is comprehensive but also complex. It is applicable to a broad range of radioactive materials, including materials containing radionuclides of natural origin (for purposes of regulation, a radioactive material is defined by the IAEA as that designated in national law or by a regulatory body as being subject to regulatory control because of its radioactivity). The IAEA defines NORM as radioactive material (irrespective of whether processed or not) that contains no significant amounts of radionuclides other than radionuclides of natural origin. In terms of IAEA Safety Standards Series No. GSR Part 3 (Interim), Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards¹ (BSS), material containing natural uranium would be regarded as radioactive material if any of the individual radionuclide concentrations exceeded 1 Bq/g — such material would be subject to some form of regulatory control, placing certain obligations on those responsible for the material. However, the BSS provide criteria for exemption from specific regulatory requirements, noting that such requirements would be warranted only when doses from exposure to the materials exceeded certain dose levels under various exposure situations. This is consistent with a risk based approach where control would depend upon the magnitude of the risk (which is estimated as potential dose). The basis for this approach is described in IAEA Safety Standards Series No. RS-G-1.7, Application of the Concepts of Exclusion, Exemption and Clearance [2], and the concepts contained therein have now been generally adopted.

The scope of regulation for materials containing radionuclides of natural origin can be summarized as follows:

- (a) Exposures are automatically **excluded** from the requirements of the BSS when they are ‘unamenable to control’ (this includes exposure to cosmic radiation, to natural levels of radon and to ⁴⁰K in the body). Any exposure to materials with individual ²³⁸U and ²³²Th decay series radionuclide activity concentrations of 1 Bq/g or less could be regarded as an excluded

¹ INTERNATIONAL ATOMIC ENERGY AGENCY, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, IAEA Safety Standards Series No. GSR Part 3 (Interim), IAEA, Vienna (2011). [Editor’s note] This has been superseded by Ref. [1].

exposure because, in terms of the BSS, such exposure would not be subject to regulatory control and the material would consequently not be regarded as radioactive.

- (b) Materials with radionuclide concentrations above 1 Bq/g may be **exempt** from regulatory requirements where it can be shown that the risks are so low as to not warrant regulatory control or that regulation does not provide any net benefit. Exemption can only be determined and granted by the appropriate regulatory body (and is thus a form of regulatory control).
- (c) Materials that are already in a regulated practice may be approved for **clearance** by the regulatory body when it can be shown that the impacts of releasing the material do not warrant regulatory control.

Even though a mechanism exists to exempt material with activity concentrations above the 1 Bq/g entry level for the system of protection for planned exposure situations, the simplest and clearest criterion is the entry level itself and, in practice, it is the easiest mechanism by which to regulate. As a result, 1 Bq/g has become the de facto 'limit' in many national regulatory systems. Even though the IAEA standards require a graded approach to regulation, with potential dose being the main driver of the radiation protection requirements, in practice the activity concentration criterion of 1 Bq/g tends to be the primary criterion for the imposition of control.

However, for a material that does contain radionuclides in concentrations above 1 Bq/g, the general IAEA framework outlined in the BSS and IAEA Safety Standards Series No. RS-G-1.7 [2] provides for those materials to be exempted from regulation when such regulation is not warranted. Within this framework, the regulatory body may decide to grant an exemption at activity concentrations many times higher than 1 Bq/g (e.g. up to 10 Bq/g), with decisions on exemption being based not on activity concentration but on a dose criterion of the order of 1 mSv per year (as determined for scenarios which represent typical exposure situations). Often, however, exemption mechanisms are not set out explicitly in national regulations, possibly because of:

- (a) The difficulty in conducting dose assessments, which may involve complex dose modelling;
- (b) The reluctance to grant exemption due to a precautionary approach, regulatory conservatism or public concern;
- (c) The criterion of 1 Bq/g being clear, easy and unambiguous as a cut-off value;
- (d) Once a material is labelled as radioactive, the difficulty in removing that label;

- (e) The ease of determining radionuclide concentration from either the product specification information sheets or from sampling, rather than carrying out complex dose modelling.

In the absence of a proper risk assessment, and in an over simplified regulatory framework, irrational situations arise. For example, a material with an activity concentration of 0.9 Bq/g would be considered to be automatically exempt, while a material with an activity concentration of 1.1 Bq/g would be defined as a radioactive material and therefore subject to regulatory consideration. In practice, a customer would more than likely reject the latter material rather than undergo the potential added regulatory burden, constraint and scrutiny.

5. POTENTIAL DOSES FROM 1 Bq/g MATERIAL

In order to understand the doses that might arise from metal concentrates containing 1 Bq/g of radionuclides of natural origin (in secular equilibrium), the following exposure scenarios were modelled:

- (a) A truck driver who transports material;
- (b) A sampler who takes a sample from each truck;
- (c) A front end loader operator who manages a stockpile of material;
- (d) A plant operator who works with smelter residues;
- (e) A member of the public living on buried smelting slag;
- (f) A member of the public exposed to fumes emitted from a smelting operation.

The dose assessment was performed using the RESRAD suite of radiological modelling software. The assumptions made and the results obtained are shown in Table 1.

The calculation parameters are conservative and show that at 1 Bq/g, occupational doses are low. The exposure scenarios giving rise to the highest dose are those for the loader and truck operators. It would be expected that doses would be lower using more realistic exposure parameters. With regard to the public, using the most conservative assumptions, the calculated doses are also low. As a result of this preliminary assessment, it can be concluded that actual doses from a metal concentrate with an activity concentration of 1 Bq/g would be low, and therefore the constraints imposed by the material being subject to regulatory control simply because it falls within the definition of radioactive material (NORM), are unjustified.

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TABLE 1. DOSE CALCULATIONS FOR MATERIAL WITH AN ACTIVITY CONCENTRATION OF 1 BQ/G: ASSUMPTIONS AND RESULTS

Scenario	Parameter	Value or assumption	Dose (mSv/a)
Truck driver	Truck load	20 t	0.18
	Exposure period	2 000 h/a	
	Protection	No shielding from truck body, air-conditioned cab	
Sampler	No. of samples	3 000 per year	0.01
	Exposure period	1 min per sample	
	Dust concentration	1 mg/m ³	
Front end loader operator	Stockpile size	6 000 t	0.62
	Exposure period	2 000 h/a	
	Location	Adjacent to stockpile	
	Protection	No shielding from loader body	
	Dust concentration	1 mg/m ³	
Plant operator	Exposure period	2 000 h/a	0.04
	Dust concentration	1 mg/m ³	
	Residue stockpile size	210 t	
	Location	Adjacent to stockpile	
Residence on smelter slag	Cover thickness	1 m	0.001 3
	Slag thickness	1 m	
	Consumption	All food/water from immediate area	
Smelter emissions	Smelting rate	60 000 t/a	0.034
	Period of operation	20 a	
	Mixing depth	10 cm	
	Consumption	All food/water from immediate area	

Safety Reports Series No. 49, Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials [3], provides indicative relationships between NORM activity and the resulting dose to a worker. The values for mineral concentrates such as zircon sand and sand containing monazite are about 0.4 µSv/h per unit NORM activity concentration (in Bq/g) for gamma exposure at 1 m and about 37 µSv/h per unit activity intake (in Bq) for inhalation

of airborne dust (the values for other minerals are in the ranges 0.004–0.82 $\mu\text{Sv/h}$ per Bq/g for gamma exposure and 1–37 $\mu\text{Sv/h}$ per Bq for dust inhalation). Care must be exercised when calculating doses from these factors. For occupational exposure, the gamma dose dominates. Applying a value of 0.4 $\mu\text{Sv/h}$ per Bq/g to a full year of exposure (2500 h) would give an annual dose estimate of 1 mSv/a, which is the level at which regulatory control might become necessary. However, it is unrealistic to assume that the worker would remain within 1 m of the source for a full year, since this rarely happens in practice (the dose assessment in Safety Reports Series No. 49 [3] assumes, more realistically, an exposure period of 400 h/a, giving a dose of 0.16 mSv/a).

6. WHAT DOES 1 Bq/g MEAN TO A PRODUCER?

For a metal concentrate at a radionuclide activity concentration of 1 Bq/g, the radioelement concentration levels corresponding to the main long lived radionuclides are shown in Table 2.

TABLE 2. RADIOELEMENT CONCENTRATIONS CORRESPONDING TO RADIONUCLIDE CONCENTRATIONS OF 1 Bq/g

Radionuclide at 1 Bq/g activity concentration	Corresponding radioelement concentration (mg/kg)
U-238	80
U-234	4.1×10^{-3}
Th-230	1.5×10^{-3}
Ra-226	3×10^{-5}
Po-210	7×10^{-9}
Pb-210	4×10^{-7}

To achieve these levels is technologically demanding and requires either low concentrations of radionuclides to start with or selective leaching or extraction. The supplier of such material is left in a conundrum and in a significantly disadvantaged position, with limited options such as the following:

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- (a) Develop processes that will selectively remove or reduce radionuclides to below the 1 Bq/g concentration level:
 - (i) These processes exist at a processing plant scale and result in significant added production costs and are complex, carrying their own risks.
 - (ii) The added costs may make projects unviable (e.g. it might involve a capital expenditure of US \$100 million to reduce the activity concentration from 2 Bq/g to 1 Bq/g).
- (b) Collaborate with the customer to seek exemption for the material:
 - (i) The intent of the IAEA approach can be explored by quantifying the risk of the material through dose and impact modelling and presenting the case to the regulatory body.
 - (ii) Any conditions on using the product may be prohibitive for the customer.
- (c) Negotiate sale terms with the customer:
 - (i) Undertake contractual discussions with the customer regarding 'out of specification' penalties.
 - (ii) Identify ownership of the material and who has responsibility for any approvals to import or export materials.

Project costing must take into account the potential options described above and if an outcome cannot be negotiated or a cost effective technical solution cannot be developed, then the project may be uneconomic. A potential alternative option is blending to obtain a total product mix that is less than 1 Bq/g. Regardless of the options, in a competitive industry where profit (and therefore viability and sustainability) is determined by marginal gains and losses, material containing radionuclides above 1 Bq/g is at a severe disadvantage. This is particularly frustrating when the actual radiological risk from the material may not warrant the restrictions.

7. ICRP CONSIDERATIONS

The International Commission on Radiological Protection (ICRP) philosophy is that radiation should be managed primarily on its risk rather than by limits. From a radiation protection perspective, the unavoidable question is whether the constraints placed on material with naturally occurring radionuclide concentrations above 1 Bq/g are justified, particularly in the absence of a proper dose assessment. The optimization principle of the ICRP would clearly direct that the magnitude of the radiological impacts and risk be assessed before controls are implemented. However, the practical implementation of the IAEA framework

has resulted in a situation where ‘limitation’ has taken precedence, even though it may not be justified in all cases. Without consideration of the actual impacts or risk of materials, the approach to radiation protection collapses from an ALARA — as low as reasonably achievable — (optimization) approach to one of mere compliance. To add to this problem, there are other unintended consequences of the 1 Bq/g ‘limit’ including:

- (a) It further reinforces the contention that even low levels of radioactivity are dangerous;
- (b) It reinforces the idea that there is a safe/unsafe level of radioactivity.

8. CONCLUSION

The IAEA framework on radiation protection of materials containing low levels of naturally occurring radioactivity is complex. In practice, this leads to the implementation of an over simplified version of the framework. While the intent of the IAEA framework is to have a graded approach to regulation of such materials, in practice most regulatory systems are based, erroneously, on the simple activity concentration level of 1 Bq/g, which results in the material being defined as ‘radioactive’ regardless of the actual risk of the material. This implementation is inconsistent with the philosophy of the ICRP and the fundamental thinking behind radiation protection, in which the level of control should be commensurate with the actual risk.

The author suggests that more emphasis be given in national regulations on providing proponents the ability to seek exemption upon demonstration of the safety of a material. This needs to occur in a manner that avoids perceptions of the material (and the risk) overriding proper decision making. To achieve this requires additional education and information on the practical processes to achieve exemption. There is a responsibility on producers and customers of raw materials to develop competencies and properly understand their products, including understanding the presence and fate of NORM radionuclides. There is also a responsibility on legislators and regulators to have mechanisms to ensure that the actual risk of a material is properly considered before restrictions or constraints are imposed.

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Invited Paper

COMPREHENSIVE EXTRACTION: A KEY REQUIREMENT FOR SOCIAL LICENSING OF NORM INDUSTRIES?

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Abstract

The paper explores the potential contribution of the policy and practice of comprehensive extraction to the achievement and maintenance of social licensing for NORM industries. Given the increasing difficulty worldwide of gaining stakeholder acceptance of mining projects of any nature, the concept of disturbing the ground only once and extracting all resources of value for current or future use is highly attractive. This approach, and the underpinning assumptions of sustainable development on which it rests, was (re)introduced by Pingru Zhong at an IAEA Technical Meeting in September 2011 and has rapidly gained acceptance. The initial focus has been on the feasibility of re-engineering flowsheets for managing and processing phosphate related resources such as phosphorus, uranium, rare earth elements and thorium in a comprehensive manner. But the recent recognition that, by virtue of geological processes as yet unexplained, oil and gas deposits are often found co-located with phosphate resources, suggests an even more radical rethink around the concept of an ‘energy basin’, managed as a single complex entity, may be the true implication of the comprehensive extraction method. At a minimum, it seems to satisfy contemporary requirements for measuring triple bottom line (economic, social and environmental) indicators of performance and outcome, so achieving a new equilibrium between stockholder (owner–investor) and stakeholder (engaged or affected party) interests. But the appeal of the concept is also cultural and contextual in the sense defined by Brundtland in *Our Common Future* (1987). It invites those exploring, reporting and extracting such resources to think of optimizing the entire value of the resource to a given group of beneficiaries at a given time, across a complete life cycle, rather than draining single commodities for a one-off return. This is likely to have significant potential for coupling the social licensing process with a commitment to comprehensive extraction.

1. INTRODUCTION

The globalization of trade has not yet been matched by the globalization of related regulations and standards. The outcome is incoherence and inconsistency in both specific regulatory but also more general taxonomic and definitional terms:

“Despite the widespread occurrence of NORM, and notwithstanding the development of guidance material in some countries and by international authorities, there is no systematic international approach to regulating NORM in commodities and products, or for the management of NORM wastes”[1].

When viewed across different markets, the so called NORM industries — which the IAEA categorizes into a list of 12 [2] and the European Commission categorizes into a list of 16 (annex V of Ref. [3]) — reflect this unsatisfactory situation from a regulatory point of view, with approaches as different as (a) regarding NORM residues as toxic wastes; (b) regarding NORM residues as valuable resources to be protected under law; and (c) having no position in law on NORM residues at all. As well as the obvious case for bringing such regulatory approaches into some kind of constructive alignment, there is a deeper underlying issue addressed in this paper: namely, the imposition of an unnecessary economic, social and environmental penalty on the use NORM that flows from a failure to anchor NORM regulations in evidence based science.

1.1. Comprehensive extraction

The term ‘comprehensive extraction’ was (re)introduced by Pingru Zhong (China) during an IAEA Technical Meeting, 26–30 September 2011, on uranium extraction from phosphate rock (UxP).¹ The concept it described was received so favourably that the term was brought into currency during the follow on UxP Training Workshop in Marrakesh, 31 October to 5 November 2011. It has now been widely adopted, with the following operational objectives:

- (a) Build and sustain human resource capability by education and training;
- (b) Disturb the ground only once during mining and extraction, optimizing returns from all the resources in an ore body, not just a single target mineral;
- (c) Foster flowsheet modifications, and innovative and, if necessary, disruptive technologies and business to achieve triple bottom line returns;

¹ See <http://www.iaea.org/OurWork/ST/NE/NEFW/News/2011/repository/New-Comprehensive-Approaches-to-Uranium-Mining-and-Extraction.html>.

- (d) Leave zero waste at the end of the project life cycle, thus eliminating long term negative externalities;
- (e) Base any mineral resource project life cycle plan on finding the ‘new point of equilibrium’ between the interests of stockholders and concerned parties, expressed in the form of a social licence and measured in financial, social and environmental returns;
- (f) Future proof mineral resources through proactive life cycle management, including recovery and recycling, as a key sustainable development outcome.

1.2. Definitional and taxonomic uncertainty

Perhaps because of the focus on the ‘radioactive material’ aspect of the NORM acronym, the equally significant designation ‘naturally occurring’ is overlooked, or seen as of secondary significance; and this definitional imbalance in and of itself gives rise to the incoherence in how NORM is understood and managed. As a result, when an industrial process is applied to NORM there is an immediate inference that the radioactive aspect must have been put there by the process; that is, the radioactivity is somehow caused or enhanced by man. This misperception allows the regulator to think of managing exposure to NORM even though in most situations this exposure is not amenable to control.

Natural radioactivity is widely distributed across the Earth, giving rise to a natural radiation background that varies by approximately two orders of magnitude. Levels may be even higher in certain localized mineral deposits. In consequence, all living species are exposed to radiation from natural sources. The ubiquitous occurrence of natural radioactivity means that many of the ores and minerals (such as coal, oil and gas, iron ore, bauxite and phosphate rock), commodities (such as water, building materials and fertilizer), products (such as ceramics) and devices (such as welding rods, gas mantles and electronic components) are likely to contain enhanced levels of radionuclides of natural origin, and even depend for their functionality on this radionuclide content.

The origins of natural radioactivity in mineral deposits such as phosphates and potash are in living organisms in the oceans. These organisms decompose, depositing the minerals contained in them on the sea bed; over millions of years this leads to the formation of sedimentary rock — rock that is now used for fertilizer manufacture. The relative radioactivity of the rock is the same as, or slightly higher than, the radioactivity levels of the world’s soils.

Activities as varied as mineral processing, coal burning and water treatment can modify the NORM concentrations in the products, by-products,

residues and waste streams they generate, giving rise to the concept TENORM² (technologically enhanced, naturally occurring radioactive materials) where the NORM content is held to be ‘technologically enhanced’ [4]. While discouraged by the IAEA, the term is still used in scientific publications, reflecting further levels of definitional uncertainty. In some NORM industry processes, specific radionuclides can separate out from the original radionuclide mixture, as for example the separation of radium and uranium during the processing of phosphate ore to produce fertilizer, which includes the majority of uranium, and phosphogypsum which retains the most of the radium.

As pointed out in Ref. [5], only 2 of the 12 IAEA listed NORM industries directly target NORM as a product. For the other ten industries, NORM is incidental. One, the phosphate industry, has produced triuranium octaoxide in the past and may do so again [6], and this same industry has the ability to not only supply its own products, but also the products of two other important industries on the list, uranium and rare earth elements, a fact some States are using to revisit the underlying business models on which particular NORM industries have been built. This is giving rise to the ‘whole energy basin’ approach to mineral management in which resources that are commonly co-located from a geological point of view such as coal, oil, gas, phosphates, uranium and rare earths are now being examined from an integrated rather than single target mineral point of view [7]. Such a holistic approach is naturally in harmony with the objectives of sustainable development.

2. SUSTAINABLE DEVELOPMENT

The classical definition of sustainable development was introduced by Brundtland in 1987 [8]: “Sustainable development is development that meets the needs of the present without compromising the ability of future generations to meet their own needs”. It contains within it two key concepts:

- (1) The concept of needs, in particular the essential needs of the world’s poor, to which overriding priority should be given;
- (2) The idea of limitations imposed by the state of technology and social organization on the environment’s ability to meet present and future needs.

As the concept is intergenerational in nature, it is intrinsically supportive of a new equilibrium between stockholder (ownership–investor) interests and

² See, for example, <http://www.epa.gov/rpdweb00/tenorm>.

stakeholder (engaged or affected party) interests. As such it aligns with, and probably derives from, the Nash cooperative game equilibrium model, which requires both parties in a game to ‘win’ rather than there to be a winner and a loser [9].

How might this apply to a NORM industry such as phosphates? Let us assume Stockholder 1 wants to focus value generation ‘upstream’ and mine and sell as much virgin phosphate ore as possible either as rock, acid or fertilizer product. Stakeholder 2 by contrast wants to achieve value ‘downstream’ by recovering and using as much phosphate as possible from residues and waste streams.

In a classical market (winners and losers) situation, Plan A for the phosphate life cycle, built on Stockholder 1’s interest to maximize production if managed in isolation is structurally in conflict with Plan B, based on Stakeholder 2’s preferences. Worse, if the two models compete, the moral hazard risk is that both profit from increasing their respective waste streams. If, however, the objective is resource conservation and efficient use of phosphate resources throughout the product life cycle, then the solution is vertical integration from the ‘virgin’ mine to the secondary ‘urban’ mine. When business performance is judged that way, a new point of equilibrium emerges which incents both parties to work to a common solution, as shown in schematic form in Table 1.

The application of Nash’s model demonstrates that only by combining their strategies (both follow Plan A) do both stockholder and concerned parties maximize their chances of achieving the best (i.e. most sustainable) rewards. In other words, a selfish strategy does not work for either party; the optimum solution is when all concerned parties follow the same plan; and no individual stakeholder maximizes his return by focusing solely on his own plan.

TABLE 1. A SIMPLE COORDINATION GAME SHOWING RELATIVE RETURNS FOR STOCKHOLDER 1/STAKEHOLDER 2 ACCORDING TO THE COMBINATION OF THEIR STRATEGIES

	Relative returns A/B	
	Stakeholder 2 adopts Plan A	Stakeholder 2 adopts Plan B
Stockholder 1 adopts Plan A	4/4	1/3
Stockholder 1 adopts Plan B	3/1	2/2

To make a negotiated, cooperative solution realistic, the critical dependency is that the returns to either or both concerned parties can be measured and verified to their satisfaction. And hitherto, this has been a key sticking point for any reasoned approach to building consensus as to optimum strategies for all concerned parties in the managed NORM resource life cycle.

The translation of sustainable development theory into business practice was initiated by Elkington in 1994 [10] in direct response to Brundtland [8] but also Nash [9]. Elkington proposed that a successful, sustainable enterprise needed to satisfy three equally significant performance measures, no one of which could be pursued to the disadvantage of the other two. These are financial, social and environmental returns, the so called ‘triple bottom line’ (TBL). This theory derives directly from Nash’s cooperative game model and has become increasingly influential in both private and public sector performance analysis, flowing naturally into the theory and practice of the social licence.

The operational consequence of applying the TBL approach is a new socioeconomic model built around what Villas-Boas has called the ‘axis of hope’. Instead of the market being driven by the traditional forces of fear and greed, Villas-Boas proposes that there must be hope at the centre, hope in a practical and pragmatic sense that a project which attracts a social licence will leave the society that grants the licence in a better state at the end that it was in at the beginning. This model is shown in Fig. 1. In such a model, the fundamental requirement of Brundtland that “the essential needs of the world’s poor” be given overriding priority is met because the focal points of meeting those needs are food security and energy security.

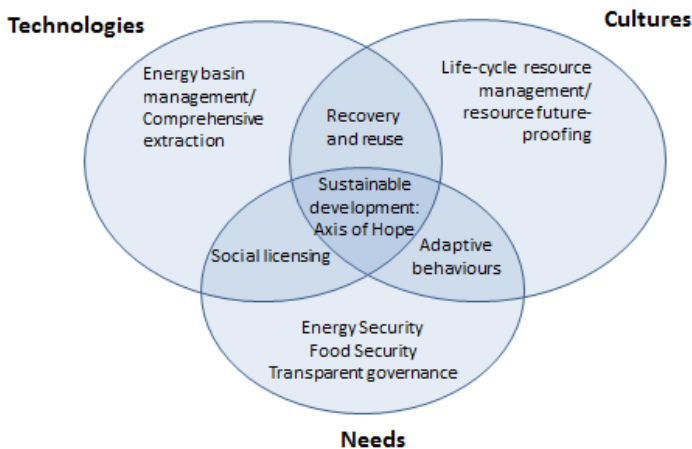


FIG. 1. The axis of hope as the core value of triple bottom line sustainable development.

3. THE SOCIAL LICENCE

A social licence has first to be won, and then maintained throughout the life cycle of a project. It is not like a driving licence or permit, which is granted on a single, static set of competencies. It is a continuous negotiation between stockholder and stakeholder, value based, and dynamic in its execution. Inevitably the measures by which the licence will be judged over time will change as the sustainable development process unfolds but these must be equitable for both stockholders and concerned parties.

At the level of an individual project, the social licence is rooted in the beliefs, perceptions and opinions held by any population local to that project but also held by other concerned parties about the project. These concerned parties may exist outside the formal jurisdiction of the project's actual location. For example, the world's phosphate resources are distributed highly unevenly around the Earth; we are all concerned parties in how these resources are managed whether we live in one of the countries holding large resources, such as Morocco or the United States of America, or one that consumes very large quantities, such as India. A social licence is granted by one or sometimes many communities and may vary in nature from community to community according to the need and capability of each. It is part tangible, part intangible, and as much effort must be made to measure the beliefs, opinions and perceptions that underpin the licence as is made for measuring TBL returns. Because of the subtleties of the interactions between tangible and intangible measures of return, its immediate outcomes are dynamic and progressive. But if there is no fundamental agreement on values that do not shift through time, the licence will be unsustainable.

3.1. Social licensing: Values

The three primary values, or intangible boundary conditions of social licensing may be described as social legitimacy, credibility and trust.

3.1.1. Social legitimacy

Social legitimacy is based on established norms, the norms of the community, that may be legal, social and cultural and both formal and informal in nature. Companies must know and understand the norms of the community and be able to work with them as they represent the local 'rules of the game'. Failure to do so risks rejection. In practice, the initial basis for social legitimacy comes from engagement with all members of the community and providing information on the project, the company and what may happen in the future and then answering any and all questions.

3.1.2. *Credibility*

The capacity to be credible is largely created by consistently providing true and clear information and by complying with any and all commitments made to the community. Credibility is often best established and maintained through the application of formal agreements where the rules, roles and responsibilities of the company and the community are negotiated, defined and consolidated. Such a framework helps manage expectations and reduces the risk of losing credibility by being perceived as in breach of promises made, a situation common where relationships have not been properly defined. A good practice for both stockholders and concerned parties is to avoid making oral commitments. In the absence of a permanent written record, oral statements may be open to question or reinterpretation at a later date.

3.1.3. *Trust*

Trust takes both time and effort to create, and can be lost in an instant. True trust comes from shared experience and commonly valued outcomes. The challenge for the stockholder is to go beyond a merely transactional model of agreement with the stakeholder to create a deeper sense of shared challenge and opportunity leading to mutual benefit. Concerned parties are much more likely to be realistic in their expectations of stockholders if they sense that the stockholders are realistic and are transparent in their sharing of risk and reward.

3.2. **The social licence life cycle**

If the social licence rests on a fundamental alignment of values between stockholder and stakeholder, there is also a life cycle to the social licence, which over times achieves equity of interest and benefit between both parties. This life cycle consists of three main stages:

- (1) Enabling investment;
- (2) Meeting local and cultural needs;
- (3) Mutually beneficial outcomes.

3.2.1. *Investment*

Investment concerns the mobilization of resources, in the form of both human and financial capital. The human capital investment is channelled through capacity building, which may consist of various types of training, ranging from deep, strategic investment in the educational infrastructure and system of a

community, or region, to competency based training for specific jobs. In both cases there must be a durable outcome.

If the capacity building is well conceived and well executed, its performance measures are relatively straightforward; for example, if the premise is one of partnership, over time there will be visible technology transfer in the form of plant, infrastructure and expertise, but also technology spill over, in that related or even unrelated sectors of the economy will start to show benefit from the capacity building process. In financial terms, the investment must be adequate to achieve the task in hand, based on a transparent, well rehearsed plan. One key component of the plan is the demonstration of mutual benefit between the investment in human capital and the physical infrastructure the project requires. Health, safety and environment measures apply in equal measure as process performance indicators and as outcome measures, but the balance between process and outcome is commonly confused.

One common point of confusion is between outputs and outcomes. Outputs such as tonnes mined or processed, or accident free hours or days worked are crucial to any project; but if the outcome is that the product cannot be sold profitably, or if the accident free labour is unproductive and inefficient, then the equilibrium between the measures of return is lost and the project is obviously unsustainable. A good practice is a balanced interest between lead and lag performance indicators, whether in terms of productivity or health and safety, a balance which always is reflected in an ongoing commitment to training and quality improvement.

3.2.2. *Meeting local and cultural needs*

If the investment objectives are clearly articulated and suitably funded, the result will be tangible local benefits, short and longer term, measurable in financial, social and environmental terms. These may affect individuals, families or the community as a whole, for example by building social capital and infrastructure, such as schools, hospitals and healthcare, roads, and enhanced communications. While such benefits may win initial support for the social licence, sustained support will depend on a fuller degree of cultural integration, leading to stakeholder acceptance and buy-in. As major projects will be of national not merely regional or local significance, success will also depend in the long term on alignment with national and local sustainable development needs and policy. This may be a particular challenge for smaller, emerging economies where the level of skill and experience in the team negotiation with the investors may not be equivalent to what the investor can afford to field. In such situations, the international community may need to assist in finding the point of equilibrium. External advisors may also be required for certain aspects of regulatory and

legal affairs, negotiations of commercial terms and risk management. Bilateral communications between concerned parties and stockholders may benefit from some initial facilitation by neutral third parties.

3.2.3. *Outcomes*

Outcomes will be expressed both in ‘synchronic’ terms (i.e. those that have immediate or ongoing impact) and also in ‘diachronic’ terms (i.e. those most manifest at the end of the project life cycle). And it is the diachronic outcomes where most bad experiences are to be found, at least as seen from a stakeholder perspective. These may be evident in damage to occupational, public and environmental health, in legacy wastes, or in a boom and bust financial cycle, leaving the stakeholder in a worse position than before the project began. But the negative outcomes may be invisible in nature, such as failures to build transferable skills, or the absence of technology transfer or spillover into other areas of economic activity. It is because of the risk of such accumulated negative impacts that transparent governance and accountability need to be in place from the earliest stage of any project, including engagement with concerned parties at the earliest opportunity. Such governance requirements must be complemented by coherent and consistent laws and regulations, with associated powers of enforcement. That will enable early stage agreement on verifiable performance measures, open reporting and equitable access to profit, including no corruption. Here governments have a key part to play in that there is too much evidence of major projects simply ‘bypassing’ the official economy of a country or region.

3.3. **Obstacles to success in social licensing**

In analysing why projects have failed, even after an initial social licence has been achieved, a number of broad conclusions have been reached. The list is not exhaustive, but the causes of failure tend to be similar in each case:

- (a) The company sees the gaining of a social licence in terms of a series of tasks or transactions (in effect, making a deal), with a strong emphasis on the initial grant of the licence. The community grants the licence on the basis of the expected nature and quality of a long term relationship. The asymmetry of expectation, and the imbalance in terms of the expected longevity of the partnership, causes profound failure, most damagingly of trust.
- (b) The company in its urgency to achieve a go-ahead:
 - (i) Confuses acceptance with approval;
 - (ii) Confuses cooperation with trust;
 - (iii) Confuses technical credibility with social credibility.

These points of confusion disturb the equilibrium at the heart of the licence.

- (c) The company:
- (i) Fails to understand the local community (social profile) and the local ‘rules of the game’ and so is unable to establish social legitimacy;
 - (ii) Delays stakeholder engagement;
 - (iii) Fails to allocate sufficient time for relationship building;
 - (iv) Undermines its own credibility by failing to give reliable information or, more commonly, failing to deliver on promises made to the community;
 - (v) Fails to respect and listen to the community;
 - (vi) Underestimates the time and effort required to gain a social licence;
 - (vii) Overestimates (or, worse, assumes) the quality of the relationship with the community.

4. SUSTAINING THE SOCIAL LICENCE: CRITICAL CONTROL POINTS IN POLICY AND MANAGEMENT

Micromanaging a whole NORM mineral life cycle, such as that for phosphate resources, is both wholly unrealistic from an operational point of view, and unaffordable. But when faced with a similar problem of quality management in a complex production environment the food industry adopted a macromanagement strategy instead, with excellent results. The technique deployed was the ‘hazard analysis and critical control point’ (HACCP) methodology. This identified a small number of ‘macros’, expressed as critical control points where progress could be monitored and change could be managed system wide in a way that maintained and even enhanced overall systemic efficiency and safety. By addressing these control points in a coherent and consistent way, systemic improvement was achieved. Applying HACCP theory to sustainable, socially licensed projects, such as for the comprehensive extraction of phosphate resources, yields a high level approach to resource management as shown in Fig. 2.

The key to success is thought to be the use of ‘one up, one down’ management techniques, where each stakeholder is required not just to focus on their own best interest but also on understanding how the stakeholder ‘one up’ and ‘one down’ in the supply chain thinks and behaves, and what implications such thoughts and behaviours have for all of them. A high level model of the phosphate supply chain is shown in Fig. 3. For a given stakeholder to function optimally in the supply chain, he or she needs all the time to understand and cooperate with stockholders or concerned parties ‘above’ or ‘below’ him in the life cycle, and work with them on a proactive, cooperative basis. Accordingly, Fig. 3 sets out some of the key concerned parties in the sustainable resource life cycle, their roles

Sustainable, socially licensed comprehensive uses of phosphate ores (P^S)

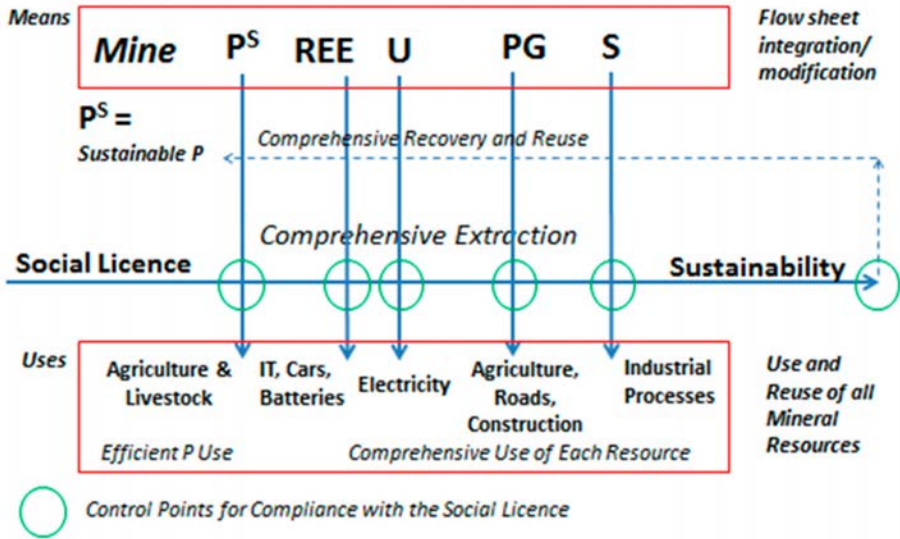


FIG. 2. Comprehensive extraction in operation.

Stakeholders	Needs	Control Point(s)	Equilibrium
Geology and mining engineers	– Exploration and discovery	– Verifiable classification, characterisation and quantification of resources/ reserves	– Energy basin management – Green mines
Beneficiation and processing specialists	– Technologies – Flowsheets	– Recovery rate – Waste stream(s)	– Near perfect recovery (“Comprehensive extraction”) – Zero waste
Fertilizer and feed manufacturers and distributors	– Market(s)	– Commodity prices	– Value add services (extension service model) – Utility prices
Farmers/ livestock producers	– Fertile soil – Market(s)	– Critical P value	– Critical P level sustained – Stable prices and margins
Food processors	– Reliable supply chain	– Effective “one up/ one down” integration	– Stable prices and margins
Consumers	– Safe, affordable food	– Storage/consumption of food – Diet/ nutrition – Waste	– Diet/ lifestyle in balance (no obesity) – Zero waste
Waste managers and processors	– Prevention/ Collection	– Recovery and reuse rates	– Reward for prevention – Waste as concept of last resort

FIG. 3. Achieving and sustaining equilibrium across the resource management life cycle.

and responsibilities and how the new equilibrium is to be achieved and sustained. The conclusion from this application of Nash's model is that the outcome of this cooperative game called social licensing is a new equilibrium. This equilibrium, by aligning the incentives of both stockholders and concerned parties, enables a 'win/win' outcome, enabling a higher level of sustainability in operations while also stimulating greater coherence and consistency in regulations and standards worldwide, as these will derive in large measure from good practices.

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REMOVAL OF URANIUM AND THORIUM FROM KVANEFJELD RARE EARTH INTERMEDIATE PRODUCT

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Abstract

Rare earth elements are commonly encountered in close geological association with radioactive minerals. In particular, thorium is often found in the same minerals as rare earth elements, making their combined treatment necessary. The paper discusses the metallurgical flowsheet developed for the Kvanefjeld rare earth and uranium deposit located in southern Greenland. The Kvanefjeld and associated deposits are well recognized as one of the largest rare earth and uranium deposits in the world. The minerals which contain the rare earth elements also contain economically viable amounts of uranium and potentially economically viable amounts of thorium. Metallurgical studies using flotation have been successful in producing a high grade concentrate which consists of 13% rare earth oxide, 0.2% triuranium octaoxide and 0.8% thorium. Due to the unique nature of the minerals contained within the deposit, a customized hydrometallurgical flowsheet was developed to treat the concentrate. The hydrometallurgical flowsheet has been well tested and is capable of producing a high grade mixed rare earth product which is very low in uranium and thorium. A separate uranium oxide product can also be produced using commercially established solvent extraction. The Kvanefjeld project is currently at the feasibility study stage, with metallurgical designs of both the concentrator and hydrometallurgical plant well advanced. The paper discusses the metallurgical flowsheet and show the deportment of NORM throughout the flowsheet. The issues of NORM deportment and strategies for removal and safe disposal are also discussed.

1. INTRODUCTION

1.1. Kvanefjeld project summary

Greenland Minerals and Energy Ltd (GMEL) is a mineral exploration and development company operating in southern Greenland. GMEL is primarily focused on advancing the 100% owned Kvanefjeld multi-element project (both light and heavy rare earth elements, uranium and zinc) through the feasibility and permitting phase and into mine development. The Kvanefjeld project area is located in southern Greenland, approximately 10 km from the town of Narsaq (see Fig. 1), and is adjacent to deep water fjords that allow year round shipping

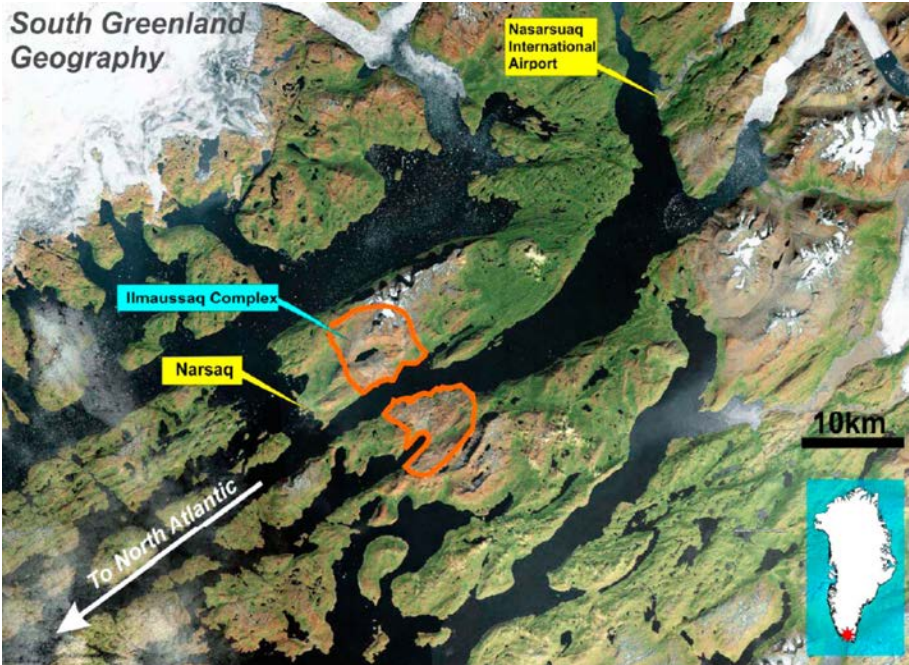


FIG. 1. Location of the Kvanefjeld deposit in Greenland.

access directly to the project area. Since acquiring the project in 2007, GMEL has completed an extensive exploration and scientific research programme which includes:

- (a) More than 57 000 m of diamond core drilling;
- (b) Geological modelling and mine design;
- (c) Metallurgical batch and continuous piloting testwork;
- (d) Engineering design, inclusive of capital and operating costs.

GMEL's exploration and research programme follows on from the extensive historical work conducted by the Danish authorities and scientists in the 1970s and early 1980s, which resulted in a pre-feasibility study being published by Risø National Laboratory in 1983. Through a rigorous technical programme, an optimum flowsheet was identified and developed by GMEL. The flowsheet consists of flotation to produce a mineral concentrate and then atmospheric sulphuric acid (H_2SO_4) leaching of the concentrate. This paper describes the hydrometallurgical treatment of the Kvanefjeld mineral concentrate. During the hydrometallurgical treatment, secular equilibrium is disturbed. The department

of the radioactive elements is described along with removal methods to prevent contamination of the rare earth intermediate product.

1.2. Kvanefjeld geology

Kvanefjeld is a unique multi-element deposit hosted within marginal phases of the Ilimaussaq Intrusive Complex, located in the south-western tip of Greenland. As shown in Fig. 2, another two deposits, Sorensen and Zone 3, are located close to Kvanefjeld and share very similar chemistry. The deposit is exposed at the surface along a series of undulating bluffs on a broad peninsula surrounded by deepwater fjords that run directly out to the Atlantic Ocean. The global resources to date include 956 million t at 273 ppm triuranium octaoxide (U_3O_8), 1.08% total rare earth oxides including yttrium oxide (TREO) and 0.23% zinc (see Table 1). The deposit is a truly world class resource for the strategic metals uranium and rare earth elements.

1.3. Mineralogy

The deposit type is known as an agpaitic nepheline syenite. Agpaitic deposits contain silica undersaturated alkaline rocks with an alkali to aluminium ratio in excess of unity. They also contain a suite of complex alkali titanio, niobio and zircono silicates. The ore rock type present is called lujavrite



FIG. 2. Ilimaussaq, showing Kvanefjeld, Sorensen and Zone 3.

TABLE 1. COMBINED KVANEFJELD, SORENSEN AND ZONE 3 RESOURCES

Cut-off (ppm U ₃ O ₈)	Resource		Grade (ppm)						
	Classification	Amount (Mt)	U ₃ O ₈	Zn	Rare earth oxides (lanthanides) (REO)			Y ₂ O ₃	REO + Y ₂ O ₃ (TREO)
					Light REO	Heavy REO	Total		
150	Indicated	437	274	2 212	9 626	402	10 029	900	10 929
	Inferred	519	272	2 468	9 438	383	9 820	867	10 688
	Total	956	273	2 351	9 524	392	9 916	882	10 798
200	Indicated	291	325	2 343	10 452	419	10 871	978	11 849
	Inferred	354	320	2 731	10 171	387	10 558	919	11 477
	Total	645	322	2 556	10 298	401	10 699	946	11 645
250	Indicated	231	352	2 363	10 950	443	11 281	1 032	12 312
	Inferred	260	355	2 871	10 548	401	10 813	957	11 769
	Total	491	353	2 632	10 737	421	11 033	992	12 025

REMOVAL OF URANIUM AND THORIUM FROM KVANEFJELD

which is an agpaitic plutonic rock dominated by aegirine or arfvedsonite, along with nepheline, alkali feldspar, zircon silicates and a host of rare minerals. The mineralogy of the Kvanefjeld deposit is different from that of other commercialized uranium and rare earth deposits. A deposit similar to Kvanefjeld called Lovozero is currently mined for rare earth elements in the Kola Peninsula, Russian Federation. Within Kvanefjeld, steenstrupine is the dominant host to rare earths and uranium in all ore domains. It is a complex sodic phosphosilicate mineral and mineralogical studies suggest that it commonly contains 0.2–1% U_3O_8 ; it is likely to host approximately 50% of the uranium at Kvanefjeld. Other minerals that are important hosts to rare earths include the phosphate mineral vitusite and to a lesser extent cerite and monazite. Aside from steenstrupine, uranium is also hosted in unusual sodic silicate minerals that are rich in yttrium, heavy rare earths, zirconium and tin. Minor amounts of uranium are also hosted in uranothorite and monazite. Zinc is almost exclusively hosted in sphalerite, which is the dominant sulphide throughout the deposit. Figure 3 shows the typical mass proportions of minerals observed in the ore. Note that the value minerals only make up a small proportion of the overall mass which renders the ore amenable to concentration.

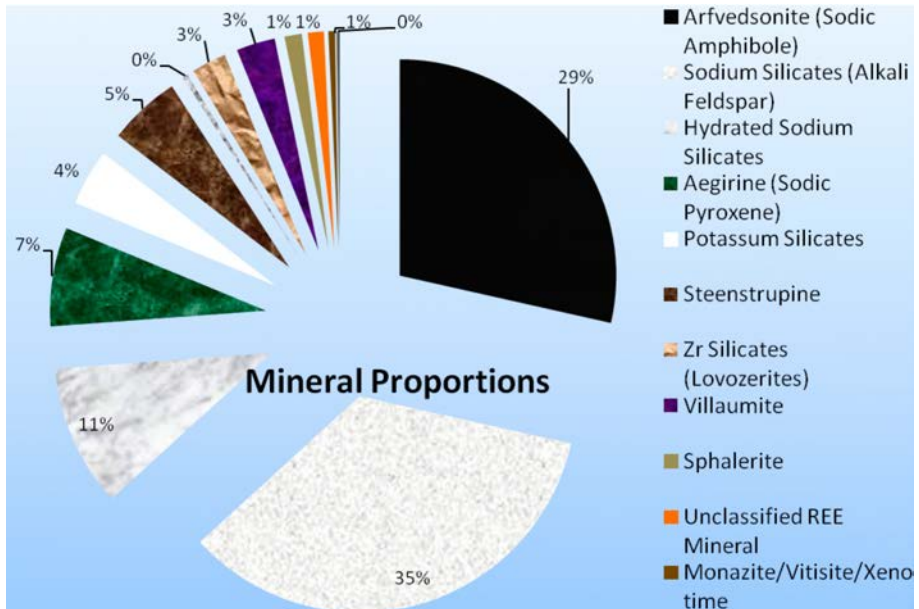


FIG. 3. Minerals and their proportions in Kvanefjeld ore.

Based on the geochemistry of minor elements, a number of discrete ore domains within the deposit have been identified. These domains approximate to horizontal layers of slightly differing compositions within the deposit. It is also clear that the mineralogy gradually varies between each of the domains. Domain A will account for approximately 40% of the process plant feedstock over the life of the project. Details of the various domains are as follows:

- (a) Domain A is closest to the surface and is relatively enriched in heavy rare earths and phosphorus.
- (b) Domain B occurs just below domain A and contains similar U_3O_8 grades to Domain A, but with lower levels of heavy rare earths.
- (c) Domain C occurs just below Domain B and has lower uranium and rare earth grades, a lower phosphorous content and increasing amounts of zirconium and tin.
- (d) Domain D occurs just below Domain C and has a high zirconium and low phosphorus content — lower grades are also observed.
- (e) Domain MC represents an area of the deposit which has undergone hydrothermal alteration and is elevated in uranium while still being relatively high in phosphorus.

2. GENERAL METALLURGY

2.1. Kvanefjeld metallurgical flowsheet

After an extensive metallurgical flowsheet selection process, a single ‘go forward’ flowsheet was selected in March 2012. The process is essentially flotation followed by a H_2SO_4 atmospheric leach on the flotation concentrate. There are additional stages following the leach which are needed to separate the minerals of value from the gangue elements in the solution and residue. These additional stages are also designed to remove radionuclides such as uranium and thorium from the rare earth intermediate product. A high level block flow diagram of the flowsheet is provided in Fig. 4.

2.2. Flotation

2.2.1. Zinc flotation

Following comminution, the first stage of flotation consists of zinc removal to produce an additional saleable product and, by doing so, to remove an unwanted contaminant from the downstream processing. The flotation conditions for zinc

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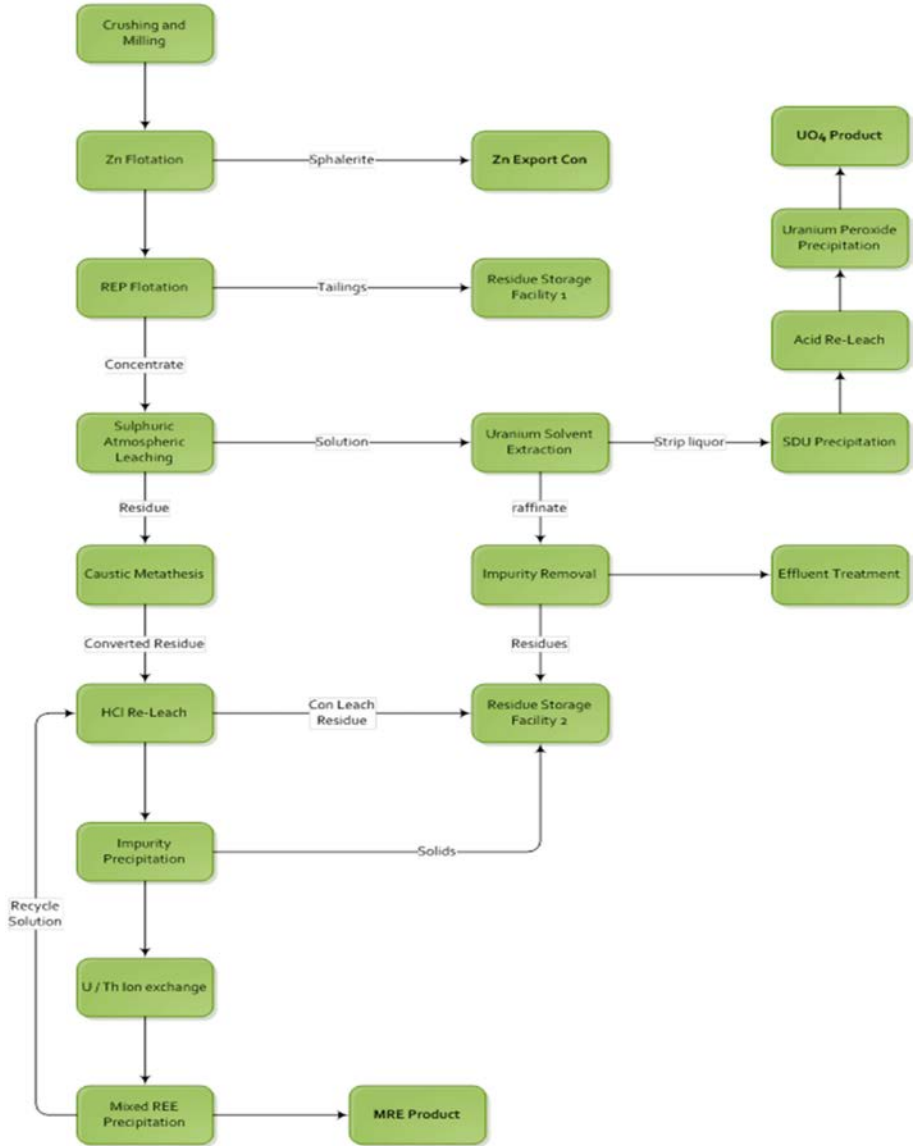


FIG. 4. Processing block flow diagram.

removal are well established commercially with copper sulphate activation and xanthate collector. Overall, approximately 70% of the zinc is removed in this stage into a high grade concentrate which will be marketed as a separate product.

2.2.2. *Rare earth phosphate flotation*

The rare earth phosphate (REP) flotation is an important part of the flowsheet as it essentially concentrates the majority of minerals of value into less than 10% of the original mass. Treating this concentrate, rather than whole of ore, dramatically reduces the capital and operating costs of the hydrometallurgical process plant. Flotation is performed with commercially available reagents which include sodium silicate and hydroximates. The flotation circuit consists of roughing, scavenging and cleaning of the concentrate. Extensive laboratory testwork has included multiple pilot plant campaigns. The concentrate derived from the first pilot plant was used as the main feed source for the hydrometallurgical investigations. Further bench scale testwork and a second pilot plant campaign completed in 2012 demonstrated a slightly higher rare earth grade concentrate than used in this hydrometallurgical programme. The concentrate produced in each campaign was derived from Domain A ore. A comparison of the composition of Domain A ore and REP concentrates from the first and second pilot plants is shown in Table 2. The leach response of concentrates derived from other domains has been investigated to determine ore variability.

In the Kvanefjeld ores, cerium can be used as a proxy for the upgrade of light rare earth elements while yttrium can be used as a proxy for the upgrade of heavy rare earth elements. The concentrate from the second pilot plant generated a higher grade concentrate with significantly lower reagent consumption through optimization of the process conditions and better equipment selection. Significantly, the ratio of TREO to aluminium of the second pilot plant concentrate is more than double that achieved in the first and will result in a very significant reduction in acid consumption in the acid leach stage.

2.3. **Leaching**

The successful generation of an enriched concentrate has enabled the use of acid leaching to recover the minerals of value. Most of the acid consuming gangue is rejected during flotation; however, sufficient reactive silica remains in the concentrate requiring precautions to be taken against the formation of silica gel. Under the leach conditions selected, a portion of the silica dissolves and precipitates in a filterable form. Most of the developmental test work was completed using concentrate generated from the first flotation pilot plant. A schematic diagram showing the main unit operations for the leach and rare earth circuit is shown in Fig. 5.

TABLE 2. COMPOSITION OF DOMAIN A ORE AND REP CONCENTRATES FROM THE FIRST AND SECOND PILOT PLANTS

	Concentration (%)										
	Al	Ce	Fe	P	Si	U	Y	Zn	Th	TREO	
Ore	6.19	0.593	10.1	0.29	24.2	0.037	0.121	0.3	0.075	1.41	
REP concentrate, pilot plant 1	4.42	4.19	8.58	2.38	18.8	0.150	0.66	1.10	0.61	11.3	
REP concentrate, pilot plant 2	2.56	5.77	7.68	3.58	16.4	0.243	0.99	0.49	0.70	15.6	

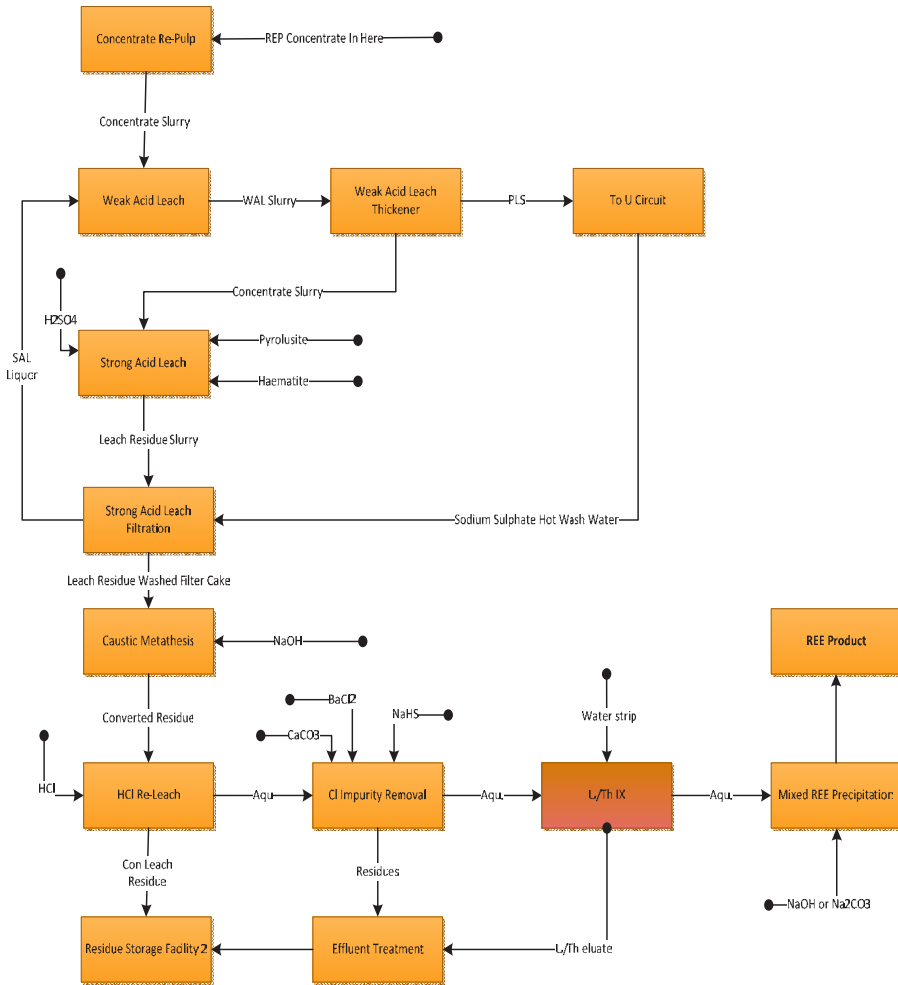


FIG. 5. Rare earth circuit block flow diagram.

3. HYDROMETALLURGICAL REFINING

3.1. Sulphuric acid leach

Damp concentrate is leached with H_2SO_4 in a two stage counter current circuit in order to minimize acid consumption and produce a uranium solvent extraction (USX) feed with low free acid. Fresh concentrate is contacted with strong acid leach discharge liquor in the weak acid leach circuit. Some of the rare earths and uranium are also partially leached. The leach conditions required

to prevent gelling of the silica also result in the precipitation of rare earths as a sodium double sulphate and maximizes the overall rare earth recovery to the final product.

The main elements leached from the concentrate include sodium, aluminium, iron, manganese, calcium, silicon, thorium, uranium and rare earths. The rare earths, calcium silicon and thorium reprecipitate into the residues while the other elements remain in solution, which provides an effective mechanism for separating the rare earths from the problematic gangue.

The residues from the weak acid leach are thickened and report to the strong acid leach circuit where fresh acid is added to increase the free acid to about 80 g/L in solution and results in further leach extraction of minerals of value. Depending on the ore treated, there may be a requirement to add oxidant to the leach to improve uranium recovery. There is also an option for adding haematite in order to increase the iron:phosphorus ratio of the weak acid leach liquor to prevent uranium precipitation; however, to date this has not been required. The strong acid leach residues are filtered and thoroughly washed to remove entrained liquor. Test work has shown that, provided that silica precipitation is controlled, the thickened slurry is amenable to vacuum filtration. Minor dissolution of rare earths occurs but these are recovered in the leach circuit. The leach provides effective separation between the rare earths and the uranium and the vast majority of the impurities.

Batch testwork was used to identify the leach conditions required for elemental dissolution but the behaviour of silica is best determined by continuous testwork. A semicontinuous programme was utilized to enable the behaviour of silica to be assessed. A two stage semicontinuous leach was conducted in order to verify the leach conditions identified in the batch programme. Acid solution and feed slurry were pumped into a 15 L (live volume) reactor at a fixed ratio to simulate the leach. Slurry was pumped out of the reactor hourly and samples taken every 4 h for assay. The leach targeted conditions under which the silica leached and precipitated without forming gels. This required the specific reactive silica feed rate to the reactor to be controlled to below 0.7 g/h of reactive silica per litre of reactor live volume.

The results for the weak acid leach and the strong acid leach stages are summarized in Table 3. It is necessary to distinguish between elemental dissolution and elemental attack. Elemental dissolution represents the proportion of the element which was leached from the solids and remains in solution. Elemental attack represents the proportion of the element which remains in solution as well as the proportion which has been leached but has reprecipitated in a sparingly soluble form. Where no sparingly soluble salts are generated the dissolution and the attack value are the same. Where significant reprecipitation has occurred, the attack value is significantly larger than the dissolution value

TABLE 3. RESULTS OF TWO STAGE SULPHURIC ACID LEACHING OF DOMAIN A ORE

	Al	Ca	Fe	K	Mn	Na	P	Th	La	Ce	Nd	Y	U
Weak acid leach (%)													
Dissolution	13	8	<0.1	2	41	17	37	8	0.3	0.3	0.3	8	68
Attack	15	69	<0.1	10	40	44	39	15	68	69	70	71	70
Strong acid leach (%)													
Dissolution	50	11	2	2	57	18	92	<1	0.2	0.2	0.2	4	83
Attack	50	88	4	11	56	53	94	57	97	97	97	88	85

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and is evident for the rare earths, thorium, sodium and calcium. The results show that the rare earths have a high degree of attack but very low dissolution. The heavy rare earths show a lower degree of double salt precipitation due to the higher solubility of their double salts and their lower initial head grade in the concentrate. Importantly, the uranium, aluminium, manganese, phosphorous and iron do not form sparingly soluble salts. The rare earths are almost entirely present in the residues as labile double salt and are readily separated from the impurities in solution.

The weak acid leach stabilizes at pH2.1 and effectively neutralizes the majority of the residual acid in the strong acid leach liquor. The strong acid leach achieves a terminal free acid concentration of about 80 g/L. Acid consumption across the leach is 323 kg/t. Most of the acid consumption is through the dissolution of aluminium (30%), sodium (21%), calcium (11%) and rare earths (30%). Iron is mainly present in arfvedsonite and is largely refractory with about 4% iron dissolution under the leach conditions. The behaviour of silica in the leach is critical for achieving reasonable solid–liquid separation. During the leach, solution samples were taken and assayed for silicon in solution. Figure 6 shows how the concentration of silicon in solution varied with leach time during a continuous leach test.

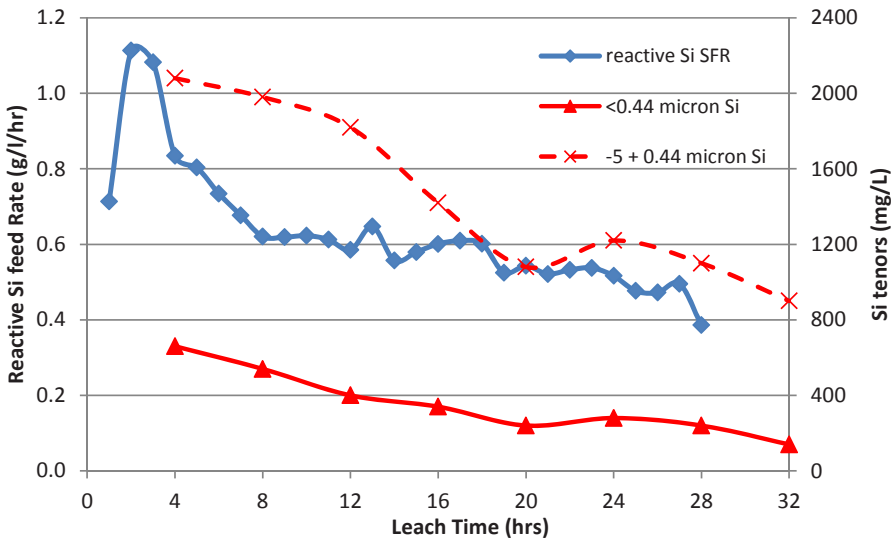


FIG. 6. Silicon tenors and reactive silicon feed rate versus leach time during semicontinuous H₂SO₄ leach.

During the course of the leach, silicon in solution is trending down, indicating that it is precipitating or growing faster than it is leaching. The $<0.44 \mu\text{m}$ silicon values are also trending down and indicate that the very fine silica particles are growing into larger more filterable particles. Filtration of the slurry is observed to improve over the course of the leach. A specific reactive silica feed rate of 0.6 g/h per litre of live reactor volume (equivalent to a 10 h residence time) is sufficient to precipitate the silica in a form which has good handling properties. The resulting slurry from these tests is amenable to vacuum filtration which will facilitate efficient washing. While the majority of the uranium reports to the weak acid leach liquor, most of the thorium follows the rare earths into the residues.

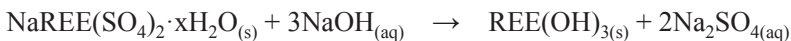
3.2. Uranium solvent extraction

Uranium is recovered from the weak acid leach liquor via conventional solvent extraction using alamine 336 as the extractant. The loaded organic will be scrubbed with dilute acid to remove impurities, particularly zirconium, prior to stripping. A schematic diagram showing the main unit operations in the uranium recovery circuit are shown in Fig. 7. The loaded organic is stripped with sodium carbonate (Na_2CO_3) which de-protonates the organic and completely strips the uranium from the organic. Uranium is precipitated from the pregnant carbonate strip liquor as sodium diuranate (SDU) using sodium hydroxide (NaOH). The SDU is re-leached in H_2SO_4 and then precipitated as uranium peroxide. The peroxide product will be dried and package as a final product.

The USX raffinate contains very low levels of rare earths but high levels of aluminium and to a lesser extent iron and manganese. The raffinate will be treated with limestone to precipitate iron, thorium and aluminium. Manganese is precipitated from the resulting slurry using lime and air, the slurry is then thickened and the underflow stored sub-aqueously in a lined tailings storage facility. The recovered water, mainly comprising sodium sulphate, is recycled to the two stage H_2SO_4 leach circuit.

3.3. Metathesis

The washed residues from H_2SO_4 leaching contain the rare earths as double sulphate salts, gypsum, precipitated silica and residual unreacted gangue (feldspars and arfvedsonite). The rare earths can be converted to hydroxides (or carbonates) via metathesis according to the following reaction:



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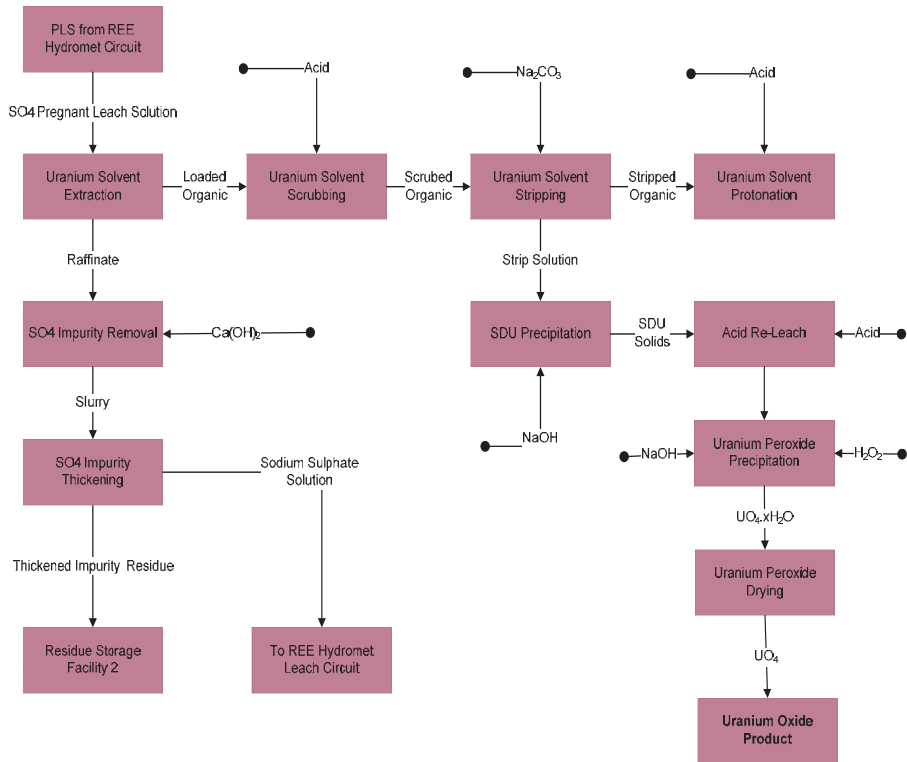


FIG. 7. Uranium circuit block flow diagram.

The conversion of gypsum to lime and silica to sodium silicate competes with the double salt reaction and increases reagent consumption and impurity levels in solution.

A series of batch tests was conducted to evaluate the impact of temperature, sodium sulphate levels, reagent type and reagent dose on metathesis performance. Metathesis conditions are selected to convert the double sulphate to the alkali precipitate while minimizing silica redissolution. As expected, the slurry pH was the most important variable; a pH above 12 was required to achieve high conversion with caustic. As the temperature increased from 10°C to 65°C, conversion improved slightly, but silica dissolution also increased significantly. At temperatures above 30°C, sodium sulphate tenors up to 40 g/L have little impact on double salt conversion. The results of metathesis using NaOH or Na₂CO₃ at ambient temperature (30°C) are given in Table 4.

Both reagents generate a similar product with about 0.5% residual sulphur in the final solids. This sulphur is not soluble in water and is not associated with rare earth double sulphate. All the elements except sulphur show an upgrade

TABLE 4. METATHESIS FEED AND PRODUCT ASSAYS

	Reagent dose (kg/t)	Solids assay (%)										
		Ce	La	Y	Th	U	Na	S	Si	Fe	Ca	Mn
Feed	—	4.05	2.28	0.45	0.43	0.024	2.80	4.54	16.4	7.42	1.45	1.08
NaOH	97	5.01	2.32	0.54	0.54	0.026	3.30	0.53	20.0	9.37	1.61	1.23
Na ₂ CO ₃	101	4.90	2.82	0.53	0.60	0.029	3.40	0.50	20.8	9.87	1.8	1.37

due to the mass loss associated with the conversion; surprisingly, sodium also shows an upgrade. The final solution has very few dissolved metals and mainly comprises sodium, sulphate, residual reagent and minor dissolved silica.

The presence of oxygen in the system oxidises cerium to the Ce(IV) state which reduces the cerium dissolution in the acid leach. Following metathesis, the converted solids are filtered and washed to remove entrained sulphate. The washed solids are then leached with hydrochloric acid (HCl) to recover the rare earths.

3.4. Hydrochloric acid leach

Following the solid–liquid separation, the metathesis residues are leached with HCl to selectively leach rare earths. The impact of pH on metal dissolution at ambient temperature was determined, and is presented in Figs 8 and 9.

Leaching is rapid with significant rare earth dissolution achieved at pH4; rare earth recovery increases slowly with decreasing pH. Most of the acid consumption (67 kg of HCl (anhydrous) per tonne) is required to achieve pH4 and is attributed to rare earth and calcium dissolution. Further acid addition, to 83 kg/t, results in the pH decreasing to 1.5 with a further ~25% (relative) increase in rare earth recovery.

The relatively low dissolution of light rare earths is due to these elements being associated with monazite in the original feed. Cerium dissolution is

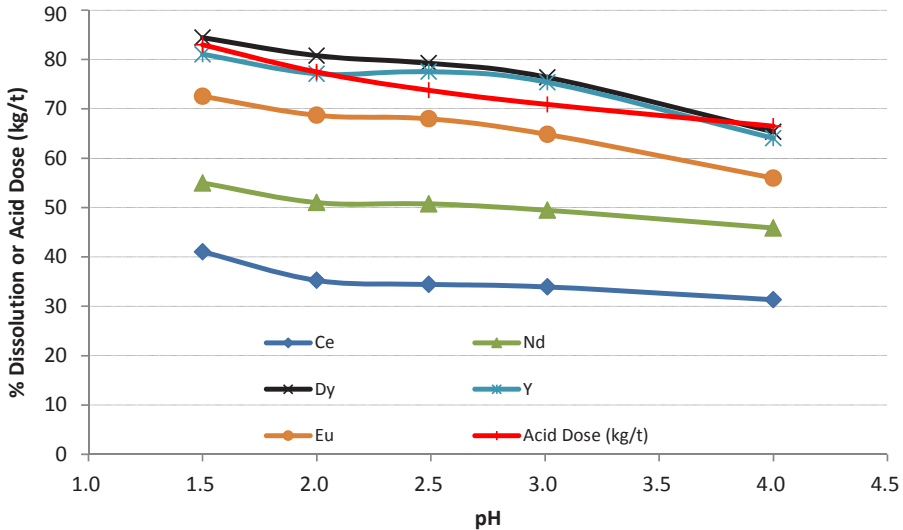


FIG. 8. Metal recovery in HCl leach of metathesis product at various pH values.

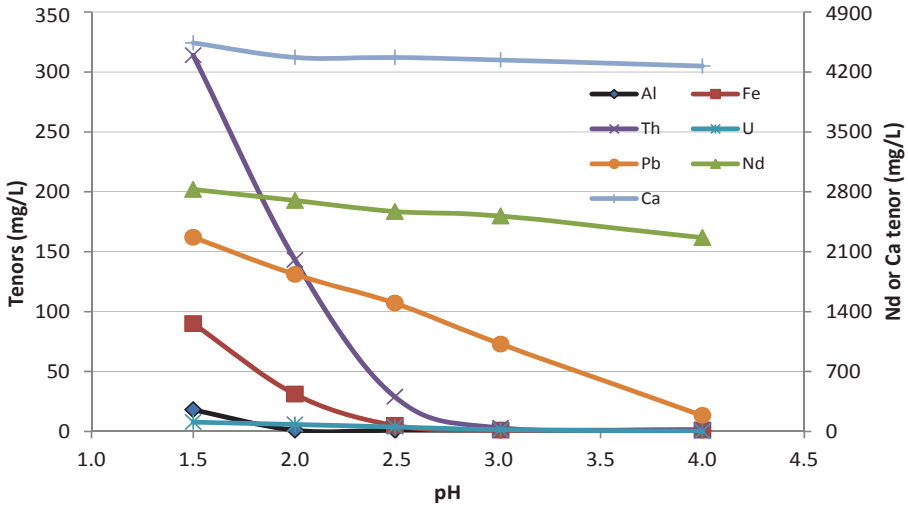


FIG. 9. Impurity dissolution in HCl leach of metathesis product at various pH values.

significantly below that of neodymium and is attributed to oxidation of the cerium with air during metathesis. With the correct precautions, this effect will be negligible in commercial operations. With the exception of cerium, the rare earth recovery achieved at pH 1.5 reflects that achieved in the H_2SO_4 leach stage for the same sample and suggests that all the rare earths reporting to the H_2SO_4 leach residue as double salt are being recovered in the HCl leach.

Calcium tenors in solution are high and relatively unchanged with decreasing pH and indicate that the gypsum is converted during the metathesis, probably to lime which readily dissolves in the HCl leach. Lead tenors steadily increase with increasing acid dose. Some of the lead is radiogenic and can represent a significant radioactive impurity in the rare earth product. Thorium tenors are low at pH values above 3, but increases dramatically below pH 3. Iron in solution increases at pH values below 2.5 but remains below 100 mg/L at pH 1.5, reflecting the refractory nature of the iron in the residues. Aluminium tenors in solution remain very low and reflect the stability of the feldspars under the leach conditions. The composition of the HCl leach products and feed are summarized in Table 5. The solution is enriched with rare earths with minor aluminium, iron, uranium and manganese in solution. Higher levels of silicon, lead and thorium are in solution and all would need to be removed prior to recovering a rare earth product.

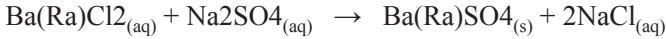
TABLE 5. RESULTS OF THE HCl LEACH OF METATHESIS PRODUCT AT pH1.5

	Assay value		
	Feed (%)	Residue (%)	Precipitated leach solids (mg/L)
Ce	5.01	3.19	5530
La	2.32	1.45	4280
Nd	1.88	0.93	2828
Dy	0.119	0.017	229
Y	0.740	0.130	1380
Th	0.540	0.610	314
U	0.025	0.031	8
Na	3.30	4.00	1190
S	0.528	0.68	534
Si	20.0	22.6	364
Al	1.66	2.07	18
Fe	9.37	9.81	90
Ca	1.61	0.269	4540
Pb	0.11	0.053	162
Mn	1.23	1.54	3

3.5. Impurity removal

Impurity removal is undertaken in a number of stages. The aluminium, iron and most of the thorium are removed by adjusting the pH to about 4 with lime. Rare earth losses at pH4 are minor especially when low levels of iron and aluminium are precipitated and represent less than 0.5% loss.

Barium chloride (BaCl_2) is also added to the iron removal stage to remove radium from solution via co-precipitation in accordance with the following reaction:



The flowsheet also includes a sulphiding step once the iron has been precipitated to remove residual lead and zinc and to provide a mechanism for polonium and bismuth removal from solution. The sulphiding reaction is:



Zinc, polonium, arsenic and bismuth undergo similar reactions and, provided the pH is controlled above 3, almost quantitative removal should be achieved. While polonium, arsenic and bismuth levels are very low, they will be removed from solution via co-precipitation. Rare earth losses are negligible, as they do not form sulphides.

The sulphide precipitate is formed and blended with the previous precipitation stage to dilute the expected high radioactivity of the sulphide precipitate. The mixed precipitate is thickened, filtered and stored in the residue storage facility.

3.6. Ion exchange

Low levels of uranium and thorium remain in solution following impurity removal and need to be removed prior to recovering the rare earths. The low impurity levels lend themselves to the use of ion exchange. A range of resins was evaluated to identify a suitable functional group to extract uranium and thorium away from the rare earths in the chloride solution. The reagent identified was capable of extracting uranium and thorium to below 0.1 mg/L with minimal rare earth loading and provides an effective mechanism for polishing the solution prior to rare earth recovery. Typical results from ion exchange tests are given in Table 6. The results are for a single batch contact at a liquor to resin ratio of 100 and shows that the resin was effective at reducing already low levels of uranium and thorium in solution to very low levels despite the high background of rare earths in solution. Some rare earth loading was observed. However, this can be minimized using a lead-lag configuration.

TABLE 6. ION EXCHANGE FEED AND PRODUCT LIQUOR ASSAYS

	Ion exchange assay (mg/L)									
	Th	U	La	Ce	Nd	Dy	Y	Ca	Pb	Zn
Feed	0.37	2.33	2240	3900	2085	150	998	2720	61	3
Product	0.03	0.02	2120	3630	1877	134	911	2640	54	3

3.7. Rare earths recovery

Following ion exchange, the solution comprises the rare earths with significant amounts of calcium, sodium and minor amounts of magnesium and manganese. Various reagents to recover the rare earths have been investigated. Both NaOH and Na₂CO₃ generated reasonable product grades. A typical hydroxide solid assay is given in Table 7. The results show that the uranium and thorium tenors in the product were low and demonstrate the effective removal of these elements using the flowsheet. Since this product was generated without sulphide precipitation, lead is a significant contaminant in the final product but this will be significantly reduced once sulphide precipitation is employed. Sulphur, representing 9.6% sulphate, is the single most significant contaminant which may be reduced by increasing the pH at which precipitation takes place.

The hydroxide product was also submitted for radionuclide analysis and the results are summarized in Table 8. The results show that the thorium decay chain contributes very little activity and that the control measures are effective in controlling these impurities. In the ²³⁸U decay chain, the current measures were effective in controlling thorium and radium but the lead and polonium remained, and are a source of activity. The sulphide precipitation unit operation should effectively remove most of the remaining activity and significantly diminish the residual activity from the ²³⁸U decay chain. The ²³⁵U decay chain is the source of approximately half the total activity. The uranium, thorium and protractinium are effectively controlled using the current process, but a high proportion of the actinium follows the rare earths into the final product and, along with its progeny, will be the most significant source of activity in the rare earth product.

TABLE 7. MIXED RARE EARTH HYDROXIDE PRODUCT ASSAYS

		Assay (%)										
Product	La	Ce	Pr	Nd	Gd	Dy	Er	Yb	Y	TREO		
NaOH	13.9	23.9	3.0	8.6	0.72	0.80	0.31	0.15	5.9	69.5		

		Assay (ppm)										
Product	Al	Fe	As	Ca	Mn	Pb	Si	S	U	Th		
NaOH	360	500	1 200	380	166	4 000	16 000	32 000	2	2		

TABLE 8. RADIONUCLIDE CONCENTRATIONS IN MIXED RARE EARTH HYDROXIDE PRODUCT

Decay chain	Radionuclide	Activity (Bq/g)
Th-232	Th-232	0.006 9
	Ra-228	<0.012
	Th-228	<0.01
U-238	U-238	0.021
	Th-230	<1.1
	Ra-226	<0.037
	Pb-210	22.7
	Po-210	27.1
U-235	U-235	<0.069
	Pa-231	<0.16
	Ac-227	8.4
	Th-227	8.4

4. CONCLUSIONS

- (a) A two stage counter current H_2SO_4 leach is effective in dissolving uranium while keeping essentially all the rare earths and most of the thorium in the residues. Conditions for preventing silica gel formation have been confirmed in continuous test work.
- (b) The uranium can be effectively separated from the impurities in the sulphate leach solution and recovered into a uranium peroxide product using commercially proven technology.
- (c) The rare earths in the H_2SO_4 leach residues can be converted to hydroxides and effectively dissolved under mildly acidic condition with HCl. The resulting pregnant chloride leach liquor comprises rare earths with relatively low levels of contaminants.
- (d) Iron, aluminium and most of the thorium are effectively precipitated from solution at pH3.5 with minimal rare earth losses. The addition of $BaCl_2$ to this precipitation stage is effective at removing radium from solution but actinium, polonium and lead remain in solution.
- (e) A sulphide precipitation stage should effectively eliminate polonium, bismuth, lead, arsenic and zinc from solution. This will significantly reduce the presence of the lead and polonium radionuclides in the final rare earth product.

- (f) Residual uranium and thorium remaining after impurity precipitation from the chloride solution are effectively removed from solution using ion exchange. Despite high background levels of rare earths in solution, the relatively low levels of uranium and thorium in solution can be reduced to less than 0.1 mg/L.
- (g) A rare earth hydroxide product containing 70% TREO can be generated from the purified solution with sulphate being the main contaminant (9.6%). Uranium, thorium and radium contamination of the product is minimal and demonstrates that the current flowsheet controls are effective. Actinium remains the sole radionuclide contaminant in the rare earth product.

ACKNOWLEDGEMENTS

Acknowledgement goes to the management of Greenland Minerals for permission to publish this paper. Special acknowledgement goes to J. Lawson of ANSTO Minerals who provided his technical expertise in dealing with silica gels. Greenland Minerals would also like to thank SGS Orestest, ANSTO and ALS AMMTEC who performed the test work presented in this paper.

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SAMPLING AND MEASUREMENT OF ^{210}Po IN THE WASTE STREAMS OF A COAL FIRED POWER PLANT

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Abstract

In the paper, a stack sampling instrument was developed to sample the aerosol radionuclides from a coal fired power plant (CFPP). Based on the sampling instrument, a 225 MW(e) coastal coal fired electric utility plant was sampled to determine radionuclide concentrations in all major process streams including coal, fly ash and bottom ash samples and six naturally occurring radionuclides: ^{238}U , ^{226}Ra , ^{210}Pb , ^{210}Po , ^{232}Th and ^{40}K were analysed by gamma spectrometry and alpha spectrometry. For this plant, radionuclide balances and the amount of aerosol ^{210}Po emitted in the stack effluent were determined. The amount of aerosol ^{210}Po emitted from the stack was normalized for the total electric power of the CFPP. It was found that $2.5682 \times 10^8 \text{ Bq}\cdot\text{GW}^{-1}\cdot\text{a}^{-1}$ of ^{210}Po was released into the environment from this coastal CFPP. In the paper, a measurement technique for ^{210}Po in seawater was also developed and the ^{210}Po in the waste desulphurization seawater was analysed. It was found that the concentration of ^{210}Po in the desulphurization seawater was an order of magnitude higher than in normal seawater. In the paper, the micromorphology of the aerosol ^{210}Po from the stack was analysed by a scanning electron microscopy (SEM) technique and a preliminary method for distinguishing the source of aerosol ^{210}Po in the air around the CFPP was developed based upon the SEM technique.

1. INTRODUCTION

Coal, like other ores found in nature, contains trace quantities of the naturally occurring primordial radionuclides. It has been observed that the radionuclides are concentrated in the smaller fly ash particles which have larger surface to volume ratios (surface effect) and the hot flue gases cool down on their way to stack [1]. Therefore, the combustion of coal results in the release to the environment of some natural radioactivity and it can modify ambient radiation fields and exposure situations for people living around a coal fired power plant (CFPP).

In 1982, the United Nations Scientific Committee on the Effects of Atomic Radiation made a compilation of the knowledge on these industries, pointing out the fact that, within the wastes generated in some of those NORM industries, an important enrichment of the natural radioactive elements with low boiling point, related to the temperature reached in the industrial processes, could be observed (see annex C of Ref. [2]). Polonium is one of those low boiling point elements and ^{210}Po is the most abundant and ubiquitous of its isotopes. With a half-life long enough and a radiotoxicity high enough, ^{210}Po is of interest in radiation protection. Meanwhile, coal accounts for over 75% of China's total commercial primary energy consumption and has been crucially important to China's rapid economic expansion. Coal will inevitably remain the overwhelming indigenous energy resource for the foreseeable future [3]. Those reasons make the study of sampling and measuring natural radioactivity from the stack of a CFPP a necessity for China. More importantly, the readily volatile radionuclides such as ^{210}Pb and ^{210}Po deserve more attention.

2. MATERIALS AND METHODS

2.1. Physical and chemical characteristics of ^{210}Po in CFPPs

In order to design a sound and efficient sampling system to sample aerosol particles of ^{210}Po , it is necessary to study the chemical and physical characteristics of ^{210}Po at high temperature. In Ref. [4], a description can be found of the behaviour of polonium, based on evaporation–condensation effects, and modelling of some key aspects under those conditions. In the work described in Ref. [4], the chemical compounds formed by polonium before and after the process were characterized. In coal, polonium is expected to be found in two main chemical forms: pyrite, associated with a metal ($\text{M}_x\text{Po}_{1-x}$), or organically bound. It is pointed out in Ref. [4] that in the first stage of combustion, all polonium present in the natural material will evaporate in the form of elemental polonium. With the temperature and pressure falling, the elemental gaseous polonium together with other gases such as air, and a quantity of solid particles, become suspended in those gases. Though some research shows that polonium will condense in the form of heterogeneous condensation and homogeneous condensation [5], much of the aerosol particles containing the ^{210}Po will be released from stack.

2.2. Design of the sampling instrument

In order to sample the aerosol particles containing ^{210}Po , a sampling system was created. The sampling system adopts a paper filter to capture the aerosol particles. Constant velocity sampling was considered in this test. The sampling head of this system is shown in Fig. 1. The sampler head was inserted into the stack from the sampling hole and fastened in the stack, and then the fly ash was pumped to the paper filter through a special plastic pipe. The sampling procedures are shown in Fig. 2.

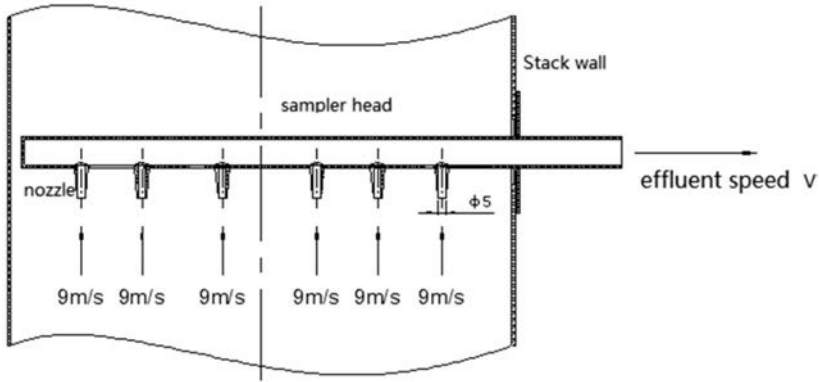


FIG. 1. Sampler head design.

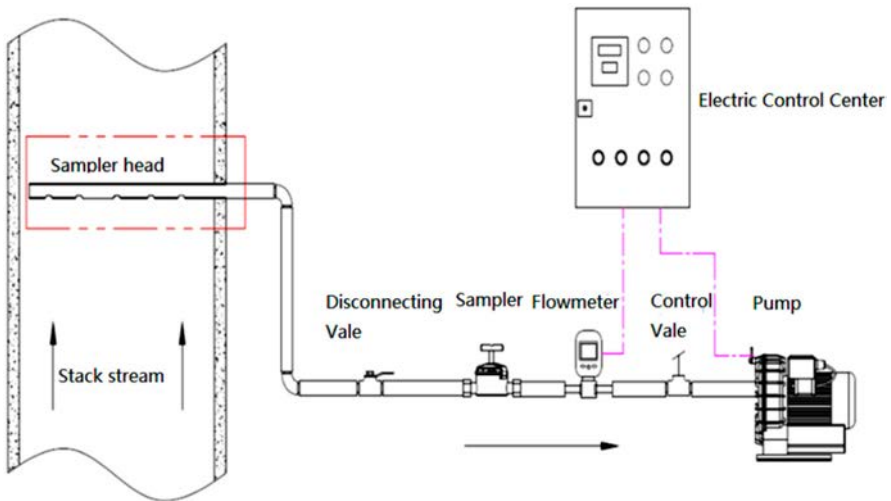


FIG. 2. Sampling system layout.

2.3. Sampling plan and procedures

During the period 15–20 March 2012, a 225 MW(e) coastal coal fired electric utility plant was sampled using the sampling instrument to determine radionuclide concentrations in all major process streams including coal, fly ash and bottom ash. The sampling experiment was done by the China Institute of Atomic Energy and the Radiation Centre of the Ministry of Environmental Protection of China. The sample representivity was considered in the sampling plan. The sampling materials account for a large portion of the radioactivity input and output for the whole plant. Coal accounts for the largest portion of activity input, and the ash in different process steps accounts for a large portion of the activity output. Also, the waste gas effluent from the stack plays a key role in determining the radiation hazard to the people living around the CFPP. The sampling plan was based on the processes within the whole plant (see Fig. 3) and is detailed in Table 1. The physical parameters of the waste gas effluent from the stack were measured using a Laoying 3012H dust collector. The flue gas had a velocity of 11.2 m/s and a flow rate of 644 756 Ndm³/h.

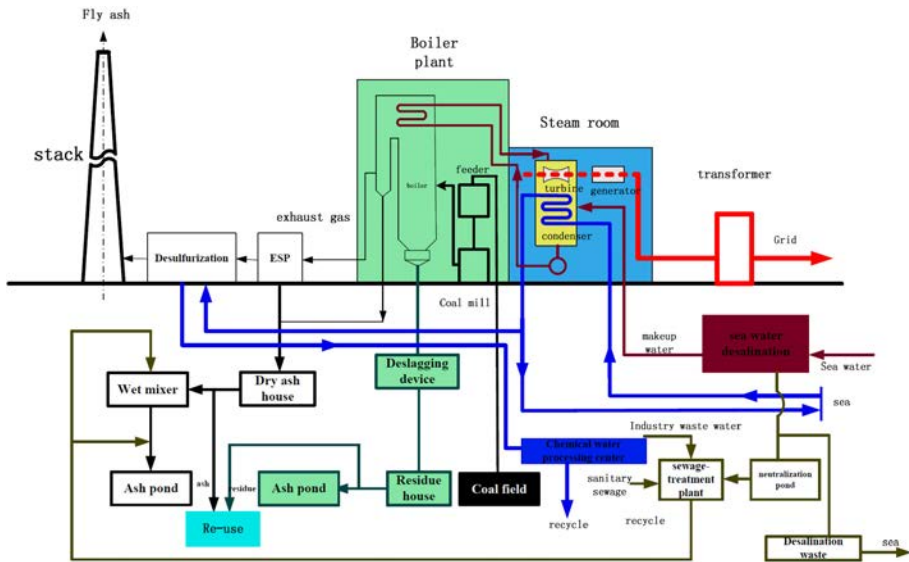


FIG. 3. Processing layout of the plant.

TABLE 1. SAMPLING PLAN

	Collection method	Analyses performed
Coal	Representative sample of each batch of coal	
Bottom ash	3 samples temporally equal in each test	
ESP ash	3 samples temporally equal	U-238, Ra-226, Pb-210, Po-210, Th-232, K-40
Water	3 samples temporally equal	
Desulphurized water	3 samples temporally equal	

3. MEASUREMENT AND ANALYSIS RESULTS

3.1. Stack sampling results

In the sampling test, nine samples were obtained using the designed sampling system. The activity concentrations of the samples were analysed using a Canberra Alpha Analyst. The sample volume was 239 m^3 , the sampling time was 63.1 h and the total ^{210}Po activity was 2.7951 Bq. The coastal CFPP uses three kinds of coal to produce electricity. The radionuclide activity concentrations in the coal for this CFPP were analysed using a gamma spectrometer and an alpha spectrometer. The ^{210}Po activity concentrations were as follows:

- Soft coal: $0.014\ 33 \pm 0.002\ 60 \text{ Bq/g}$;
- Lean coal: $0.032\ 87 \pm 0.004\ 00 \text{ Bq/g}$;
- Lignite: $0.022\ 31 \pm 0.004\ 16 \text{ Bq/g}$.

The electrical power of the sampled CFPP is $2.2 \times 10^5 \text{ kW}$ and the coal usage rate is 100 t/h. According to the average ^{210}Po activity in the coal, the total input of ^{210}Po into the boiler can be determined. The average discharge of ^{210}Po to the atmosphere was 7760.5 Bq/h and the fraction of discharged ^{210}Po to the total input of ^{210}Po was 0.3%, obtained according to the measurement of the coal. The amount of aerosol ^{210}Po emitted from the stack was normalized for the total electric power of the CFPP. It was determined that $2.568\ 2 \times 10^8 \text{ Bq}\cdot\text{GW}^{-1}\cdot\text{a}^{-1}$ of ^{210}Po was released into environment from this CFPP.

3.2. Desulphurization seawater sampling results

It was found that the concentration of ^{210}Po in the desulphurization seawater was an order of magnitude higher than in normal seawater, indicating that part of aerosol ^{210}Po is soluble in water, in line with previous research [4].

3.3. Environmental air sampling results

Besides the samples taken inside the CFPP, the air outside the CFPP was also considered in this paper, so as to investigate the deposition of aerosol particles from the stack in the nearby environment. The results of previous air sampling work conducted in the main wind direction at various distances from the stack of a CFPP [6] indicated that there is a deposition peak for ^{210}Po at a certain distance from the stack. So in order to be able to sample at the point of maximum deposition, a simulation was carried out to locate this point of maximum deposition. The deposition curve was determined using the Gaussian dispersion model by MATLAB and is shown in Fig. 4. The sampled air was analysed and the ^{210}Po concentration is shown in Table 2.

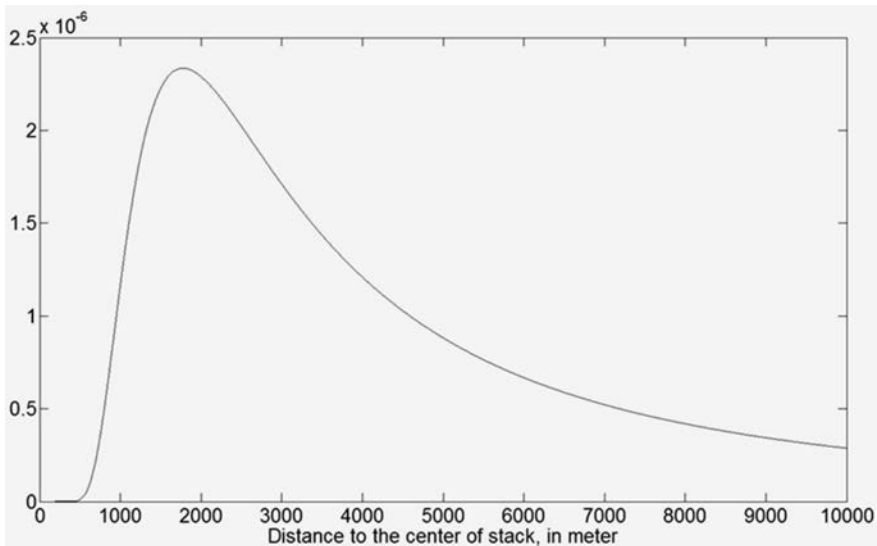


FIG. 4. Theoretical deposition curve.

TABLE 2. ENVIRONMENTAL AIR SAMPLING RESULTS

	Sample volume (m^3)	^{210}Pb activity concentration (mBq/m^3)
Reference location	800	0.061 9
Theoretically determined point of maximum deposition	2 000	0.213 3

3.4. Mass balance study

In order to evaluate the sampling efficiency, a mass balance study was carried out for the ^{210}Po activity distribution throughout the whole combustion process, shown in Fig. 5. Previous studies indicated that the radionuclide mass balance in a CFPP is important for determining the portion of ^{210}Po in each part of the process [4, 7, 8]. The mass balance was determined from the analyses of the coal, bottom ash, electrostatic precipitator (ESP) ash and stack effluent. The following results were obtained:

- Coal: 2 317 000 Bq;
- Bottom ash: 41 500 Bq;
- ESP ash: 2 164 050 Bq;
- Stack flue gas: 7760.5 Bq;
- Difference: 4.4%.

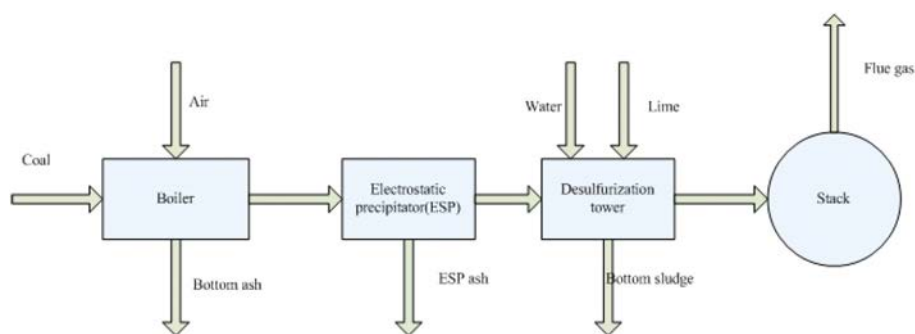


FIG. 5. Combustion process.

3.5. Scanning electron microscopy analysis

A scanning electron microscopy (SEM) image of an aerosol sample from a CFPP stack is shown Fig. 6, indicating clearly the presence of spherical particles. According to previous research, the presence of such spherical aerosol particles can be used to identify a CFPP power plant as the source of emission, since such spherical particles do not occur naturally. Domestic furnaces have to be excluded because glass is viscous above a temperature of 700°C and formable above 1200°C. Domestic furnaces do not reach such high temperatures [6]. So, based upon this philosophy, the study of the morphological features of aerosol particles in the air sample might be used to indicate the original source of radionuclide contamination.

In order to verify whether aerosol particles from the stack of the CFPP can be found in the air outside the CFPP, an SEM image of a sample taken at the reference point was obtained (see Fig. 7). The image shows the presence of spherical aerosol particles, confirming that there is definitely ^{210}Po deposition from the CFPP at the reference point. This finding demonstrates the value of this technique for investigating the radiological hazards from a CFPP to the nearby environment.

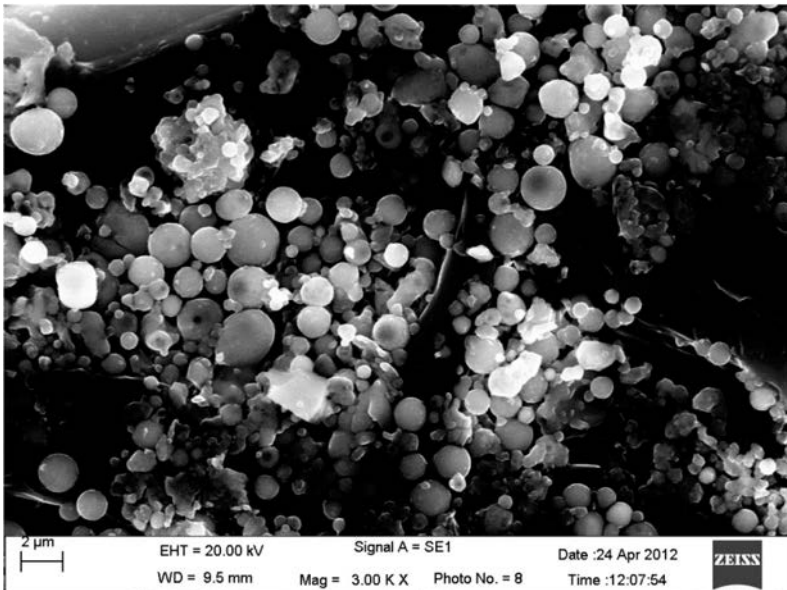


FIG. 6. SEM image of an aerosol sample from a CFPP stack.

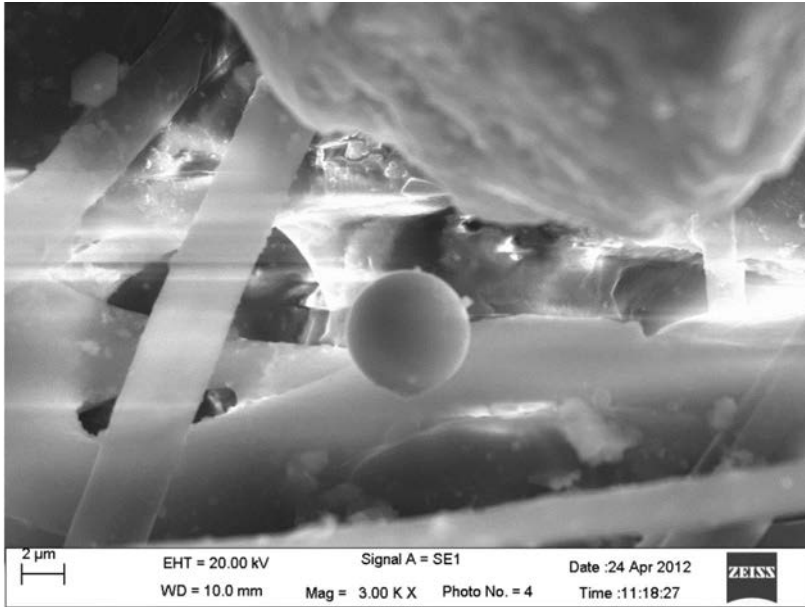


FIG. 7. SEM image of an aerosol sample from the reference point outside the CFPP.

4. CONCLUSION

The designed stack sampling instrument is suitable for sampling the aerosol radionuclides from a CFPP. A 225 MW(e) coastal coal fired electric utility plant was sampled to determine radionuclide concentrations in all major process streams. The ^{210}Po and some other radionuclides were analysed by gamma spectrometry and alpha spectrometry. The amount of aerosol ^{210}Po emitted from the stack was determined and normalized for the total electric power. A measurement technique for measuring ^{210}Po in seawater was also developed. The ^{210}Po concentration in the waste desulphurization seawater was determined and found to be an order of magnitude higher than in normal seawater. A mass balance study for ^{210}Po in the CFPP was determined and the micromorphology of the aerosol ^{210}Po from the stack was analysed using an SEM technique. This technique provides a preliminary method for distinguishing the source of aerosol ^{210}Po in the air around a CFPP.

ACKNOWLEDGMENTS

This paper was sponsored by the China Institute of Atomic Energy and the China National Nuclear Corporation — many thanks to them.

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NORM WASTE FROM OIL EXPLORATION

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Abstract

Exposure to natural sources accounts for the majority of the dose received by a person each year at approximately 80% of the total dose. However, there is a noticeably higher concentration of radioisotopes present in NORM, which is associated directly with human industrial activities. NORM is encountered during the process of mineral mining, including phosphate production, where the end goal is to concentrate large quantities of metals or elements (e.g. phosphorus). However, NORM has also become a widely recognized problem in the oil and gas industry. Enviroklean Product Development Inc., the Nuclear Engineering Teaching Laboratory and Florida Memorial University have been involved in the cleanup and identification of NORM waste in west Texas. An overview of the characterization and technical challenges of properly evaluating the activities of the various types of NORM waste is given. A description of the educational challenges to the training of personnel in the west Texas oilfields handling NORM is also presented.

1. INTRODUCTION

It is approximately 150 years since oil was discovered in the continental United States of America and the mention of radioactivity in mineral oils and natural gases occurred in 1904, just eight years after the discovery of

radioactivity by Henri Becquerel in 1896. In just over three decades, the problems associated with NORM waste arising from the oil and gas industry have been much more intensely scrutinized. In the 1980s, ^{226}Ra began to be noticed when scrap metal dealers would detect unacceptably high levels of radiation from oilfield piping [1]. In 1991, an article was published on the new 'hot' wastes in NORM [2] and in 1992, the health physics aspects of radioactive petroleum piping scale were described [3]. NORM will develop in high concentrations in by-product oil and gas waste streams [4–7]. The NORM will chemically separate from other piped material in the process of the extraction of oil, resulting in high concentrations of ^{226}Ra , ^{228}Ra and ^{210}Pb and other radioisotopes in a densely caked layer on the inner surfaces of the piping [1]. The activity of ^{226}Ra in this NORM ranges from 0.185 to several tens of becquerels per gram of sample. By comparison, the NORM concentrations of radium in rock and soil are, at a natural level, 0.018 5–0.185 Bq/g [1]. Disposal of NORM becomes more problematic as higher concentrations of radioactivity demand even higher degrees of separation from the general populace. Very low levels of NORM can be dispersed along the surface, but higher concentrations require containment in abandoned wells or salt domes [1]. Radon-222, a product of the decay of ^{226}Ra , is also a major component of dose to oil and gas workers from NORM [1]. Radon buildup is particularly hazardous in places where air ventilation is limited, such as underground mining operations. In the oil and gas industry, radon tends to preferentially follow gas lines, and thus is a major concern in the extraction of natural gas where it reaches concentrations of 5–200 000 Bq/m³ [8].

In 1979, the United States Environmental Protection Agency (EPA) set the current rules and guidelines for different radionuclides in drinking water to be at a limit of 0.185 Bq/L. These guidelines were set so that all water systems could meet health standards without creating a financial burden to the towns and counties with the potentially contaminated water arising from NORM. The Texas Commission on Environmental Quality¹ is in charge of monitoring the drinking water systems throughout the state. Both water testing and enforcement of any violations that may occur fall within their mandate. The radionuclides of most interest in Texas drinking water are ^{226}Ra and ^{228}Ra , which emit alpha and beta particles, respectively, along with their associated gamma emissions. Radium is efficiently absorbed into the human body, with intake by way of food and water as a chemical analogue of calcium, and is incorporated into bones. There it can potentially cause an array of health effects including bone sarcoma, leukemia, cancer of the mastoid and paranasal sinuses, cancer of the upper digestive tract and orofacial cleft [9–13].

¹ See www.tceq.state.tx.us.

The EPA has also placed the set rules for contaminated soil to be at 1.11 Bq/g. The obvious concern is that any elevated concentrations in radionuclides in soil may eventually leach into the groundwater. Enviroklean Product Development Inc. is a company in Houston, Texas, that is involved in radioactive waste cleanup of large amounts of residues from the oil and gas exploration industry in west Texas. Due to the absence of uniform legislation regulating oil industry NORM disposal, various methods exist for dealing with it [14–17]. Along with the University of Texas and Florida Memorial University, we have formed a team to better characterize ^{226}Ra , ^{228}Ra and ^{210}Pb and metals in solid NORM wastes. Below is an overview of the analytical work as well as the health physics training given to the workers in the oilfields arising from a close industry–university collaboration in the past three years.

2. ANALYTICAL DETERMINATION $^{226,228}\text{Ra}$, ^{210}Pb AND METALS IN NORM RESIDUE SAMPLES

NORM samples from oil and gas exploration contain a complex matrix as a result of the precipitation of radium, barium, calcium and strontium sulphates. Barium, calcium and strontium are in Group II of the periodic table, thus they have very similar chemical characteristics. Thus the determination of low energy photons of ^{226}Ra (186 keV) and ^{210}Pb (45.6 keV) may be severely compromised due to self-attenuation of the gamma radiation in the sample. This is especially true if the samples are gamma counted in large volumes such as Marinelli containers which may hold up to 250–500 g of material. To overcome this problem, we used Petri dishes containing 20 g of material.

In our experiments, we used a 37 000 Bq calibrated water based solution containing isotopes from 46 to 1836 keV to derive a standard efficiency curve. A program called SELFABS [18] was used to estimate the attenuation in the water and soil. For soil, the estimated concentrations of elements above 0.1% were based on published crustal earth data [19]. The transmission factors were as follows: 45.2% for the 46.5 keV photon belonging to ^{210}Pb ; 76.8% for the 186.2 keV photon belonging to ^{226}Ra ; and 87.3% for the 911 keV photon of ^{228}Ac , which is in secular equilibrium with ^{228}Ra . All samples were heat sealed and allowed to stand for one month to achieve secular equilibrium, were counted for 12 h on a hyperpure germanium detector with an efficiency of 28% and an FWHM (full width at half maximum) resolution of 2.0 keV for the 1332.4 keV photon belonging to ^{60}Co . The radioactivity results for soil, scale, sludge and processed water samples are shown in Table 1.

TABLE 1. RADIOACTIVITY RESULTS FOR ^{210}Pb , ^{226}Ra AND ^{228}Ra

	Activity concentration (Bq/g)			
	Sludge	Scale	Soil	Water
Pb-210	5.148 ± 0.222	1.370 ± 0.074	22.889 ± 0.74	0.815 ± 0.037
Ra-226	59.00 ± 0.300	2.630 ± 0.148	65.296 ± 3.296	1.481 ± 0.074
Ac-228 (Ra-228)	28.501 ± 1.493	0.565 ± 0.040	0.154 ± 0.012	0.251 ± 0.016

A quality assurance check on the efficiency curve was performed by analysing the IAEA certified reference material IAEA-375 Radionuclides and Trace Elements in Soil. Our result of 5.024 ± 0.163 Bq/g for ^{137}Cs (with all uncertainties propagated) is in good agreement with the consensus value of 5.280 ± 0.080 Bq/g, which is $\sim 5\%$ higher. A simple sensitivity analysis revealed that the dominant factor for any variation in a result is the calculation of the calculated transmission factor (or self-absorption) of photons. The analytical uncertainties based on counting statistics for ^{210}Pb , ^{226}Ra and ^{228}Ra for the sludge, soil and scale varied between 0.1 and 6%. Detection limits for ^{210}Pb , ^{226}Ra and ^{228}Ra varied depending on the type of matrix that was determined. However, in all four cases there were adequate statistics to achieve good precision. Two additional interferences were taken into consideration. One is the interference of the 185.2 keV gamma ray of ^{235}U on the 186.2 keV of ^{226}Ra . This interference is well known and explained in detail in Ref. [20]. However, in these NORM samples the activity of ^{235}U is from one to several orders of magnitude lower than that of ^{226}Ra , thus having a negligible interference effect. The second systematic problem is coincidence summing. Again, this is a well known effect, and for natural radioactivity these corrections have been tabulated for Marinelli and Petri holders [21]. For ^{210}Pb and ^{226}Ra , the correction factors are negligible, but for the 911 keV photon for ^{228}Ac used to determine ^{228}Ra , there is a 9% effect. The results in Table 1 reflect this correction factor. More recently, we have used a more standard way of determining the self-attenuation by using a ^{152}Eu source and placing it on top of the Petri dish with and without the NORM sample. Once an attenuation curve is plotted, self-absorption factors for photons for any sample less or more dense can be easily calculated by normalizing the original plot with just one photon. Results for ^{210}Pb , ^{226}Ra and ^{228}Ra are shown in Table 1.

3. NEUTRON ACTIVATION ANALYSIS

3.1. Sample collection and preparation

Soil, solids and piping scale samples were collected from an oil producing facility in western Texas and homogenized using a 250 µm sieve and vacuum dried for 2 h in a 100°C oven. Each sample was weighed to approximately 0.5 g and placed in a polyethylene vial for neutron irradiation.

3.2. Methods

A complete description of the methods of thermal and epithermal neutron activation analysis in conjunction with Compton suppression used in this work is well described in previous work [22, 23]. Ten samples each were prepared for thermal and epithermal neutron activation analysis (NAA). Each set of ten contained two solids samples, two soil samples, two scale samples, and four National Institute of Standards and Technology (NIST) reference materials, including coal (NIST 1632c), two coal fly ash (NIST 1633a and NIST 1633b), and San Joaquin soil (NIST 2709). It was quickly recognized that for short lived NAA, the reactor power levels needed to be significantly reduced by an order of magnitude because of the very high dead times arising from some of the radionuclides. A complete description of the various irradiation, decay and counting times is seen in Table 2.

TABLE 2. IRRADIATION, DECAY AND COUNTING TIME CHARACTERISTICS FOR THE DIFFERENT ELEMENTS

	Count type	Type of irradiation	Irradiation time	Decay time	Count time
Sc, Fe, Co	Normal	Thermal (950 kW)	1 h	2 weeks	2 h
Zn, Cr, Ni	Compton	Thermal (950 kW)	1 h	2 weeks	2 h
Na, Cl, Al, Mn, Ca, V	Normal	Thermal (100 kW)	30 s	10 min	10 min
U, Br, Si, Ba, Sr, I, In	Compton	Epithermal (100 kW)	10–60 s	5–10 min	20–30 min

3.3. Flux measurements

To control the variation in flux measurements in short lived NAA, which can vary up to 7%, aluminum wires were irradiated with each sample. A description of this methodology is presented elsewhere [24].

3.4. Calibration and quality control

Calibration was mostly done using NIST 2709 San Joaquin Soil as the primary standard with the exception of uranium, bromine, chlorine and indium concentrations which were taken from NIST 1633a, NIST 1632c, NIST 1632c and a liquid standard, respectively. Once a calibration library was set up, the same NIST samples were used as unknowns. In general, all the NAA results agreed to within 3–10% of the certified or information NIST values.

3.5. Results and discussion

The results for 19 elements (aluminium, barium, bromine, calcium, chlorine, chromium, cobalt, indium, iodine, iron, manganese, nickel, scandium, silicon, sodium, strontium, uranium, vanadium and zinc) for scale, solid and soil samples are shown in Table 3.

It can be seen that the elements in the same group as radium have elevated amounts of barium, calcium and strontium for the scale, soil and solid samples. Surprisingly, there are also elevated amounts of indium in the scale and solid samples. This is most probably due to the indium activated aluminum anodes used in the separation of crude oil from the brackish water. Because of the low solubility of the uranium, as compared with that of ^{226}Ra , the concentrations were very low or not detected at all. The other striking feature of the samples was the very high amounts of iron. In the case of scale, 25% of the elemental composition was made up of iron. This may be a result of the corrosion of the holding tanks for these products. When compared with concentrations in soil, it would also appear that zinc, manganese, bromine, chlorine, sodium and iodine are also very elevated. In the case of bromine, chlorine, sodium and iodine, this may be a result of the brackish water or seawater component found in oil in west Texas. The results shown in Table 4 reveal that chlorine:sodium and bromine:chlorine ratios agreed reasonably well with those ratios in seawater. However, the iodine:chlorine ratio was about two orders of magnitude greater than the ratio found in seawater. It would be hard to speculate on the reason for this large difference other than that the mobility of iodine in an oil environment may be significantly enhanced.

NORM WASTE FROM OIL EXPLORATION

TABLE 3. ELEMENT CONCENTRATIONS

	Concentration		
	Scale	Soil	Solid
Al (%)	0.53 ± 0.02	1.88 ± 0.02	0.46 ± 0.01
Ba (µg/g)	8 936 ± 717	34 435 ± 2 721	177 121 ± 1 000
Br (µg/g)	223± 11	2.9 ± 0.6	48 ± 3
Ca (%)	11.2 ± 0.7	2.4 ± 0.2	1.5 ± 0.1
Cl (%)	6.41 ± 0.48	0.04 ± 0.01	0.71 ± 0.05
Cr (µg/g)	53 ± 2	13.7 ± 0.7	131 ± 5
Co (µg/g)	14.6 ± 0.6	1.8 ± 0.1	2.8 ± 0.2
In (µg/g)	1.64 ± 0.08	Not detected	0.84 ± 0.05
I (µg/g)	31.9 ± 2.5	3.2 ± 0.3	10.5 ± 0.9
Fe (%)	24.9 ± 0.5	0.94 ± 0.02	15.7 ± 0.3
Mn (µg/g)	1 053 ± 38	80 ± 4	207 ± 9
Ni (µg/g)	54 ± 9	11.7 ± 2.5	73 ± 9
Sc (µg/g)	0.21± 0.02	1.81 ± 0.03	0.12 ± 0.01
Si (%)	1.08 ± 0.14	39.0 ± 1.3	2.40 ± 0.13
Na (%)	3.24 ± 0.09	0.14 ± 0.01	0.85 ± 0.03
Sr (µg/g)	12 484 ± 1389	4 142 ± 464	48 288 ± 5 362
U (µg/g)	0.08 ± 0.02	0.80 ± 0.03	Not detected
V (µg/g)	Not detected	21.8 ± 1.5	5.2 ± 0.7
Zn (µg/g)	288 ± 8	50 ± 2	683 ± 15

TABLE 4. HALOGEN RATIOS IN A SCALE SAMPLE

	Scale	Seawater, for comparison
Cl/Na	1.97	1.8
Br/Cl	3.5×10^{-3}	6.2×10^{-3}
I/Cl	5×10^{-4}	3.3×10^{-6}

4. NORM AWARENESS TRAINING

The training is not only limited to employees directly dealing with the NORM or in the field. It is also beneficial for employees in management positions to have training in health and safety and current regulations. It is a valuable resource for other companies in the oil and gas field to have their employees trained in NORM awareness so that they understand the risks and regulations of working with this part of the oil and gas industry. These other companies may then train other field or environmental workers as well as executives. The class needs to comprise a general introduction to radiation as well as worker safety and health concerns. This includes going back to the fundamentals of general chemistry, including explaining what an atom and its components are. Taking the extra time and care to go over the basics will help provide benefits in both health and safety and avoid any potential violations. Next, it is important to determine where the radiation in the oil and gas field comes from using decay series and visual aids. A general description of where NORM collects during the process is also useful.

One of the most important aspects in the training course is worker safety. This includes but is not limited to on-the-job site safety, emergency preparedness and personal protective equipment (PPE). The PPE varies depending on what the particular job entails and can include fire retardant clothing, gloves, safety goggles, mask, steel toed boots, a thermoluminescent dosimeter badge, a hydrogen sulphide (H₂S) monitor, a hard hat and self-contained breathing apparatus. Stressing the importance of PPE is not only vital to the safety of the workers but important for adhering to state and national guidelines. This part of the training should ideally be ongoing and emphasized everyday by the radiation safety officer (RSO) of the job site. For example, forgetting to wear an H₂S monitor can lead to potentially serious health effects which can easily be prevented. It is mandatory for the onsite RSO to go over the safety guidelines for the specific job site each day. Greater precautions should be exercised

with regard to closed container operations. The radionuclides, especially ^{222}Rn , tend to build up in the lower levels of the tanks or containers where a self-contained breathing apparatus is always necessary. The class also highlights the importance of monitoring for low levels of radiation each time someone leaves the restricted area. This method, called ‘frisking out’, is when the RSO on duty runs a pancake probe Geiger–Müller (GM) counter over the person’s body, focusing especially on the hands. If the readings are above background, the employee is required to wash the affected area and change clothes if necessary. This procedure is outlined with great importance in order to prevent accidental ingestion of radionuclides from the job site. Another imperative section of the training is the education pertaining to the health effects of NORM. Since all high doses of radionuclides can potentially cause damage, the class focuses on the four radionuclides that pose the most concern in the field: ^{226}Ra , ^{222}Rn , ^{228}Ra and ^{210}Pb . Emphasis is also placed on how NORM is indistinguishable from non-radioactive material. Providing pictures of scale and its buildup on pipes helps reinforce proper safety when dealing with these radionuclides. Explaining how radiation enters the human body is also a key point. Using a chart to describe the simplistic interactions between the gamma rays, alpha and beta particles is also advantageous. This supports the point that the highest risk of working with NORM comes from ingesting the radionuclides containing alpha and beta particles and helps highlight the necessity of using gloves and frisking out when working around this material.

The class ends with a review of the important facts followed by an examination of the material learned. After passing the test, employees can work with NORM knowing the proper procedures and protocols required for its handling. For new workers, it is important to stress the key facts learned during the course to ensure proper safety procedures. A refresher course for employees that have worked with NORM for a number of years is beneficial, since it is easy to become complacent and forget the safety aspects of NORM and its associated health effects. An overview of the implementation of these rules and regulations is depicted in Fig. 1.

4.1. Training different levels of education

A big hurdle in training for NORM in the oil and gas field in Texas is the wide array of academic backgrounds that one may come across among those taking the course. Often, the knowledge of NORM or radiation is limited. This then necessitates spending more time on the basics of radiation or even chemical science before the employees are able to properly understand the meaning of radiation. Taking the time to teach the individual class members the fundamentals helps them immensely to understand the class material. It is useful to show how a

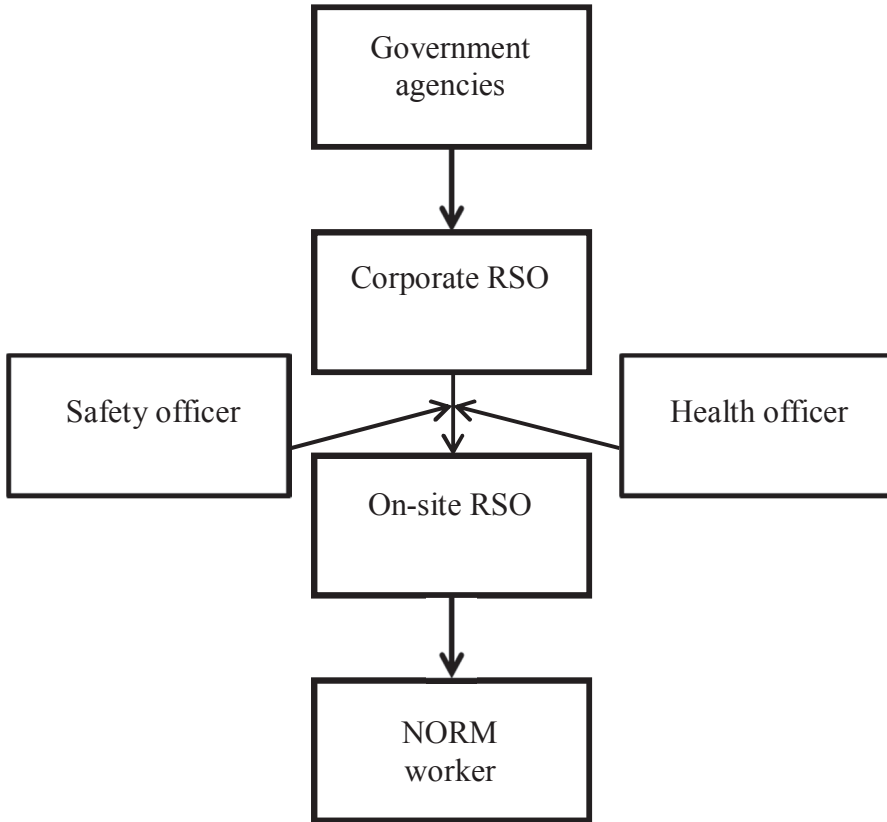


FIG. 1. Implementation of rules and regulations for NORM workers.

typical person interacts with low levels of radiation in their everyday life and that working with radioactive materials does not have to be a hazard, providing proper precautions are closely followed. The population of Spanish speaking residents is growing in Texas. The need to train employees who are not fluent in English is becoming more common. During these classes, it is important to make sure that everyone follows the instructions. This is best accomplished by ensuring that there are bilingual people in the class who can help with translation. The training of employees with a non-scientific background also entails that they understand the importance of taking samples and correctly labelling them for further off-site analysis. Receiving samples that are improperly labelled or not labelled at all can be a major problem especially when dealing with radionuclides such as ^{222}Rn that have a short half-life. In order to get an accurate reading of radionuclides, it is important that the air samples be labelled with the date they were taken and

length of sampling time. This portion of the training needs to be refreshed at least once a month to ensure that the employees adhere to proper sample protocols.

4.2. On-the-job site monitoring and training

Each job site needs to be monitored at all times because of the nature of the NORM that is encountered during decontamination and is having to be cleaned up. On-the-job training may be necessary if a new or different situation is encountered. If this is the case, it is the responsibility of the RSO on the job site to help train and monitor the site. There may be a requirement for a portion of the actual job training to take place on site to better inform the employees. Hands-on training in addition to in-class instruction helps to reinforce the material being taught. Training workers on NORM is valuable for the health and safety of the employees in the oil and gas industry and for the general public. Refreshing all employees on the subjects they have learned will help create a safer work environment and minimize accidents. It is important to update and make changes to the ongoing training as new regulations and guidelines are put into effect. With the training course and proper on site job monitoring, the risks of working with NORM are greatly reduced, making it a safe working environment for all employees.

5. CONCLUSIONS

Our collaboration with the industry on NORM in oil exploration has been very fruitful. We have implemented analytical protocols in determining radionuclides in the waste products and determining self-attenuation properties for samples that exhibit unusually high atomic number elements such as iron, barium and strontium. We have established a protocol to determine up to 19 elements in the NORM samples using neutron activation analysis. Equally important has been the putting into practice of various training modules and health safety practices.

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NORM AND MINERAL DEVELOPMENTS IN THE NORTHERN TERRITORY OF AUSTRALIA

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Abstract

The Northern Territory (NT) of Australia has a long history of involvement with NORM, primarily through an association with uranium mining that goes back over 50 years. The paper describes this history briefly, including the more modern developments in uranium mining including the operating Ranger Mine and the recently closed and remediating Nabarlek mine. However, in the past few years there has been an expanded interest in uranium and other deposits that have a NORM aspect. Current projects relating to oil and gas, uranium, rare earths, phosphates and mineral sands, as well as some metalliferous deposits, are at various stages of development from early exploration to development and implementation. The paper discusses these various activities from a regulatory viewpoint, in particular how the supervising authorities cooperate and implement the various aspects of NORM regulation in the NT.

1. INTRODUCTION

The mining and/or processing of any natural material or mineral will almost always have the potential to increase the exposure to radiation of the workforce or the community as the raw materials all contain radionuclides of natural origin. While only a few of these situations will require the introduction of radiation protection plans, it is important that the risk is assessed in order to ensure the safety of individuals and the environment as well as enabling the appropriate level of regulatory supervision to be applied. In 2006, the IAEA published Safety Reports Series No. 49, Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials [1], in which the whole issue was discussed. The report also listed those industries and activities, apart from uranium mining, where the matters of NORM and subsequent risk of radiation exposure were most likely to be encountered. The economy of the Northern Territory (NT) of Australia is based in a significant part on the minerals sector and thus the issue of NORM needs to be taken into account by the various authorities and agencies responsible for regulating the industry.

2. NORM INDUSTRIES IN THE NORTHERN TERRITORY TODAY

The NT has a significant history of association with NORM which stems mostly from the uranium mining industry. However, in recent years other NORM resources have been investigated and for which, in some cases, development plans are being prepared. The range of NORM projects under consideration includes rare earths, mineral sands and phosphate deposits as well as ongoing activities related to all parts of the uranium production cycle. The major elements of the uranium related activities are described most recently in Ref. [2] and so will not be discussed at length here.

2.1. Oil and gas

The NT has an active and developing oil and gas industry with three active oil fields in the southern NT at Palm Valley and East and West Mereenie with three more not yet operational. In addition, there are offshore oil and gas production fields which use Darwin as a supply base as well as the operating liquefied natural gas (LNG) plant at Wickham Point and the new LNG plant presently under construction for the Icthus project at Blaydin Point, both located on Darwin harbour. Offshore oil and gas activity is regulated by the Australian Government.

All of these developments and projects have the potential to produce NORM related exposures, primarily arising from the cleaning of pipes and tanks involved in the pumping of associated or formation waters. These potential exposures are managed in accordance with the requirements of the regulating authorities and taking heed of the guidance provided by the IAEA in Safety Report Series No. 34, Radiation Protection and the Management of Radioactive Waste in the Oil and Gas Industry [3]. The radiation protection issues are regulated through the Radiation Protection Act, which is administered by the Department of Health.

2.2. Rare earths

The NT has a good prospectivity for rare earth deposits and extensive exploration activities are taking place in several areas. The most advanced prospects include the Nolans Bore deposits, being developed by Arafura Resources Limited, and the Charley Creek deposits being evaluated by Crossland Uranium Mines Limited. The NT Department of Mines and Energy (DME) is responsible for regulating mines through the Mining Management Act (MMA)

and refers all operators, be they explorers or miners, to Safety Reports Series No. 68, Radiation Protection and NORM Residue Management in the Production of Rare Earths from Thorium Containing Minerals [4], when considering the preparation of their Mining Management Plan (MMP) and the included Radiation Management Plan (RMP).

Published resource information for the Nolans Bore deposit is shown in Table 1, indicating that there may be possibilities for by- and co-production of other materials. However, much of that will depend on market conditions for the respective materials in the future. The current plan is for the project to produce a beneficiated concentrate on site, about 500 000 t annually, which would then be transported to the company’s rare earths complex which will be built at Whyalla, in South Australia. This will require transport permits and appropriate licences for authorities in both the NT and South Australia. One further point to note is that the residues from the plant at Whyalla will be returned to the mine site at Nolans Bore for disposal in containment structures specifically built for that purpose. These residues are likely to be classified as radioactive since they will contain essentially all the thorium, and uranium if it is not recovered, as well as other possible radionuclides not recovered. The project hopes to recover 848 000 t of rare earths (lanthanum, cerium, praseodymium, neodymium, samarium, europium, gadolinium, terbium, dysprosium and yttrium). In addition, the deposit contains 3.9 million t of phosphorus pentoxide (P₂O₅) and 13.3 million lb (6045 t) of uranium oxide. The mining operation would be regulated under the MMA, which will require an annual MMP to be submitted for approval and that MMP will be required to include an RMP.

TABLE 1. NOLANS BORE RESOURCE CLASSIFICATION

	Rare earth oxides			P ₂ O ₅ grade (%)	U ₃ O ₈ grade (lb/t)
	Ore (Mt)	Grade (%)	Product (kt)		
Measured	4.3	3.3	144	13	0.57
Indicated	21	2.6	563	12	0.42
Inferred	22	2.4	511	10	0.37
Total	47	2.6	1217	11	0.41

Source: <http://www.arafuraresources.com.au>.

The Charley Creek project is being investigated by Crossland Uranium Limited in partnership with Pancontinental Uranium Corporation of Canada with a scoping study currently being completed. The project was originally exploring for uranium, but it became apparent that the deposit contains significant amounts of rare earth oxides and is thought to contain 17% of the heavier, and more keenly sought after, elements amongst the rare earths, rather more than is usual. The other interesting point is that the deposit appears to contain low levels of radionuclides associated with the rare earths and it is thought unlikely that there will be any significant radiation protection issues.

2.3. Mineral sands

The NT has only one minerals sands project which is operated by MZI Resources Limited. The company has interests elsewhere in Western Australia and is on the point of completing operations at the Lethbridge South site in the Tiwi islands which are in the Arafura Sea just to the north of Darwin. MZI has operated in the Tiwi islands since 2009 and has produced some 33 000 t of heavy mineral concentrates, which were sold mostly to China. The mines in the Lethbridge series on Melville Island are due to end operations in 2013 but the approvals process for a new mine at Kilimiraka on Bathurst Island has begun with a full feasibility study expected to be completed in 2014. Recent statements by the company suggest an inferred resource of 56.2 million t grading at 1.6% heavy minerals and an anticipated heavy mineral content of 894 000 t. The process is a standard excavation and wet separation in spirals; and to date, there have been no significant radiation protection issues. However, the operator is aware of the potential for such issues to arise and appropriate measures are in place. In early 2013, the company submitted a notice of intent for the new project at Kilimiraka and the environment impact assessment process was set in train by the NT Environmental Protection Authority in March 2013 with the decision on the required level of assessment due to be handed down before the middle of the year.

2.4. Phosphates

Phosphate deposits are being investigated at a number of locations within the NT. The evidence has been that all the deposits identified to date have been found to be low in natural radionuclides and radiation protection issues have been minimal. One deposit is the Amaroo project being developed by Rum Jungle Resources Limited where the total of measured, indicated and inferred resources has been reported as 238 million t with a cut-off at 10% P₂O₅. Throughout the

deposit, the uranium levels are reported as being between 19 and 31 ppm, which is why the radiation risk is deemed to be low.

Minemakers Limited are the owners of Australia's largest undeveloped rock phosphate deposit which has 78 million t of measured resources at 20.8% P₂O₅, and inferred and indicated resources of 933 million t at 13% P₂O₅ with a 10% cut-off. The ore body contains uranium at an average grade of 18 ppm although the maximum recorded concentration was 75 ppm. Work carried out by the company as part of the preparation of the environmental impact statement have indicated that the levels of uranium are not sufficient to warrant further investigation as the ore will be neither beneficiated nor concentrated. Thus, the product and the residues are not considered to present a radiological risk for the proposed project with direct shipping of ore.

However, for both these projects the possibility exists that there may be a change from an emphasis on direct shipping of crushed ore to some form of processing which is likely to involve phosphoric acid production and possibly even further downstream processing to fertilizers or similar products. Such developments would require a reassessment of the radiological implications, especially in relation to NORM scales building up in pipework and reaction vessels, among other things. This may be of particular interest in the case of Minemaker's project where they have recently announced (March 2013) the possibility of having a slurry pipeline from the mine to the town of Tennant Creek, a distance of about 240 km, at an estimated cost of \$500 million. This project could also include a fertilizer plant which would also need to be assessed for radiological risks.

2.5. Other mining

One issue that has to be considered when looking at the list of NORM related industries in Safety Report Series No. 49 [1] is the possibility of radiation protection issues arising in mining operations other than uranium or the industries described above. Much of the mineralization that exists in relation to uranium, for example, is often associated with other valuable deposits. The uranium mine at Guratba (Coronation Hill) and the other South Alligator Valley deposits also produced gold in the 1960s [5]; these sites were also explored in the 1990s, when a gold, platinum and palladium mineralization was identified at a number of locations. However, the sites are all located in an area that became stage 3 of the World Heritage listed Kakadu National park and no further mining activity will be permitted. The site at Rum Jungle was primarily a copper mine that also produced uranium (p.142 of Ref. [6]); in fact, several of the uranium related radiological anomalies in and around that district are now areas where gold mining has either taken place or is still in progress. For this reason, all underground mines

in the Pine Greek geosyncline are regarded as having the potential for exposing workers to doses in excess of 1 mSv/a and the DME recommends and advises that operators carry out suitable monitoring and screening programmes and then discuss the results with the Department of Health where appropriate. To date, there have been no results of significance reported, although the presence of uranium in some drainage waters indicates that the matter cannot be ignored and ongoing surveillance is required. The DME's Environmental Monitoring Unit checks some legacy sites as well as active mines which are also required to submit monitoring data to the DME periodically. Any anomalous results, usually in water chemistry, are discussed with the operators, where applicable. Such data may also be referred to the Department of Health and/or the NT Worksafe authority as appropriate.

3. CONCLUSION

The NT has a long and strong association with the mining industry that continues to the present time. Many of the minerals mined in earlier times contained NORM, although the risks associated with these substances were not always recognized. More recently, the NORM industry in the NT has been dominated by uranium mining where world class leading practices in radiation protection are a regular part of operations. As the world has become more interested in rare earth elements and the demand for fertilizers has grown, so have suitable mineral deposits related to these commodities been discovered in the NT. Most of these deposits are associated with NORM minerals that occur in varying concentrations. Several of these deposits are likely to be developed in the near to medium future and the DME is ensuring that the potential risks associated with NORM are clearly explained to operators who in turn are adopting suitable monitoring programmes and radiation protection plans where appropriate.

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CONFINEMENT AS A SUSTAINABLE ROUTE TO PHOSPHOGYPSUM MANAGEMENT IN THE TAPARURA ZONE

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Abstract

The phosphogypsum (PG) management policy implemented within the Taparura project at Sfax, Tunisia, allowed a reduction of the impact of this residue by finding better waste management procedures with an extremely limited radiological impact. The PG samples used in this study came from a fertilizer factory in Sfax, Tunisia, and were compared with samples from Huelva, Spain. The paper reviews the impacts associated with the storage and disposal of PG and the confinement procedure carried out to minimize the negative effects of this residue and compares results with those obtained from a similar investigation carried out on Spanish PG. The semiquantitative chemical composition of the PG samples was identified. Mineral species were determined by X ray diffraction. The morphological aspects of the PG were analysed using scanning electron microscopy with energy dispersive spectroscopy. Selected uranium and thorium decay series radionuclides and ^{40}K present in the PG samples have been quantified. The PG samples from Tunisia have low activity levels compared with those in PG samples from Huelva, Spain. In both cases, the values are below the activity concentration screening levels recommended by the European Commission for use in common building materials and industrial by-products used for building. Measurements made in the environment surrounding the Taparura site after completion of all excavation and confinement work show no radiological effects. The gamma dose rates were at natural background levels.

1. INTRODUCTION

Phosphogypsum (PG) is a residue from the processing of phosphate rock by the 'wet acid' method of fertilizer production, which currently accounts for over 90% of phosphoric acid production. Huge quantities of PG are generated (about 5 t per tonne of phosphoric acid production), and worldwide PG generation is estimated to be around 280 million t/a. This residue has applications as a by-product, but most of it is deposited in stockpiles without any treatment.

These stockpiles are generally located in coastal areas close to phosphoric acid plants, where they occupy large land areas and cause serious environmental damage. PG is mainly composed of gypsum but also contains significant levels of impurities such as phosphates, fluorides, sulphates, heavy metals, other trace elements and radionuclides of natural origin.

This paper describes an important PG deposit remediation project — the Taparura project — initiated in 2006. It is a large coastal project associated with PG containment by means of confinement and development into an urban park. The aim of this project is to address the environmental pollution of the northern coastal area at Sfax, the second city of Tunisia. The area has undergone significant urbanization, with a strong industrial presence. The remediation of the area will allow the reconciliation of the city with its beach areas by reclaiming 420 ha of land from the sea to allow for the extension of the urban centre.

1.1. Genesis of the Taparura project

Known for its entrepreneurial spirit, its economic dynamicism and its agricultural and energy production, the region of Sfax is the second pole of activity in Tunisia. A few decades ago, the northern coastline of Sfax comprised a series of clean beaches that were the most prominent destination of the city dwellers. The installation of the chemical plant and the industrial zone along the coast close to the city centre resulted in pollution of the marine environment and the degradation of the area due to various industrial discharges, mainly PG from the phosphate treatment industry.

The aim of the Taparura project was to allow reconciliation of the city with its beaches and marine ecosystems. Its overall objective is to reclaim and rehabilitate 6 km of beaches. The realization of the Taparura project is divided into two steps:

- Step 1: Cleanup of the northern coastline of the city of Sfax and reclamation of 420 ha land from the sea;
- Step 2: Development of the remediated area and creation of a new urban centre.

The extent of the project is shown in Fig. 1.



FIG. 1. Works zone of the Taparura project.

1.2. PG confinement procedure

The phase of remediation completed in May 2009 is illustrated in Fig. 2 and consisted of:

- (a) Excavation of the PG layer which surrounds the deposit;
- (b) Removal of contaminated materials from the land and the sea;
- (c) Remodelling of the PG deposit into a double eccentric truncated cone and its development into a 60 ha urban park;
- (d) Isolation of the PG deposit by a radial screen consisting of concrete and high density polyethylene sheeting anchored in a tight hydrogeological barrier formed by a bed of subjacent clay 12 m below ground;
- (e) Installation of a pumping system comprising extraction wells under the bed of subjacent clay and a drainage system for shallow subsoil water and surface water.



FIG. 2. Design of the remodelled PG deposit and radial screen anchoring work.

1.3. Pumping system

The role of the pumping system is to prevent the migration of polluted water to the surrounding area. The pumping system is designed such that the underground water reaches and sustains a certain level inside the screen about 0.25 m lower than the underground water outside the screen. Due to the effect of the tide on the groundwater outside the screen, the difference between the water table inside and outside the screen is not constant. Based on an on-site investigation and pumping test, the pumping system, with extraction wells inside the screen, was developed using the Visual MODFLOW hydrogeological model.

The installation of the screen allows the effect of the tide on the groundwater inside the screen to be controlled. Taking into account the balance of the groundwater inside the screen, two water flows are considered: the first is the groundwater beneath the deposit, and the second is rainwater. Rainwater is collected and evacuated to the outside of the deposit. The prevention of

infiltration of rainwater into the PG in this way avoids contamination of the water and any increase in water level inside the screen.

2. MATERIALS AND METHODS

The PG samples used in this study came from a fertilizer plant in Sfax. The semiquantitative chemical composition of the PG samples was determined using an X ray fluorescence analyser (Philips model PW-1404 sequential wavelength dispersion unit). Mineral species were determined by X ray diffraction (Siemens model D5000, with a copper tube and lithium fluoride monochromator). The morphological aspect of the PG was analysed using scanning electron microscopy (SEM) (Joel model JXA-840) with energy dispersive spectroscopy. Uranium and thorium decay series radionuclides and ^{40}K present in the PG samples were quantified as follows [1]:

- (1) The uranium content in the samples was determined using two different methods: (a) direct measurements by gamma spectrometry with high purity germanium detectors; and (b) laser induced kinetic phosphorimetry. The direct measurements were carried out on 700 g aliquots of the samples packed in standard Marinelli beakers. The ^{238}U activity concentration was determined through the photopeaks of its immediate decay product ^{234}Th (63 and 92.5 keV), whereas ^{235}U was measured directly from its 143.8 and 163.4 keV gamma peaks. Concerning the laser induced kinetic phosphorimetry technique, 1 g of the sample was completely digested in 15.6 mol/L nitric acid (HNO_3) and the measurements were performed using a kinetic phosphorescence analyser (KPA-11) (Chemcheck Instruments Inc., Richland, WA) [2]. In order to compare the results obtained by both techniques, the total uranium concentration obtained by laser induced kinetic phosphorimetry, expressed in $\mu\text{g/g}$, was then converted to the activity concentration of each uranium isotope. Theoretical values of the isotopic composition of natural uranium (99.3% ^{238}U , 0.72% ^{235}U and 0.0055% ^{234}U) and the specific activities of these isotopes in natural uranium (Bq/g) were used for this purpose [3].
- (2) The polonium activity concentration in the samples was determined by alpha spectrometry, by means of a ^{210}Po separation procedure [4]. An aliquot of 1 g was digested on a hot plate at a controlled temperature ($<90^\circ\text{C}$), using 8 mol/L HNO_3 . Polonium-209 standard dissolution was added to the dissolved samples as a tracer to estimate the recovery of the whole process. The polonium isotopes were self-deposited on silver discs in accordance with Flynn's method [5].

- (3) The concentrations of ^{226}Ra , ^{232}Th , ^{210}Pb and ^{40}K were quantified by gamma spectrometry analysis using a high purity germanium detector. The detector was shielded from external radiation by a 15 cm thick iron wall. The emission gamma spectrum was analysed using Genie-2000 application software. To ensure radioactive equilibrium between ^{226}Ra and its short lived decay products, the samples (700 g aliquots) were packed in standard Marinelli beakers, hermetically sealed and stored for about four weeks prior to counting. The concentrations of ^{226}Ra and ^{232}Th were estimated from the gamma photopeaks of their progeny ^{214}Bi (609 keV) and ^{228}Ac (911.2 keV, 969.0 keV), respectively. The concentrations of ^{210}Pb and ^{40}K were measured directly from their gamma emissions at 46.5 keV and 1460.8 keV, respectively. The minimum detectable activity limit (DL) was also calculated.

3. RESULTS AND DISCUSSION

3.1. Phosphogypsum characterization

The chemical composition of both types of PG sample is summarized in Table 1. The data show that sulphate (expressed as SO_3), calcium oxide (CaO), silicon dioxide (SiO_2) and phosphorus pentoxide (P_2O_5) are the major elements — 50.7%, 41.24%, 1.38% and 1.2%, respectively, for Tunisian PG and 52.6%, 42.82%, 2.72% and 0.7%, respectively, for Spanish PG.

TABLE 1. COMPOSITION OF THE PG SAMPLES

Country of origin	Concentration (wt%)								
	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	SO_3	Na_2O	P_2O_5	F
Tunisia	41.24	1.38	0.11	0.09	0.02	50.7	0.59	1.2	4.9
Spain	42.82	2.72	0.40	0.22	0.07	52.6	0.24	0.7	—

The morphological study of the PG samples using SEM, illustrated in Fig. 3, shows two different sections of the sample. The micrographs reveal a homogeneous and prismatic PG piling arrangement and a well defined crystalline structure with a majority of orthorhombic shaped crystals [6–7]. Similar results

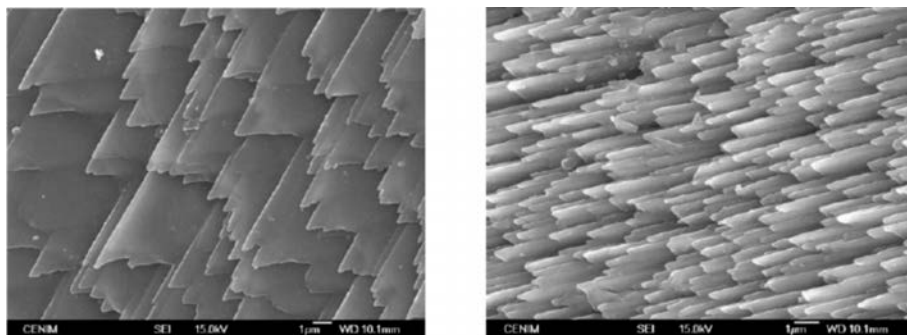


FIG. 3. SEM micrographs of two different sections of a PG sample (15 keV).

can be observed in the study performed by Miloš and Dragan [8], in which they reported that the marked crystal structure of PG indicates that PG presents a more complex composition than natural gypsum (characterized by a poorly expressed crystalline structure), which may eventually influence its chemical behaviour.

3.2. Radionuclide concentrations

The results of the radionuclide analyses of each of the PG samples are given in Table 2 and show that, in the samples originating from both Tunisia and Spain, ^{226}Ra , ^{210}Pb and ^{210}Po are the main sources of radioactivity.

The results obtained were compared with those reported for other countries [9–12] (see Table 3). It is evident that the activity concentrations in PG of Tunisian origin are lower than those in PG originating from most other countries. Moreover, the concentrations of ^{238}U , ^{232}Th and ^{40}K are significantly lower than the worldwide average values for soil (0.05 Bq/g for ^{238}U and ^{232}Th and 0.5 Bq/g for ^{40}K [13]). The lower activity concentrations compared with those for PG from other countries may be attributed to the nature of the phosphate rock, the depth of sampling [14] and differences in the industrial process applied to obtain phosphoric acid. A recent analysis of natural radioactivity in the different phases of the production process [15] showed that isotopes of lead, radium and, to a certain extent, thorium are transferred directly from the phosphate rock and remain associated with the PG particles, while the uranium is present at significantly lower concentration values, depending on the number of washings of the PG. The data obtained in the present study show that the activity concentrations in the PG samples originating from both Tunisia and Spain are lower than the activity concentration screening levels recommended by the European Commission for common building materials and industrial by-products

TABLE 2. ACTIVITY CONCENTRATIONS IN THE PG SAMPLES

	Activity concentration (Bq/g)	
	PG from Tunisia	PG from Spain
U-238, phosphorimetry	0.030 7	0.102 ± 0.001
U-238, gamma spectrometry (²³⁴ Th)	0.027 ± 0.004 9 (DL = 0.04)	0.081 ± 0.028
U-234	0.0316	0.105 ± 0.005
Ra-226 (Bi-214)	0.188 ± 0.009 5	0.520 ± 0.023
Pb-210	0.163 ± 0.081	0.881 ± 0.058
Po-210	0.194 ± 0.078	0.820 ± 0.043
U-235, phosphorimetry	0.001 4	0.004 7 ± 0.000 2
U-235, gamma spectrometry	<0.006 5 (DL)	0.008 ± 0.003 (DL = 0.02)
Th-232 (Ac-228)	0.012 4 ± 0.001 4	0.008 ± 0.002
K-40	<0.013 5	<0.039 (DL)

Note: Ranges represent plus or minus two standard deviations. DL — detection limit; PG — phosphogypsum.

used for building materials [16]. This finding suggests that the PG in this study could be used as a building material.

Concerning the measurement techniques used in this study, it was found that the use of gamma spectrometry for uranium determination allowed the analysis of a more representative aliquot of the whole sample than in the case of the kinetic phosphorimetry technique. The kinetic phosphorimetry technique has a lower detection limit (i.e. greater sensitivity) and lower uncertainty (6%), but owing to the limitations of wet digestion until total dissolution and chemical interferences, only 1 g can be analysed. Nevertheless, the results obtained from both techniques are in good agreement.

TABLE 3. ACTIVITY CONCENTRATIONS IN PG, BY COUNTRY [9–12]

	Activity concentration (Bq/g)				
	U-238	Th-230	Ra-226	Pb-210	Po-210
Spain	0.22	0.008 2	0.67	0.52	—
China	0.015	—	0.085	0.082	0.082
Indonesia	0.043	—	0.473	0.48	0.45
India	0.06	—	0.510	0.49	0.42
Egypt	0.14	0.008 3	0.459	0.323	—
USA (Florida)	0.13	0.113	1.14	1.37	1.03
Australia	0.01	—	0.5	—	—
Sweden	0.39	—	0.015	—	—
Tunisia (from Table 2)	0.03	—	0.188	0.163	0.194

A radioactive environmental follow-up campaign, led by the French company Algade and the Tunisian National Centre for Radiation Protection, was carried out before, during and after completion of the excavation and confinement work of the PG deposit in the Taparura zone. The measurements made within the site environment showed that there were no residual radiological impacts.

4. CONCLUSION

The PG samples from Sfax exhibited low activity concentrations (0.03 Bq/g for ^{238}U , 0.188 Bq/g for ^{226}Ra , 0.163 Bq/g for ^{210}Pb and 0.0124 Bq/g for ^{232}Th) compared with those in the PG samples from Huelva, Spain (0.102 Bq/g for ^{238}U , 0.520 Bq/g for ^{226}Ra , 0.881 Bq/g for ^{210}Pb and 0.008 Bq/g for ^{232}Th). The results provide further support for the safety of the confinement of PG and the development of the confined deposit into an urban park. Nevertheless, a medium and long term follow-up is deemed necessary to guarantee the environmental

quality of the confinement system and the absence of any health impacts in the Taparura zone.

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RAPPORTEUR SUMMARY OF OPENING SESSION AND TOPICAL SESSION 1

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1. INTRODUCTION

This report briefly summarizes the sessions of day 1 (22 April 2013) of the Seventh International Symposium on Naturally Occurring Radioactive Material (NORM VII), in Beijing, China. From the opening ceremony, it was clear that the NORM VII symposium aims to create a forum for discussion on best available practices for sustainable development and radiation protection for NORM processing industries.

2. MANAGING EXPOSURE TO NATURAL SOURCES: INTERNATIONAL STANDARDS AND NEW CHALLENGES

The radiation protection standards and regulatory approaches being adopted at the national level still need to be further harmonized, especially in developing countries with limited regulatory resources. The International Basic Safety Standards (BSS), published by the IAEA in 2011, provide requirements reflecting the concepts of planned, existing and emergency exposure situations. The BSS promotes a graded approach. A legislator perspective on NORM: “If it is not defined in national legislation as needing to be regulated, then it is not NORM”. Important challenges and issues that need to be dealt with when adopting the revised BSS in national regulatory approaches are listed below:

- (a) Practicality is a very important issue when regulating exposure to NORM;
- (b) A challenge is to adopt recycling or by-product use to reduce the amount of NORM waste;
- (c) More attention needs to be paid to stakeholder involvement and communication;
- (d) Standardization of measurement methods and protocols is important for the evaluation of practices.

These challenges and issues were considered by several speakers. Clearly these are some of the central issues that were considered in Topical Session 1.

3. MINING: MOVING TOWARDS COMPREHENSIVE EXTRACTION

China is a mineral rich country with more than 168 types of mineral resource. The United States of America has the largest thorium reserves in the world. Australia is characterized by large distances and at least 96 mines. Australia hosts more known uranium resources (1 673 000 t of uranium) than any other country. A large amount of exploration for oil and gas, uranium, rare earths, phosphates and mineral sands, and metalliferous deposits is ongoing in Australia.

In the past, radiation protection was considered to be ‘an interesting idea’ in uranium mining. Remediation of the Alligator Rivers Region was eventually dealt with by ‘putting everything back in the pit’. Modern mining companies have to assure cleanup from the start of the operation. In Australia, a company needs to provide a deposit of 100% of the estimated costs for the remediation before startup.

Bad ventilation and high radon concentrations are common in underground mines in China. In 15% of non-uranium mines, radon concentrations can exceed 1000 Bq/m³. Health effects are reported for miners exposed to the highest radon concentrations. Effective regulations and measures are required to protect the miners.

A study involving risk perception among non-uranium miners was reported. In this study, 2836 miners were interviewed and only 1.8% of them had a correct perception of health risk resulting from exposure to radon. Risk and harm perception are complicated issues. For comparison purposes, it can be useful to ask similar questions regarding the perceived risks to various groups: experts and non-experts.

Mining of NORM containing ores presents specific challenges for the supervising authorities. Guidance for industry is required to support the implementation of various aspects of NORM regulations. Currently, we have a global market without global regulation or standards and this is a cause of instability. If one fulfils the requirements of the collaboration model, which means to find an equilibrium between the interests of stockholders and concerned parties, then a new ‘social licence’ can be obtained. In this context, the idea of ‘comprehensive extraction’ is rapidly gaining ground. Comprehensive extraction requires a long term approach. Three key features of comprehensive extraction are:

- (1) Disturb the ground only once;
- (2) Extract all useful materials;
- (3) Leave no waste behind (use or recycle residues).

In the future, there is a good chance of another uranium mine in Australia: ‘The future involves U’. Stakeholder and stockholder involvement is important when starting a new mine. An evidence based approach to risk management should be incorporated into a long term strategic resource plan.

4. THE NORM PROCESSING INDUSTRY: HOW TO HANDLE MEASUREMENT PROTOCOLS AND PRACTICAL ISSUES

Throughout several presentations, a treasure of information — including many facts and figures on NORM exposure for millions of workers in China — was discussed. The average dose received by millions of workers in China is 2.1 mSv/a. Several radiation protection issues associated with NORM exposure were considered. Out of a survey consisting of on-site investigations of 11 000 enterprises in China from 11 types of NORM industry, it was found that more than 300 enterprises have raw materials or residues with ^{238}U or ^{232}Th activity concentrations exceeding 1 Bq/g.

A radionuclide balance was determined for a coal fired power plant and the stack emissions of the aerosol ^{210}Po were determined. A stack sampling instrument was developed to sample the aerosol radionuclides. Micromorphological analysis of the aerosol ^{210}Po from the stack was performed by means of scanning electron microscopy.

NORM has become a widely recognized issue in the oil and gas industry. Different restoration activities, the characterization of NORM residues and the technical challenges of properly evaluating the activity concentrations in such residues were discussed. New NORM related issues are arising. In the United States of America, hydraulic fracturing techniques have seen a massive increase and oil and gas companies need to implement NORM related education and awareness (in both English and Spanish) and proper analytical techniques for characterization of NORM. A large amount of personal protective equipment is required when working with NORM in the oil and gas industry.

When companies are dealing with ores, products and residues with enhanced concentrations of radionuclides of natural origin, many practical issues arise:

- What is a radioactive material and when do we have to do something?
- The problem of ‘greater than 1 Bq/g’ = ‘radioactive?’ = ‘dangerous?’ = ‘extremely low concentration?’

- The label ‘radioactive’ is difficult to remove! Is it possible (useful?) to seek exemption for the materials? Is it possible to negotiate sale terms with the customers for these materials?
- How can we develop our process to reduce the radionuclide concentrations?
- Blending is a normal industrial process: Why is this such an issue with NORM?

A very important aspect that was dealt with is that we need radiation control appropriate to the magnitude of the risk.

5. FROM MANAGEMENT OF NORM RESIDUES AND CONTAMINATED SITES TO RESIDUE RECYCLING/USE?

Iron and steel manufacturers are a large industrial source of environmental contamination — 3100–6200 million t of waste are produced in the United States of America. A study was reported regarding the distribution of radionuclides in the various incoming and outgoing material flows during processing steps of iron ore in Egypt. In this study, samples from four companies were analysed. Exposure pathways for the public and workers were assessed. An important finding was that the concentration of other toxic elements such as cadmium, lead and zinc turned out to be much more problematic than the NORM content of the investigated materials. When evaluating the recycling/use options of NORM residues, the presence of other toxic components might present a much bigger problem that can block several opportunities for such recycling/use.

The phosphogypsum management policy implemented within the Taparura project in Sfax, Tunisia, was aimed at the reduction of the impact of the phosphogypsum residue by finding a better waste management approach with a limited radiological impact. The Taparura project involves the reclamation of 6 km of beaches, removal of contamination, backfilling and environmental and radiological follow-up. Attention is given to the concept of a ‘sustainable city’: the remediation project considers the social, economic and environmental role of region.

Removal options for uranium and thorium from the Kvanefjeld (southern Greenland) rare earth and uranium deposit were discussed and more details regarding the hydrometallurgical flowsheet developed for the Kvanefjeld deposit were given. The current status of the Kvanefjeld multielement project was proposed: currently a feasibility study is ongoing. The department of NORM throughout the flowsheet was discussed in detail. It is the goal to produce uranium as a separate product.

In the United States of America, the phosphate production process is under consideration as a source of co-extraction of uranium, rare earths and thorium. In regulation and dose modelling, a conservative approach is generally used. A drawback of an over-conservative approach to NORM residue recycling/use is that such an approach will generate too much waste, so care needs to be taken to find an equilibrium between radiation protection of the population and allowing recycling/use options for residues.

6. CONCLUSIONS

Mining is moving towards long term approaches such as comprehensive extraction. Stakeholder–stockholder cooperation is important when starting a new mine, also in the NORM processing industry and when dealing with the recycling/use of NORM residues.

Practicality is a very important issue when regulating exposure to NORM. There is a need for radiation control appropriate for the size of the risk. An important challenge is to go for recycling/use of NORM residues to reduce the amount of NORM waste. The control measures need to assure radiation protection of the population and workers while allowing recycling/use options for NORM residues. When implementing protective measures and looking for recycling/use options, there is a need to consider NORM issues and the presence of other toxic elements in these residues. The standardization of measurement methods and protocols is important for the evaluation of practices.

REGULATORY ASPECTS OF NORM:
MEETING THE NEW BSS REQUIREMENTS

(Topical Session 2)

Chairpersons

P.P. HARIDASAN

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Invited Paper

RADON CHALLENGES IN URANIUM MINES AND OTHER WORKPLACES

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Abstract

Uranium ore is one example of NORM, but the mining and processing of uranium ores have been regulated for many years as part of the nuclear fuel cycle. Many other minerals and raw materials fall within the definition of NORM and the mining and processing of such materials have radiological hazards and radiation protection issues similar to those arising from the mining and processing of uranium ore. The magnitude of the hazard depends on the level of radioactivity in the ore or raw material and the details of how the ore or raw material is mined or processed. One important radiological hazard in mines and enclosed spaces is radon. The recent work of the International Commission on Radiological Protection (ICRP) on the risks associated with radon, the ICRP's move toward a fully dosimetric approach for regulating workplace exposure to radon, and the ICRP's recent draft guidance on radiation protection against radon have generated much discussion on the requirements for radiation protection against radon (and thoron) in the workplace and the costs of compliance. The objective of the paper is to review the sources and exposures in uranium mining and other NORM industries, with a focus on radon, and to provide an industry perspective on associated issues which include, among other things, deficiencies in the ICRP's proposed dose coefficients arising from incomplete consideration of the carcinogenic effect of smoking and practical issues associated with the lack of dosimetrically relevant data to support a dosimetric approach.

1. INTRODUCTION

All soils and rocks, and hence all ores, contain radionuclides of natural origin. The concentrations of such radionuclides in a wide variety of materials have been reported by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [1] and are shown to vary over a wide range. Examples of ores that have been found to contain relatively elevated radionuclide concentrations include the ores of uranium, tin, tantalum, niobium, rare earths and aluminium, as well as some copper and gold occurrences and phosphate rock.

The mining and processing of these resources and production of consumer items can lead to further enhancement of the radioactivity in the products, by-products, residues or wastes arising from the industrial processing. A few examples of activity concentrations in such materials are summarized in Table 1 [2].

TABLE 1. EXAMPLES OF ACTIVITY CONCENTRATIONS IN MINERALS AND THEIR PROCESSING RESIDUES

	Radionuclides(s) with the highest activity concentration	Typical range of activity concentrations (Bq/g)
Monazite sand	Th-232 series	40–600
Metal ores (e.g. Nb–Ta, Cu, Au)	U-238 and Th-232 series	Up to 10
Zircon sand	U-238 series	2–4
Phosphate rock	U-238 series	0.03–3
TiO ₂ feedstocks	Th-232	0.001–2
Bauxite	Th-232 series	0.035–1.4
Red mud (alumina production)	U-238, Th-232	0.1–3
Phosphogypsum (H ₂ SO ₄ process)	Ra-226	0.015–3
Nb extraction slag	Th-232	20–120
Sn melting slag	Th-232	0.07–15
Scale (oil and gas production)	Ra-226	0.1–15 000
Residue (rare earth extraction)	Ra-228	20–3 000
Scale (TiO ₂ pigment production)	Ra-228, Ra-226	<1–1 600
Scale (rare earth extraction)	Ra-226, Th-228	1 000
Sludge (oil and gas production)	Ra-226	0.05–800

TABLE 1. EXAMPLES OF ACTIVITY CONCENTRATIONS IN MINERALS AND THEIR PROCESSING RESIDUES (cont.)

	Radionuclides(s) with the highest activity concentration	Typical range of activity concentrations (Bq/g)
Residue (Nb extraction)	Ra-228	200–500
Coal	U-238 and Th-232 series	0.01–0.025
Scale (coal mines with Ra rich inflow water)	Ra-226, Ra-228	Up to 200

Source: Adapted from Ref. [2].

During the mining and processing of ores, workers and nearby members of the public may be exposed to elevated levels of radiation and radioactivity (see annex B of Ref. [3]). Elevated levels of natural background radiation are seen in many occupational settings, notably in underground workplaces for the mining of minerals such as uranium, coal and gold. In addition, in other enclosed workplaces such as those in process plants, exposure to radon, thoron and their decay products can be an important source of occupational exposure.

This paper provides a brief review of the sources and exposures in uranium mining and other selected NORM industries, with a focus on radon, and of the limitations and challenges arising from the implementation of the new approach to protection against radon recommended by the International Commission on Radiological Protection (ICRP).

2. URANIUM MINING

Until recently, epidemiological studies of miners provided the main basis for estimating the risks from exposure to radon (see annex E of Ref. [4]). Today, case control studies of residential exposure to radon also show a risk of lung cancer increasing with increasing exposure to radon [4, 5]. To implement its system of radiation protection, the ICRP established an epidemiologically based

dose conversion convention for radon (i.e. mSv per working level month)¹ to allow doses from exposure to radon to be added to doses from other sources of exposure [6]. The ICRP now recommends a doubling of its nominal risk coefficient for radon induced lung cancer based on its review of uranium miner epidemiology [5]. The ICRP also notes that the risk of lung cancer from radon is substantially greater for smokers than for non-smokers [5]. This is an important observation, as the current radon risk projection models are all relative risk models and thus risk estimation is strongly influenced by smoking prevalence in the reference populations.² According to UNSCEAR (see annex B of Ref. [3]), radon exposure accounts for about 60% of the annual effective dose received by underground uranium miners in Canada. Assuming the dose conversion factors are to be doubled and all other factors remain the same, the total dose received by a Canadian uranium miner would increase by about 60% over current levels. UNSCEAR also reports average annual doses of about 8 mSv from radon exposure for three underground uranium mines in India (see annex B of Ref. [3]). In terms of the ICRP's proposal, the annual average effective dose received by those miners would increase to about 16 mSv.

In addition, the ICRP now proposes to treat radon in the same fashion, as it deals with other radionuclides in its system of radiation protection, namely, using ICRP's biokinetic dose models, and that it will provide 'nominal' dose coefficients calculated using its biokinetic dose models and 'nominal' parameter values [5, 7]. In considering the use of biokinetic dose conversion factors, it is important to understand that mine environments vary widely, not only between mines but also within mines, and with time as a result of changes in the ore body and mining method. Moreover, methods for practical measurement of dosimetrically relevant parameters, notably the activity size distribution of radon progeny, are currently very limited. In addition, this variability presents challenges to the development of measurement programmes, dosimetric evaluations and the interpretation of these evaluations.

¹ It should be noted that since the risk from radon exposure is to certain cells in the respiratory tract, the comparison is, in effect, a comparison of risk of lung cancer and risk of cancer from whole body exposure.

² That is, the epidemiologically based radon risk models are applied to the risks of lung cancer in the underlying, baseline or reference populations. In other words, the increased risk assigned to radon is actually the risk arising from the combination of underlying lung cancer rates in the reference population which arise primarily from smoking and exposure to radon.

3. PHOSPHATE

Phosphate fertilizer is used throughout the world in agriculture to help sustain food production. Phosphate rock which contains low concentrations of radionuclides of natural origin, primarily the uranium series radionuclides, is mined and then processed to produce phosphate fertilizers. Phosphogypsum (PG) is formed as a by-product of fertilizer production when sulphates from the acid react with the calcium from the base rock of the phosphates. The PG is pumped as a slurry to ponds where the solids settle out. The water is drained to the cooling pond and then returned to the plant for reuse. Once the solids fill up the ponds, containment dykes are built up with the dewatered PG. This process of stacking the dewatered PG forms what is known as a PG stack [8].

The main radiological concern with phosphate mining and processing is from indoor radon and gamma radiation. While workers can be exposed to gamma radiation, dust and radon, the major concern appears to have been exposure of the public from radon from PG stacks [9–13]. With the majority of the PG in the United States of America centralized in Florida, that state established the Florida Institute of Phosphate Research (FIPR) to study phosphate issues that impact Florida.³ Studies by the FIPR and others have confirmed the low risk arising from the use of PG as a by-product, including as an agricultural soil amendment and fertilizer, as a road base and as a daily landfill cover.

It is considered important to comment briefly on some of the current work on using PG as a resource. According to Ref. [14], the amount of PG in stacks is growing at more than 100 million t/a and the total amount worldwide will double within 30 years or so. The land based stacks such as those in central Florida are often in prime, highly sensitive and increasingly populated areas [14]. The FIPR, working with the IAEA and others, initiated a global effort referred to as the ‘Stack Free’ project to achieve eventual equilibrium between production and consumption of PG across the full life cycle of a producing facility, with the aim of considering PG more as a resource than as a waste [13, 14].

4. RARE EARTHS

Ore deposits associated with rare earth elements (REEs) contain moderate concentrations of radionuclides in the thorium and uranium decay chains. Owing to the need for regulatory control as a result of this radioactivity content, REE ores and process materials are generally regarded as NORM [15, 16]. Although

³ See <http://www.fipr.state.fl.us/about-fipr-general.htm>.

mining and processing strategies for REEs concentrate production are resource and location specific, the radionuclide concentrations are generally much higher than natural background levels and regulatory control has to be considered. In addition, specifications for hydrometallurgical REE concentrates typically stipulate very low levels of radioactivity, resulting in essentially all of the activity ending up in the mineral concentration rejects (tailings) and the metallurgical residues. The accumulation of activity in such residues is an additional reason for having to consider radiation protection during the production of REE concentrates.

In some projects, consideration may be given to the recovery of uranium (or thorium) as a by-product. While this may be technically and economically feasible, the recovery of uranium and/or thorium as a by-product could, in some countries, change the regulatory environment. In Canada, for example, if uranium and thorium are recovered during the process, the Canadian Nuclear Safety Commission (CNSC) regulations will apply and the project would be subject to licensing by the CNSC and to a federal environmental assessment process. However, if neither uranium nor thorium is recovered, the material would be specifically excluded from the regulatory mandate of the CNSC. Instead, it would most likely be subject to Health Canada's Guidelines for NORM [17]. In the end, a decision on whether to extract uranium or thorium as a by-product would be made on the basis of a number of factors, among them technical feasibility, regulatory and environmental issues and, importantly, project economics. The measurement and management of worker exposures and the provision of secure long term waste management arrangements are important components that have to be taken into account when considering the authorization and public acceptance of REE resource development.

5. DISCUSSION

Human activities can redistribute and concentrate radionuclides of natural origin in products, by-products and residues. Legal frameworks, regulations and guidelines for the management of various types of NORM in the mining and mineral processing industry (i.e. feedstocks, products, by-products and residues) are provided by various national agencies such as Health Canada [17] and regional bodies such as the European Commission [18, 19]. These are generally based on IAEA standards, guidelines and supporting documents for protection of workers and members of the public.

The anticipated reduction of exposure limits for radon and its progeny will complicate the development of new underground uranium mines and has the potential to result in radiation protection issues in other NORM related mines and

process plants. As previously noted, there are many potential uses of by-products of NORM mining and processing — among them the agricultural use of PG from phosphate fertilizer production. Other facilities that may be impacted by the forthcoming changes to radiation protection against radon include tourist caves and water treatment facilities.

Social acceptability has become an increasingly important component of any type of mine and industrial development anywhere in the world. The presence of NORM can render the acceptability of such developments (i.e. the by-product use of phosphogypsum) much more challenging. Recent experience in Canada and other countries has shown that in general terms, the following issues need attention when considering new NORM projects:

- (a) A detailed understanding of the distribution of radionuclides of natural origin in all phases of the process and a complete pathway assessment of all such phases by qualified independent third parties;
- (b) For projects involving REE production, a full discussion with the local public and the regulatory body on the radiological characteristics of the REE resources in relation to local natural background radiation;
- (c) A discussion of dose and risk with workers;
- (d) A review, with members of the public and the regulatory body, of the options for use of by-products and the options for managing any wastes arising from mining or processing, as well as any potential radiological risks in the long term.

REE resources contain radionuclides from the thorium and uranium decay series, usually at concentrations significantly above natural background levels. Exposure management in REE ore processing, concentrate production and element separation involves the minimization of exposure of workers and members of the public. Credible expertise and experience is available to ensure that radiation exposures are kept below very conservative levels. Public concerns about REE developments, whether based on intuition, facts or fears fed by third party exaggeration can in large part be addressed by the development of an understanding of exposure to natural sources and by dose assessments coupled with the application of proven and robust waste management strategies.

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RADON CHALLENGES IN URANIUM MINES AND OTHER WORKPLACES

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Invited Paper

RADIOLOGICAL PROTECTION AGAINST RADON EXPOSURE

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Abstract

At its meeting in Porto, Portugal, in November 2009, the Main Commission of the International Commission on Radiological Protection (ICRP) approved the formation of a new Task Group, reporting to Committee 4, to develop guidance on radiological protection against radon exposure. The paper is a description of the Task Group's draft report which has been posted on the ICRP website for public consultation. In this report, the Commission provides updated guidance on radiological protection against radon exposure. The report has been developed considering the recently consolidated ICRP general recommendations, the new scientific knowledge about the radon risk and the experience gained by many organizations and countries in the control of radon exposure. The report describes the characteristics of radon exposure, covering sources and transfer mechanisms, the nature of the risk, the exposure conditions, the similarities with other existing exposure situations and the challenges to manage radon exposure. To control the main part of radon exposure, the Commission recommends an integrated approach focused as far as possible on the management of the building or location in which radon exposure occurs, whatever the purpose of the building and the types of its occupants. This approach is based on the optimization principle and a graded approach according to the degree of responsibilities at stake, notably in workplaces, and the level of ambition of the national authorities. The report emphasizes the importance of preventive actions. The report also considers how to control radon exposure in workplaces when workers' exposure can reasonably be regarded as being the responsibility of the operating management. In such a case, workers' exposures are considered as occupational and controlled using the corresponding requirements on the basis of the optimization principle and the application, as appropriate, of the dose limit.

1. INTRODUCTION

The objective of the International Commission on Radiological Protection (ICRP) future publication on radon is to describe and clarify the application of the Commission's system to the protection of members of the public and workers

against ^{222}Rn and ^{220}Rn exposures in dwellings, workplaces and other types of locations.

Radon-222 is a radioactive decay product of ^{238}U which is present in the Earth's crust in varying concentrations. Because radon is a gas, it is capable of movement from the soil to indoors. This movement is dependent on the type of building and/or location. Radon-220 is a radioactive decay product of ^{232}Th , also present in the Earth's crust. Both ^{222}Rn and ^{220}Rn may also come from some building materials. The concentration of radon in a building may vary by several orders of magnitude.

Because radon is inert, nearly all of the gas inhaled is subsequently exhaled. However, when inhaled, the short lived radon progeny can deposit within the respiratory tract. Depending on the diffusion properties of the particles (size distribution of the aerosols), the decay products present in the air deposit in the nasal cavities, on the walls of the bronchial tubes and in the deep lung. Two of these short lived progeny, ^{218}Po and ^{214}Po , emit alpha particles, and the energy deposited by these alpha particles may lead to health effects, principally lung cancer.

The Commission recently made a thorough review and analysis of the epidemiology of radon for both workers (underground miners) and the general population [1]. There is now compelling evidence that radon and its progeny can cause lung cancer. For solid tumours other than lung cancer, and also for leukaemia, there is currently no convincing or consistent evidence of any excesses associated with radon and radon progeny exposures. For radiological protection purposes the Commission now recommends a detriment adjusted nominal risk coefficient for a population of all ages of 8×10^{-10} per $\text{Bq}\cdot\text{h}\cdot\text{m}^{-3}$ for exposure to ^{222}Rn gas in equilibrium with its progeny (i.e. 5×10^{-4} WLM $^{-1}$), which is approximately twice the value previously used by the Commission in Publication 65 [2]. It should be noted that the dose conversion factor will depend on the new dose coefficients currently under revision.

2. CHARACTERISTICS OF RADON EXPOSURE

Radon exposure situations have the characteristics of existing exposure situations, since the source is unmodified concentrations of ubiquitous natural activity in the Earth's crust. Human activities may create or modify pathways increasing indoor radon concentration compared with the outdoor background. These pathways can be controlled by preventive and corrective actions. The source itself, however, cannot be modified and already exists when a decision on control has to be taken. Some workplaces, however, may be deemed to be planned

exposure situations from the outset by national authorities. Such workplaces may include uranium mines associated with the nuclear fuel cycle.

Radon is not likely to give rise to an emergency exposure situation even though the discovery of very high concentrations in a place may require the prompt implementation of protective actions. The philosophy of Publication 103 [3] compared with that of Publication 60 [4] is to recommend a consistent approach for the management of all types of exposure situations. This approach is based on the application of the optimization principle implemented below appropriate constraints or reference levels.

Several characteristics of radon exposure in dwellings (and in many other locations) are similar to those of exposures arising from other existing exposure situations such as exposures to NORM or exposures in a long term contaminated area after a nuclear accident or a radiation emergency. Radon exposure affects nearly all living places of a population. The ubiquity of radon and the variability of its concentration result in a very heterogeneous distribution of exposures. Day to day life or work inevitably leads to some exposure to radon. The persistence or reduction of the risk is mainly dependant on individual behaviour. Domestic radon exposure management is to address several considerations such as environmental, health, economic, architectural and educational considerations. A large spectrum of parties is concerned. The role of self-help protective actions is also crucial.

Control of indoor radon exposure poses many challenges. As a given individual can move from place to place in the same area, the radon policy is to provide consistency in the management of the different locations in an integrated approach. As the radon risk is mainly due to domestic exposure, the radon policy is to address primarily exposure in dwellings from a public health perspective. As the radon concentration in many buildings is above the level at which the risk has been demonstrated, a real ambition is needed to reduce both the overall risk for the general population and the highest individual exposures. Radon policy is not to be in contradiction with the growing role of energy saving policies. It is to be as simple as possible, properly scaled with other health hazards, supported and implemented on a long term basis and involving all the concerned parties.

A national radon policy has also to address many challenges in terms of legal responsibilities, notably the responsibility of the individual householder towards her/his family, of the seller of a house or a building towards the buyer, of the landlord towards the tenant, of the employer toward the employee, and generally speaking of the responsible person for any building towards its users. The degree of enforcement of the actions that are warranted is very much related to the degree of legal responsibility for the situation.

3. A REALISTIC, INTEGRATED, GRADED AND AMBITIOUS APPROACH

The responsibility dimension calls clearly for the need of a graded approach in defining and implementing a radon policy. Such a graded approach is to be based on realism, effectiveness and ambition. Any radon policy thus aims to maintain and/or reduce radon concentration as low as reasonably achievable in an effective way keeping in mind that it is not possible to totally eliminate indoor radon concentration.

3.1. Application of the principles

The Commission considers that a national radon protection strategy appears to be justified since radon is a significant source of radiation exposure (the second greatest cause of lung cancer after smoking), radon exposure can be controlled and a radon policy has positive consequences on other public health policies (indoor air quality or anti-smoking policies). The Commission considers that radon strategies should address both smokers and non-smokers together.

As with other sources of exposure, it is the responsibility of the appropriate national authorities to establish their own national reference levels, taking into account the prevailing economic and societal circumstances, and then to apply the process of optimization of protection in their country. The objective is to reduce both the overall risk of the general population and, for the sake of equity, the individual risk, in particular the risk of the most exposed individuals. In both cases, the process is implemented through the management of buildings and will ideally result in radon concentrations in ambient indoor air as low as reasonably achievable below the national reference level.

According to the characteristics of the radon exposure and the approach to its control — control by actions on pathways, benefit for individuals due to the use of buildings, general information provided to enable individuals to reduce their doses — the appropriate reference level is to be set, corresponding to an annual dose in the range 1–20 mSv (see table 5 of Publication 103 [3]). Further, the value of 10 mSv, which is the middle of this range, remains the upper value of the dosimetric reference level for radon exposure as set in Publication 65 [2].

Reference levels for radon are typically set in terms of the measurable quantity becquerels per cubic metre of air. The Commission therefore recommends an upper value of the reference level for radon gas in dwellings of 300 Bq/m³

(see the ICRP Statement from the Porto meeting [1]).¹ The measurement will be representative of the annual mean concentration of radon in a building or location. For the sake of simplicity, considering that a given individual going from place to place in the same area during the course of the day is to be protected on the same basis whatever the location, the Commission recommends to use a priori the same upper value of 300 Bq/m³ in mixed use buildings (with access for both members of the public and workers).

Within a graded approach, the radon protection strategy would start with a programme aiming at encouraging relevant decision makers to enter into a process of self-help protective actions such as measurement and, if needed, remediation, with more or less incentive and helping provisions and, if judged necessary, even requirements. Then the degree of enforcement of these various actions would be increasing depending on the degree of legal responsibility for the situation and the ambition of the national radon protection strategy.

3.2. Specific graded approach for workplaces

A specific graded approach should ideally be implemented in workplaces. Where workers' exposures to radon are not considered as occupational exposures — that is, when workers exposures to radon cannot reasonably be regarded as being the responsibility of the operating management (typically office buildings) — the first step is to reduce the concentration of ²²²Rn to a level as low as reasonably achievable below the same reference level as set for dwellings (even though the corresponding level in dose is below 10 mSv per year because the conditions of exposure in the workplace are different from those in dwellings). If difficulties are encountered in this first step, a more realistic approach could follow as the second step. It means optimizing exposure on the basis of a dose reference level of 10 mSv per year taking into account the actual parameters of the exposure situation.

In workplaces, if, despite all reasonable efforts to reduce radon exposure, the exposure continues to remain above the dose reference level of 10 mSv per year, and/or where workers' exposure to radon can reasonably be regarded as being the responsibility of the operating management (e.g. some underground workplaces and spas), the workers should be considered as occupationally exposed. In such cases, the Commission recommends applying the optimization principle and the relevant requirements for occupational exposure.

¹ This numerical value may change according to the revised dose coefficients.

The dose limit would apply when the national authorities consider that the radon exposure situation is to be managed like a planned exposure situation. In any case, using either the occupational dose limit or a reference level, the upper value of the tolerable risk for occupational exposure (of the order of 20 mSv per year, possibly averaged over five years) is not to be exceeded.

3.3. National action plan

A national radon action plan is to be established by national authorities with the involvement of relevant concerned parties in order to frame the implementation of the national radon protection strategy in dwellings, places open to the public and workplaces. The action plan would establish a framework with a clear infrastructure, determine priorities and responsibilities, describe the steps to deal with radon in the country and in a given location, identify concerned parties (who is exposed, who should take actions and who could provide support), address ethical issues (notably the responsibilities) and provide information, guidance, support as well as conditions for sustainability. To be efficient, the national radon protection strategy should ideally be established on a long term perspective. The process to significantly reduce the radon risk of the general population is rather a matter of several decades than several years. The national action plan would be reviewed periodically, including the value of the reference level.

The Commission now considers that for the sake of clarification, when dealing with existing exposure situations, the distinction should be made between prevention aiming at maintaining exposure as low as reasonably achievable under the prevailing circumstances and mitigation aiming at reducing exposure to a level as low as reasonably achievable. As a consequence, a radon protection strategy should ideally include a prevention part. Whatever the indoor location is, the category of individuals inside and the type of exposure situation, it is possible to optimize radon exposure by taking into account the issue of radon exposures during the planning, design and construction phase of a building. Preventive actions mean land planning and building codes for new buildings and for renovation of old buildings. They also mean the integration of the radon protection strategy consistently with other strategies concerning buildings such as indoor air quality or energy saving in order to develop synergies and avoid contradictions.

The mitigation part of a national radon protection strategy concerns mainly existing buildings or locations. Then ideally, the control of exposure should be ensured as far as possible through the management of the building (or location) and the conditions of its use, whatever the category of individuals inside. The main steps are measurement and, when needed, corrective actions. The action plan would also deal with radon measurement techniques and protocols, national

radon surveys to identify radon prone areas, methods for mitigating the radon exposure and their applicability in different situations, support policy including information, training and involvement of concerned parties as well as assessment of effectiveness. The issues of buildings with public access and workplaces, with specific graded approaches, is also to be addressed.

A schematic representation of the general approach to the management of radon exposure is given in Fig. 1.

4. CONCLUSION

People are exposed to radon at home, in workplaces and in mixed use buildings. The detriment adjusted nominal risk coefficient recommended by the Commission is now approximately twice the value previously used in Publication 65 [2]. Radon exposure situations are existing exposure situations since the source is unmodified concentrations of ubiquitous natural activity in the Earth's crust. Only pathways can be controlled.

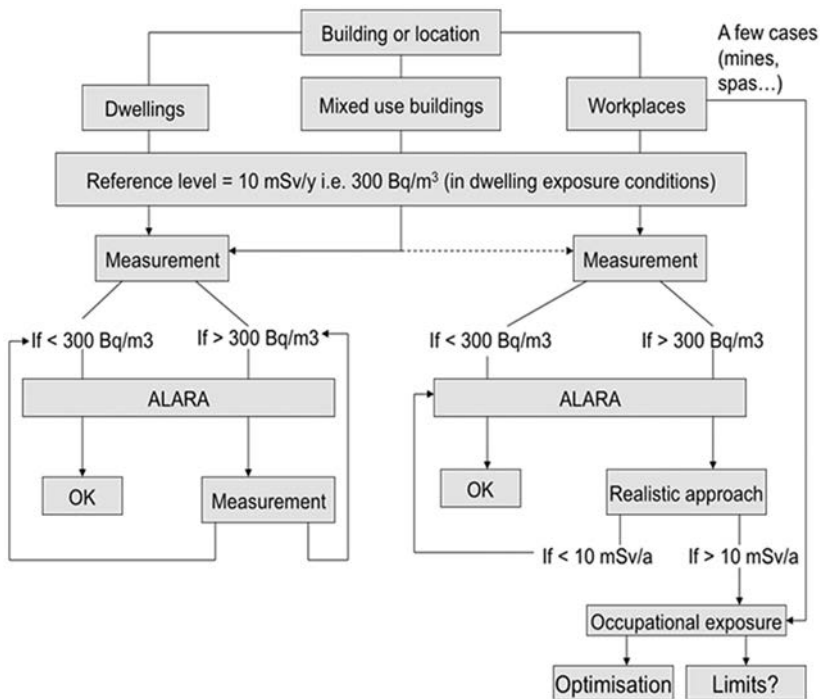


FIG. 1. General approach for the management of radon exposure.

Radon exposure has key characteristics: it is mainly due to domestic exposure (public health perspective); radon concentration in many buildings is above the level at which the risk has been demonstrated; radon policy may be in contradiction with other policies such as energy saving policy; the persistence or reduction of the risk is mainly dependant on individual behaviour (self-help protective actions); efficiency can only be achieved in a long term perspective; exposure in workplaces may be adventitious (i.e. they cannot reasonably be regarded as being the responsibility of the operating management) and not occupational.

The justification of launching a national radon strategy (national action plan) is a decision for the national authorities. The radon strategy should ideally be simple and realistic (same approach for smokers and non-smokers), integrated (consistent for all buildings), graded (according to the situation and the legal responsibilities) and ambitious (choice of the reference level; addressing both the highest exposures and the global risk). The radon strategy should ideally include both preventive (new buildings) and corrective (existing buildings) actions.

The management of radon exposure is mainly based on the application of the optimization principle below an appropriate reference level. The Commission recommends 10 mSv per year as an appropriate dosimetric reference level for radon exposure. The upper value of the reference level recommended in dwellings is 300 Bq/m³ (annual mean concentration). For the sake of simplicity, the same value is recommended by the Commission for mixed use buildings.

A specific graded approach is recommended by the Commission in workplaces:

- (1) Application of the same reference level (radon concentration) as for dwellings (although the corresponding dose is below 10 mSv/a mainly because of the time of exposure);
- (2) Application of the dosimetric reference level (10 mSv/a) taking into account the actual conditions of exposure;
- (3) Application of the relevant requirements for occupational exposure when, despite all reasonable efforts, the exposure remains above 10 mSv/a (quantitative criterion) or when the work activity is included in a national list of radon prone work activities (qualitative criterion).

The dose limits may be applied when the national authorities consider that the radon exposure situation is to be managed like a planned exposure situation.

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Invited Paper

RADIATION SAFETY REGULATORY POLICY AND RULE FOR NORM INDUSTRIES IN CHINA

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Abstract

Background information and the basic status concerning NORM industries in China is briefly introduced in the paper, including current natural radiation levels and the main results of a general survey related to NORM industries implemented by the Ministry of Environmental Protection. An introduction to the radiation safety regulatory policy and rule for NORM industries in China is also briefly presented. Finally, some considerations concerning NORM regulatory issues are discussed.

1. BACKGROUND

In accordance with the Law on Prevention and Control of Radioactive Pollution, the Ministry of Environmental Protection (MEP) in China is responsible for the regulatory control of NORM industries with respect to radiation safety and environmental protection.

Natural radiation exposure levels vary widely across different regions of China. The average annual effective dose to the public from natural background exposure is about 3.1 mSv/a. A Geochemistry Integrated Map for uranium, thorium and potassium in China for gamma dose rates at 1 m above ground is shown in Fig. 1.

Natural radiation associated with human activities is the major contributor to public and occupational exposure. The annual effective doses to the public are 0.64 mSv/a from exposure to indoor radon, 0.2 mSv/a from medical exposure, 0.01 mSv/a from coal fired power generation and only 8×10^{-6} mSv/a from nuclear power generation. The use of mineral waste or slag as a building material gives rise to elevated exposures to indoor radiation.

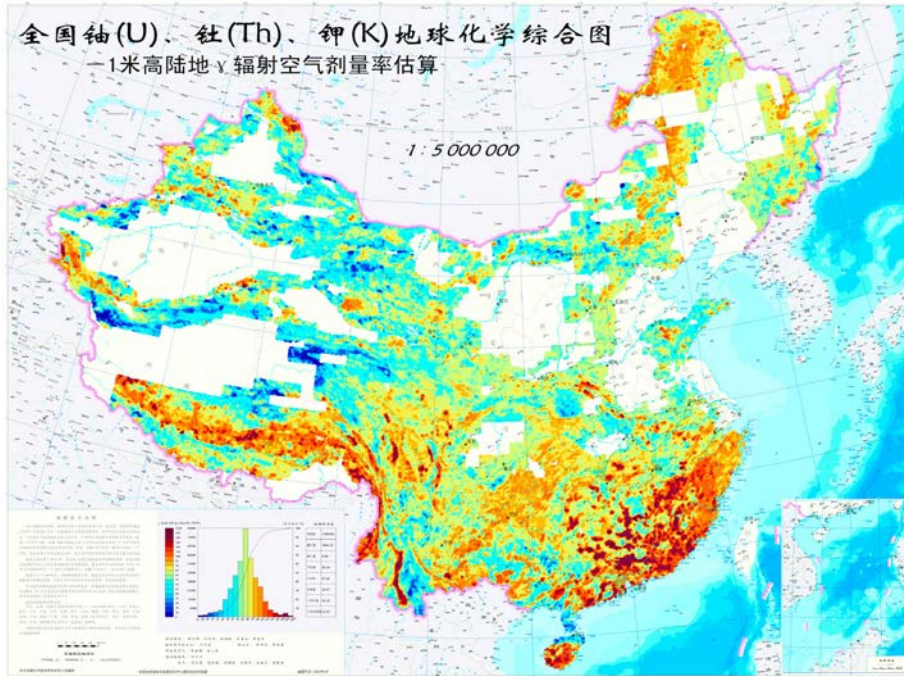


FIG. 1. Geochemistry Integrated Map for uranium, thorium and potassium in China for gamma dose rates at 1 m above ground.

2. REGULATORY FRAMEWORK RELATING NORM INDUSTRIES

The Law on Prevention and Control of Radioactive Pollution was established by Communist Party of China in 2003. It is a major law relating to nuclear and radiation safety and radiation in the environment. Among others, this law also establishes the basic regulatory principles that are applied to NORM industries. The law requires that the owner of a non-uranium mine containing elevated levels of radionuclides of natural origin should conduct an environmental impact assessment and obtain approval and supervision from the local provincial environmental protection agency. Under the framework of this law, a regulation on the safe management of radioactive waste was implemented by the State Council in 2011.

The ‘Chinese BSS’, entitled Basic Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (GB18871-2002), was implemented by the Government in 2002. The Chinese BSS clearly defines the human activities involving natural sources that are not to be excluded or exempted and that should be incorporated into the scope of the management

of radiation protection. In addition to the above mentioned law, regulation and national standards, other regulatory rules or guides were published respectively, such as:

- (a) Regulations for radioactive waste management (GB14500-2002);
- (b) Administrative rules on the prevention and control of pollution by tailings;
- (c) A list (first batch) of regulations on the radiological environmental impact associated with the exploration and utilization of mineral resources (2013);
- (d) A requirement on the control of radioactive substances for building material products and industrial by-products used in building materials (GB6763-2000).

The list mentioned in (c) above is shown in Table 1 and was published as a notice by the MEP in February 2013. The purpose of this notice is to protect the environment and health of the public in accordance with the law on prevention and control of radioactive pollution and the law on assessment of environmental impact. The scope of this notice, which covers the exploration and utilization of mineral resources, applies to NORM industries. A very important criterion in this regard is whether or not the activity concentration of any radionuclide in the uranium or thorium decay series exceeds 1 Bq/g in the ore, intermediate products or tailings (slag or other residues). The specific regulatory procedure is for any new project or activity which falls into the above mentioned scope and for which the activity concentration criterion of 1 Bq/g is exceeded, the operational organization should prepare two documents. The first document is a special report for the assessment of the radiological environmental impact and is required at the planning stage of the project or before construction or beginning of activities. The second document is a special report for monitoring and acceptability of the radiological environmental impact and is required at the stage of operation. Both documents need to be submitted to the relevant environmental protection agency for approval.

3. EXPOSURE TO NATURAL SOURCES

3.1. Exposures

The normalized collective effective dose from airborne discharges from a coal fired power plant to members of the public within 80 km was found to be 16.5 man Sv/GWa. For a coal gangue power plant, the corresponding value was 7000 man Sv/GWa. The annual collective effective dose to the public arising from buildings constructed with bone coal bricks countrywide was 3300 man Sv/a.

TABLE 1. LIST (FIRST BATCH) OF REGULATIONS ON RADIOLOGICAL ENVIRONMENTAL IMPACT FOR EXPLORATION AND UTILIZATION OF MINERAL RESOURCES

	Industrial activities
1. Rare earths, including monazite, bastnaesite, xenotime and rare earth ion absorption clays	Ore mining, ore dressing and smelting
2. Niobium and tantalum	Ore mining, ore dressing and smelting
3. Zirconium and zirconium oxide	Ore mining, ore dressing and smelting of zircon sand and baddeleyite
4. Vanadium	Ore mining and smelting
5. Anthracite	Coal mining and utilization

Occupational exposures in mines over the period 1996–2000 are shown in Table 2.

TABLE 2. OCCUPATIONAL EXPOSURES IN MINES

	Annual av. No. of monitored workers	Annual collective effective dose (man Sv)	Av. individual effective dose (mSv/a)
Coal mines	6 500 000	14 600	2.40
Metal mines	1 000 000	5 530	5.53
Other mines	3 000 000	2 060	0.69
Total or av.	10 500 000	220 000	2.1

Indoor radon concentrations measured in various cities in China in past years are shown in Table 3.

TABLE 3. INDOOR RADON CONCENTRATIONS

Study	No. of cities	No. of samples	Av. radon concentration (Bq/m ³)
1	28		43.8
2	8	2808	34.1
3	9	1994	31.7

3.2. The first China pollution sources survey

The First China Pollution Sources Survey (FCPSS) was a nationwide comprehensive survey for the target year 2007 of all kinds of pollution sources from industry, agriculture, daily life and pollution treatment centres. The survey was organized by the MEP from 2006 to 2009. For non-uranium mines, 11 types of mines were surveyed during the FCPSS, which included mines for rare earths, niobium–tantalum, zircon and its oxides, tin, lead–zinc, copper, aluminum, vanadium, iron and steel, phosphate and coal (including coal gangue).

The FCPSS included about 1433 companies producing ores, concentrates or wastes (slag and tailings). These 1433 companies comprised 876 mining companies producing 267 million t of ore and 587 mineral processing companies processing 191 million t of raw materials. The mineral processing companies generated 171 million t of solid waste and 831 million t of wastewater, of which 24.6 million t had a gross alpha activity exceeding 1 Bq/L or a gross beta activity exceeding 10 Bq/L.

About 773 companies (53.9% of the total) were located in five provinces: 310 companies in Shanxi; 143 companies in Sichuan; 131 companies in Hunan; 107 companies in Yuannan; and 82 companies in Chongqing. Most of the coal, non-ferrous and ferrous metals are produced in these five provinces. The distribution of companies according to different kinds of mining and milling was as follows: 819 companies (44.8%) were coal producers (including coal gangue); 142 companies (8.9%) were iron producers; and 97 companies (6.1%) were lead–zinc producers. The full distribution of enterprises included in the FCPSS is shown in Fig. 2.

The mineral resources associated with higher radioactivity levels are rare earths, niobium–tantalum and zircon. The average gamma dose rate was 5.709 $\mu\text{Gy/h}$ for rare earths, 3.263 $\mu\text{Gy/h}$ for niobium–tantalum and 1.592 $\mu\text{Gy/h}$ for zircon. The average concentration of ²³⁸U, ²²⁶Ra or ²³²Th in these mineral resources exceeds 1 Bq/g. However, other industries — coal mining, coal

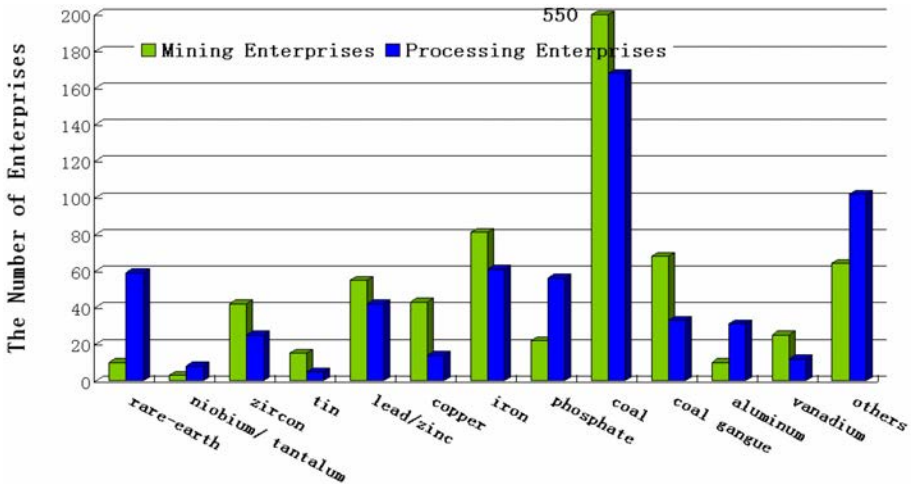


FIG. 2. Distribution of enterprises included in the survey.

utilization and phosphate — are also considered to have significant radiological impact because of their residues. Generally, activity concentrations in NORM solid residues tend to be similar to those in mineral resources.

Radioactivity levels in various mineral resources and solid residues are shown in Tables 4 and 5, respectively.

For uranium, there were 117 companies in which the activity concentrations of their ores or raw materials exceeded 1 Bq/g, and they account for 8% of the total non-uranium companies. The annual production of ores and raw materials in these companies is 5.9 million t and the amount of raw materials used is 2.5 million t. The amounts of solid residue generated are 82 000 t in niobium–tantalum processing, 1.44 million t in rare earths processing and 59 000 t in zircon processing.

For ^{232}Th , there were 68 companies in which the activity concentrations of their ores or raw materials exceeded 1 Bq/g, and they account for 4.6% of the total non-uranium companies. The annual production of ores and raw materials in these companies is 0.61 million t and the amount of raw material used is 6.44 million t. The amounts of solid residue generated are 1.44 million t in rare earths processing and 82 000 t in niobium–tantalum processing.

For ^{226}Ra , there were 123 companies in which activity concentration of their ores or raw materials exceeded 1 Bq/g, and they account for 8.4% of the total non-uranium companies. The annual production of ores and raw materials in those companies is 2.01 million t and the amount of raw material used is

TABLE 4. RADIOACTIVITY IN MINERAL RESOURCES

Element or mineral	Activity concentration (Bq/g)						Gamma dose rate (μGy/h)	
	U		Ra-226		Th-232		Av.	Max.
	Av.	Max.	Av.	Max.	Av.	Max.		
Rare earths	3.972	78	2.529	30.2	5782	137	5.709	32.671
Nb-Ta	4.476	21.5	18.131	57.486	2.015	9.242	3.263	8.023
Zircon	1.289	6.5	3.51	13.935	1.733	10.2	1.592	5.830
Sn	0.218	0.778	0.540	4.276	0.133	0.872	0.272	0.838
Pb-Zn	0.649	34.749	0.465	16.274	0.069	2.729	0.173	1.029
Cu	0.142	1.065	0.163	0.874	0.034	0.183	0.170	0.493
Fe	0.270	6.978	0.288	14.265	0.068	2.597	0.162	1.234
Phosphate	0.396	2.735	0.404	2.072	5	0.346	0.273	1.241
Coal	0.383	167.403	0.212	24.021	0.051	0.910	0.153	2.552
Coal gangue	0.171	1.321	0.118	0.682	0.082	0.241	0.135	0.242

TABLE 4. RADIOACTIVITY IN MINERAL RESOURCES (cont.)

Element or mineral	Activity concentration (Bq/g)						Gamma dose rate ($\mu\text{Gy/h}$)									
	U		Ra-226		Th-232		Av.	Max.								
Al	Av.	0.482	Max.	1.220	Av.	0.289	Max.	0.798	Av.	0.24	Max.	0.638	Av.	0.323	Max.	0.910
V		1.036		12.2		0.908		3.98		1.501		27		0.280		0.968
Others		0.503		5.029		0.744		8.048		0.508		16.535		0.422		5.940

TABLE 5. RADIOACTIVITY IN SOLID MINERAL RESIDUES

Element or mineral	Activity concentration (Bq/g)						Gamma dose rate (μGy/h)	
	U		Ra-226		Th-232		Av.	Max.
	Av.	Max.	Av.	Max.	Av.	Max.		
Rare earths	2.081	83.044	1.24	53.7	4.876 3	85.6	3.249	48.344
Nb-Ta	7.725	36.31	7.212	34.751	4.191	21.775	1.624	7.634
Zircon	1.026	9.908	0.945	7.202	0.327	2.12	0.358	1.5
Sn	0.922	2.85	1.377	4.35	0.802	3.16	0.601	1.482
Pb-Zn	0.118	0.715	0.195	2.049	0.038 4	0.137	0.13	0.303
Cu	0.142	1.63	0.155	2.38	0.036	0.145	0.153	0.385
Fe	0.246	6.028	0.247	9.908	0.135	2.289	0.189	1.177
Phosphate	0.123	0.724	0.191	1.042	0.035 3	0.202 4	0.144	0.331
Coal	0.225	7.6	0.326	92.178	0.091	0.91	0.162	0.987
Coal gangue	0.191	0.763	0.079	0.415	0.092	0.212	0.115	0.328

TABLE 5. RADIOACTIVITY IN SOLID MINERAL RESIDUES (cont.)

Element or mineral	Activity concentration (Bq/g)						Gamma dose rate ($\mu\text{Gy/h}$)	
	U			Ra-226			Th-232	
	Av.	Max.	Av.	Max.	Av.	Max.	Av.	Max.
Al	0.402	1.398	0.282	0.581	0.349	0.694	0.300	0.615
V	0.813	2.096	0.675	1.692	0.073	0.771	0.264	0.760
Others	0.338	2.968	0.435	4.72	0.119	1.39	0.2	1.18

1.35 million t. The amounts of solid residue generated are 82 000 t in niobium/tantalum processing and 3.40 million t in tin processing.

3.3. Case study: Baiyun Obo mine

Baiyun Obo mine is the largest rare earth deposit in China and has been in operation for more than 50 years. The ores are rich in radioactive elements, containing 0.01–0.05% thorium oxide and 0.0005–0.002% triuranium octaoxide. The ores are refined near Baotou to recover iron and steel along with rare earths.

About 560 million t of waste rock have been produced and are stored on site, 149 million t of tailings are stored in a tailings pond, 55 million t of ferrous slag are stored in a slag dump and 437 300 t of rare earth slag are stored in a radioactive waste storage facility. Liquid effluent is treated and reused. Waste gas is treated and discharged and some iron slag has been used as a building material.

The background radiation level in Baotou city is about 0.065 $\mu\text{Gy/h}$. Higher radiation levels are found in areas of the Baotou iron and steel plant, the rare earths plant, and at a NORM residue site. There is an area of about 7 km^2 where the typical radiation level is in the range 0.5–1 $\mu\text{Gy/h}$, with a maximum of 1.518 $\mu\text{Gy/h}$. In the central area of the rare earths plants, where the radiation level is in the range 0.2–0.6 $\mu\text{Gy/h}$, a few hotspots were found. In the slag stockpile, the radiation level is in the range 0.6–2 $\mu\text{Gy/h}$. At the tailings pond without water cover, the radiation level is 0.65–1.2 $\mu\text{Gy/h}$, with a maximum of 1.316 $\mu\text{Gy/h}$.

In the upper 20 cm layer of contaminated soil, the thorium activity concentration 0.05–0.35 Bq/g.

A preliminary dose assessment for external gamma exposure of workers gave doses in the range 0.29–0.61 mSv/a. For internal exposure to ^{220}Rn progeny, the doses were 1.05–5.27 mSv/a. For members of the public, external gamma exposure at the southern tailings pond gave a dose of 0.043 mSv/a. For indoor exposure in buildings constructed from normal materials, the dose was 1.86 mSv/a. For buildings constructed from slag, the dose was 2.0 mSv/a. For internal exposure to outdoor ^{220}Rn progeny, the dose was previously 1.26 mSv/a, but has fallen recently to 0.02 mSv/a. Inhalation of airborne dust from the tailings pond may cause internal exposure.

4. DISCUSSION AND CONCLUSIONS

Exposure to natural sources as a result of human activities is becoming the major cause of additional exposure of both the public and workers. Today, there are about 10 million people working in mining in China, and 100 million people living in houses with elevated concentrations of radon.

Exposure to natural sources has become an urgent issue. From the case study at Baiyun Obo mine, it has been shown that the additional dose received by workers from external gamma exposure is 0.24–1.0 mSv/a. If the inhalation of aerosols and dust containing thorium is considered, the dose probably exceeds 1.0 mSv/a. For members of the public, indoor radiation levels are higher in buildings using slag residues. Internal exposure control is quite important. Operational organizations are to take measures to lower the exposure of workers, and governments are to take some measures to lower the public exposure. In cases of high concentrations of indoor radon, remedial measures are to be undertaken.

It is necessary to strengthen regulatory control of non-uranium mines. The regulatory body should consider developing appropriate regulations and rules.

For NORM residues, the management system needs to be improved and NORM residue management rules need to be developed.

The Environmental Protection Department should consider implementing measures for regular supervision of the working sites and surrounding areas to ensure the safety of the workers and the control of waste streams.

Radiation safety training courses should ideally be implemented regularly for employees in production and management departments to strengthen the knowledge of safe work practices and radiation protection.

PREPARING THE BUILDING INDUSTRY FOR THE NEW EURATOM BSS

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Abstract

In the proposed new Euratom Basic Safety Standards (EU-BSS), a reference level of 1 mSv/a is set for indoor external exposure to gamma rays emitted by building materials. An activity concentration index (ACI) is proposed as a conservative screening parameter for identifying building materials that may cause the reference level to be exceeded. An alternative in situ ACI determination method was developed and the validation of this method is discussed. This screening method was used to analyse numerous building materials available on the Belgian market. An attempt was made to identify building materials containing residues from naturally occurring radioactive material processing industries that pass through Belgium, by analysing results from a large scale radiological study of the container traffic in the port of Antwerp. In addition to the regulation provided on the gamma exposure from building materials, specific parts of the proposed new EU-BSS are dealing with radon issues separately. A reference level of 300 Bq/m³ for indoor radon concentration is proposed for the EU-BSS. Relationships between radon exposure and type of building material used, the airtightness of the building and the stage of construction work were determined.

1. INTRODUCTION

The European Commission is in the process of recasting the Euratom Basic Safety Standards (EU-BSS), which provide for the control of exposure of the public and workers to radioactivity [1]. The recasting process is nearing its conclusion and adoption by the Council is expected by the end of 2013.

The proposed new EU-BSS in their current form (draft version April 2013) provide for the regulation of building materials incorporating residues from NORM processing industries, such as fly ash, phosphogypsum, phosphorus/tin/copper slag and red mud, as well as building materials with of natural origin. In the proposed new EU-BSS, the reference level for indoor external exposure to gamma emissions from building materials is 1 mSv/a. For verification of this

reference level, the following screening parameter in the form of an ‘activity concentration index’ (ACI) is proposed:

$$ACI = \frac{C_{Ra-226}}{0.3 \text{ Bq/g}} + \frac{C_{Th-232}}{0.2 \text{ Bq/g}} + \frac{C_{K-40}}{3 \text{ Bq/g}} \quad (1)$$

where C_{Ra-226} , C_{Th-232} and C_{K-40} are the concentrations (Bq/g) of ^{226}Ra , ^{232}Th and ^{40}K , respectively.

It is stated in the proposed new EU-BSS that the ACI relates directly to the incremental gamma dose in a building constructed from a specified building material (i.e. the dose above the dose typically received from outdoor exposure). The ACI applies to the building material, not to its constituents, except when those constituents are building materials themselves and are separately assessed as such, in which case an appropriate partitioning factor needs to be applied. The ACI is used as a conservative screening parameter. If the $ACI \leq 1$, the building material can be marketed without any restrictions associated with the radionuclide content. If the $ACI > 1$, a detailed dose assessment is required to demonstrate that the 1 mSv/a reference level is not exceeded. The dose assessment needs to take into account factors such as density, thickness of material and factors relating to the type of building and the intended use of the material (bulk or superficial). Once the proposed new EU-BSS is approved and becomes adopted in national legislation, cost efficient techniques for the measurement of activity concentration in an industrial context will be needed to support the building industry in the determination of the ACI.

NORM residues such as fly ash produced in large quantities from coal burning, slags from steelworks and metal recycling industries, phosphogypsum from the phosphate industry and red mud from the bauxite processing industry are being investigated for use in building materials. In Table 1, a summary of European and worldwide production is given for some NORM residues that can be used as constituents of building materials. In addition, Table 1 provides a proposal for the codification of the listed NORM residues in the European Union Waste Catalogue. The use of NORM residues in the production of new types of synthetic building materials raises concerns among authorities, the public and the scientific community on the potential increase in gamma exposure of building occupants and on indoor air quality when considering radon and other chemical hazards.

Specific parts of the proposed new EU-BSS deal with indoor radon exposure separately from exposure related to gamma exposure of occupants. The proposal requires that Member States establish a national action plan addressing the management of long term risks from radon exposure in dwellings, buildings

TABLE 1. NORM RESIDUES WITH POTENTIAL FOR USE IN BUILDING MATERIALS

	Source of residue	Estimated annual amount (million t)	EU Waste Catalogue codification	Proposed EU Waste Catalogue codification
Fly ash	Coal fired power generation	44 (Europe) [2]	001 02 or 10 01 16	
Slag and bottom ash	Coal fired power generation	8 (Europe) [2]	10 01 01 or 10 01 14	
Phosphorous slag	Thermal phosphorus production	—	06 09 02	
Phosphogypsum	Phosphoric acid production	180 (worldwide) [3]	—	
Red mud	Bauxite processing (alumina production)	120 (worldwide) ^a	01 03 07	
Slag	Primary iron production	260–310 (worldwide) [4]	10 02 02	
Slag	Steel and lead smelting	130–210 (worldwide) [4]	10 04 01	
Slag	Primary and secondary copper production	24.6 (worldwide) [5]	10 06 01	
Slag	Primary and secondary tin production	—	—	
Pyro and hydrometallurgy residues	Production of platinum group metals and rare earth elements	—	—	

^a See <http://www.redmud.org/Disposal.html> (accessed on 14 May 2013).

with public access and workplaces for any source of radon ingress, whether from soil, building materials or water. A reference level for indoor radon concentration of 300 Bq/m^3 is proposed. If the radon concentration exceeds the reference level, the necessary technical and financial means have to be employed to reduce it.

2. MEASUREMENT EQUIPMENT AND METHODS

2.1. Portal monitor study in the port of Antwerp

A large scale radiological study of container traffic in the port of Antwerp was conducted during the period 23 April 2007 to 31 August 2010 by means of portal monitors. The alarm level of the portal monitors was set at five times the standard deviation of background variations. More details on the instrumental method used can be found in Refs [6–8].

2.2. In situ and laboratory based measurements to determine the ACI of building materials

Geometry corrected in situ measurements were performed to determine the ACI of building materials with an $\text{LaBr}_3(\text{Ce})$ probe. The experimental method used for the analysis of the building materials is discussed in more detail in Ref. [9]. A detailed comparison of the properties of $\text{LaBr}_3(\text{Ce})$ and $\text{NaI}(\text{Tl})$ probes is given in Ref. [10]. The results of the in situ method were compared with laboratory analyses of the samples using a high purity germanium (HPGe) detector for 21 days in a sealed container to achieve secular equilibrium before measurement while, for the in situ measurements with the $\text{LaBr}_3(\text{Ce})$ probe, this was not the case.

2.3. Indoor radon measurements

A study was performed to determine the relationships between radon concentration and the type of building material, the airtightness of the building and the stage of construction work. During the winter of 2012, integrating passive radon detectors were exposed for three months and indoor radon concentrations were measured over a period of three days using a Sun Nuclear 1029 continuous radon monitor and a Sarad Radon Scout PMT. Airtightness measurements were performed with a Minneapolis blower door system and supporting Tectite software. The V50, v50 and n50 values were used to quantify the airtightness. The experimental approach is described in more detail in Ref. [9].

3. RESULTS AND DISCUSSION

3.1. Results for in situ determination of the ACI of building materials

A new industrially useful protocol for measurement of the ACI was developed to assess the applicability of newly developed materials for the European building market. The applicability of an in situ measurement methodology was investigated with the specific objective of aiding the industry in its search for cost efficient measurement techniques. An important aspect of the investigation was the validation of the so called B-NORM method that had been developed. A good correlation was found between the B-NORM in situ method and the certified laboratory based analysis using an HPGe detector (see Fig. 1). The slope of the regression line in Fig. 1 is 0.95 ($r^2 = 0.98$), indicating that the B-NORM method can provide a good estimate of the ACI for the materials within the measured range of values.

During the measurement campaign of the B-NORM investigation, the highest ACI found (for a natural stone product) was 3.65 when determined by the in situ method and 3.91 when determined by the laboratory method (see Fig. 1). This means that, on implementation of the proposed new EU-BSS in Belgium, a more detailed dose assessment will be required to determine whether the dose is below the 1 mSv/a reference level before allowing this natural stone to be put on the Belgian market for use in dwellings. A more detailed discussion of the results is given in Ref. [9].

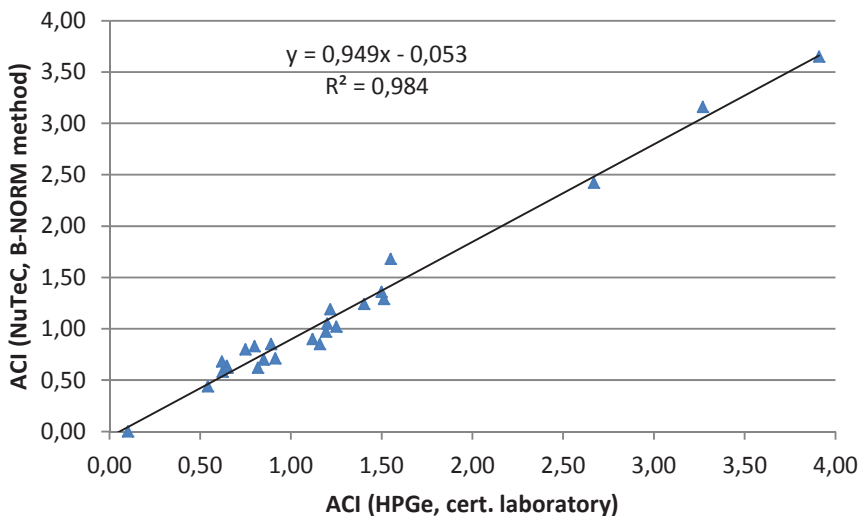


FIG. 1. Comparison of ACI determined by the B-NORM in situ method and by the certified laboratory based HPGe method for 14 tiles, 11 natural stone products and 1 gypsum product.

3.2. Database of building materials and potential NORM constituents

Figure 2 gives a summary of the results from a large scale portal monitor study at the port of Antwerp. Alarms at the detection portals (103 600 events) were attributed to various categories of materials on the basis of the shipping information. The database was compiled during the Megaports project [6] and the NuTeC-NORM project [8] by combining several databases with in-house measurement information. All percentages considered in the discussion below refer to percentages of the 103 600 events.

Figure 2 indicates that 23% of the events were attributed to the category ‘stones and tiles’ that contained, among other things, bricks, slate, sandstone, natural stone, a wide variety of tiles, paving stones and marble. Since not all of these subcategories were well defined on the basis of recorded shipping information, a quantitative assessment of the subcategories was difficult. It was found that 1% of the events could be assigned to the category

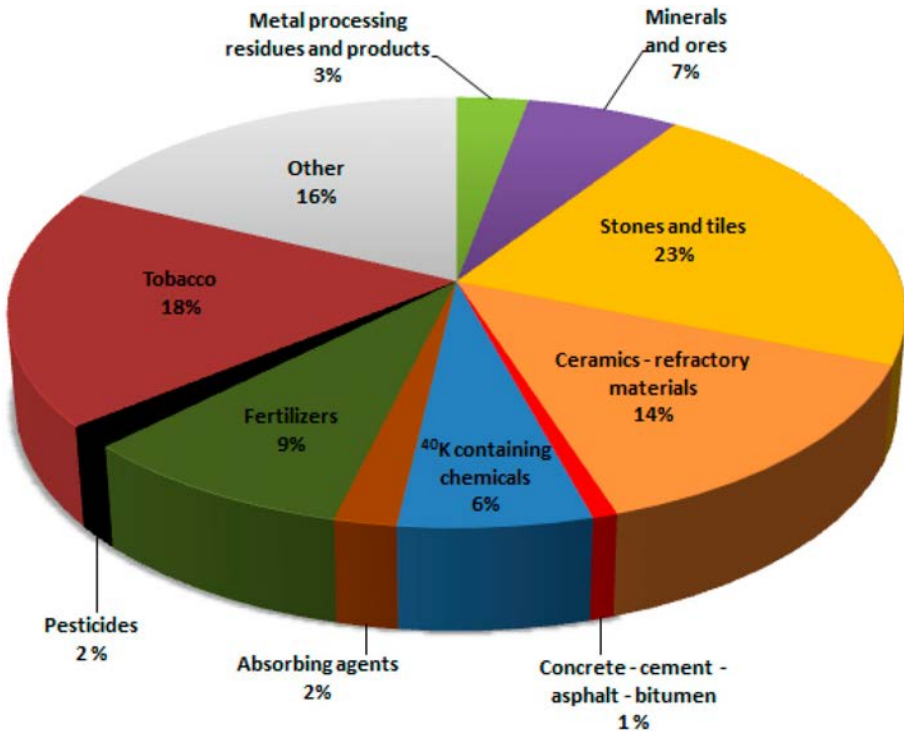


FIG. 2. NORM database relating to 103 600 alarms at detection portals, classified according to shipping information, for the period 23 April 2007 to 31 August 2010.

‘concrete-cement-asphalt-bitumen’ and 14% of events to the category ‘ceramics-refractory materials’. Furthermore, it should be noted that about 1% of events in the category ‘other’ were classified as “building materials” (without further specification) and that it was unclear if these events needed to be attributed to the category ‘stones and tiles’ or one of the other categories. A more detailed description of the database and its different categories is given in Ref. [8] and more detailed publications are being prepared.

3.3. Indoor radon measurements

A group of six neighbouring dwellings in Neeroeteren, Belgium, were selected for this study. The soil in this area consists of moist sand. At the time of the experiment, the dwellings were at different stages of construction (see Fig. 3). This resulted in different levels of airtightness. During the study, no one entered the building. In this way, differences in radon concentration due to external parameters were excluded. In Dwelling 1, the floor screed and plastering of the walls had been completed; while in Dwellings 2 and 3, the plastering was complete but no screed had been applied. In Dwellings 4, 5 and 6, neither the plastering nor the screed had been applied.

The results are presented in Table 2. The indoor radon concentration is clearly higher in buildings with a better airtightness. Both parameters seemed to be inversely proportional, as shown in Fig. 4. These results confirm the importance of a good, functioning ventilation system, especially in dwellings with limited natural ventilation such as low energy and passive dwellings. In this way the radon risk can be minimized.



FIG. 3. Various construction stages of the dwellings.

TABLE 2. RESULTS OF INDOOR RADON MEASUREMENTS

	Dwelling					
	1	2	3	4	5	6
Screed	Yes	No	No	No	No	No
Plaster	Yes	Yes	Yes	No	No	No
n50 (h ⁻¹)	3.28	8.79	9.40	31.86	27.93	29.68
v50 (m ³ ·h ⁻¹ ·m ⁻²)	3.69	8.34	9.68	32.50	26.46	33.89
Radon concentration (Bq/m ³)	57	48	29	13	12	26

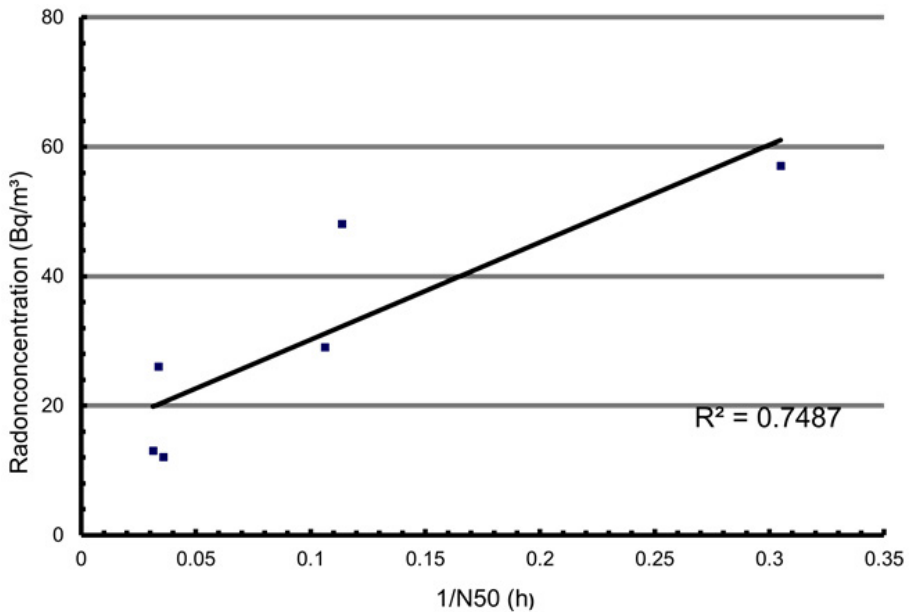


FIG. 4. Inverse proportionality of radon concentration to airtightness.

The contribution of radon to the radiation exposure is not fully accounted for in the ACI used in the proposed new EU-BSS. It is shown in Refs [11–13] that the assessment of the radon situation in dwellings is a multidimensional problem to solve. In houses, distinguishing between radon emanation from the soil and radon from the building materials is not straightforward and therefore the

approach of the proposed new EC-BSS in dealing with radon issues separately from exposure related to gamma exposure is supported.

4. CONCLUSIONS AND OUTLOOK

4.1. Conclusions

An alternative, in situ ACI determination method has been developed and validated. This method was used to analyse various building materials available on the Belgian market and can serve as a screening tool to support the industry when the proposed new EU-BSS are implemented.

A NORM database of building materials has been established, which gives an idea of which materials, both those of natural origin and those incorporating NORM residues, pass through Belgium. However, owing to limitations on the quality of the shipping information, a systematic investigation of such building materials could not be carried out.

In the proposed new EU-BSS, a reference level for indoor radon is established in addition to the reference level for gamma exposure from building materials. The relationship between indoor radon concentration and airtightness of buildings was investigated and confirmed the importance of a good, functional ventilation system to assure that indoor radon concentrations stay well below the reference level proposed in the EU-BSS.

4.2. Outlook

The depletion of energy resources and raw materials has a huge impact on the building market. In the development of new synthetic building materials, the use of various industrial residues becomes highly desirable. Therefore a new 'COST' network proposal has been submitted to stimulate the collaboration of scientists, industry and regulators in the development of knowledge, experience and new technology to stimulate research on the use of NORM residues in tailor made building materials in the construction sector, while considering the impact on both external gamma exposure of building occupants and indoor air quality. By improving radiological impact assessment models for the use of NORM residues in building materials, the proposed COST action aims to further stimulate justified uses of NORM residues in different types of newly developed building materials. Based on these models, the proposed COST action aims at investigating realistic legislative scenarios so that the authorities concerned can allow uses of NORM residues that are acceptable from a radiation protection

point of view in accordance with the Lead Market Initiative and sustainable construction. The COST proposal is awaiting final approval in May 2013.

ACKNOWLEDGEMENTS

Funding for the research projects mentioned in this paper was provided by the Ministry of the Flemish Community (Hermesfonds: 45% of the NuTeC-NORM and B-NORM projects), EFRO (40% of the NuTeC-NORM and B-NORM projects) and LRM. In addition, the authors would like to acknowledge the contribution and support from P. Fias (Controloatom), F. Biermans and T. Peeters (Federale Overheidsdienst Financiën), M. Bruggeman (SCK-CEN), A. Poffijn and S. Pepin (FANC), S. Verhoeven, S. Schepens, P. Vannitsen, L. Hulshagen, C. Vandervelpen and T. De Clerck (XIOS Hogeschool Limburg).

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PROGRESS IN THE REGULATION OF RADIATION ENVIRONMENTAL SAFETY IN THE UTILIZATION OF MINERAL RESOURCES IN CHINA

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Abstract

In the paper, the background to the progress made in the regulation of radiation environmental safety in the utilization of mineral resources other than uranium (thorium) ores in China is first introduced. The regulatory experience of radiation environmental safety in the utilization of mineral resources in international organizations and industrial countries (e.g. International Commission on Radiological Protection, IAEA, European Union and United States of America) is surveyed and analysed. Finally, the scope and requirements of regulation on the radiation environmental safety in the utilization of mineral resources in China are stated.

1. INTRODUCTION

In recent years, the regulation of radiation environmental safety in the utilization of mineral resources other than uranium and thorium ores (also referred to internationally as NORM) has attracted widespread attention both in the international organizations and in most countries. It has been found that, during the mining and processing of mineral resources such as rare earth ore, niobium–tantalum, zircon, non-ferrous metal, oil and gas, and coal, radionuclides in the uranium (and thorium) decay series may be enriched in the products, residues and waste in the workplace, leading to enhanced natural radiation levels and radioactive contamination of the environment. Although there are differences in regulatory approach for radiation environmental safety in the utilization of mineral resources, for instance differences in clearance levels and scope of regulation, between the International Commission on Radiological Protection (ICRP), the IAEA, the European Union and the United States of America, the urgency and necessity for regulation of radiation environmental safety have been widely recognized [1].

The Government of China had recognized the problem of radiation environmental safety in the utilization of mineral resources other than uranium (thorium) ores since the 1990s. In the People's Republic of China Law on the Prevention and Control of Radioactive Pollution, implemented in 2003, the radiation environmental safety in the utilization of mineral resources has already been embodied in the scope of regulation of radioactive pollution prevention and control [2]. The law requires that those intending to exploit radioactive minerals other than uranium (and thorium) are, before applying for a mining licence:

- (a) To draw up an environmental impact report and submit it to the competent environmental protection administrative department of the people's government at or above the provincial level, for examination and approval;
- (b) To design, construct and put into operation integrated radioactive pollution prevention and control facilities at radioactive mines at the same time as the main part of the project;
- (c) To construct tailings repositories for the storage and disposal of the tailings produced during the operation of radioactive mines; such tailings repositories are to conform to radioactive pollution prevention and control requirements.

The People's Republic of China Law on the Prevention and Control of Radioactive Pollution has established the legal foundation for the regulation of radiation environmental safety. However, there are some problems with the enforcement of the law, for example the scope and requirements of regulation on the radioactive mines are not explicit, leading to difficulties in implementation which need to be solved for radiation environmental safety.

In China, the first national survey of pollution sources in 2007–2009 by the Ministry of Environmental Protection indicated the following:

- (a) In over 300 mining and mineral processing enterprises, which are classified into 11 groups such as rare earth ore, niobium–tantalum, among others, the concentration of natural radionuclides contained in the mineral products and waste has exceeded the international activity concentration criterion of 1 Bq/g for application of the requirements for planned exposure situations.
- (b) The solid waste discharged each year reaches millions of tonnes.
- (c) The regulation of radiation environmental safety in the utilization of mineral resources is inconsistent [3].

In recent years, several radiation environment disputes have already taken place in China. Consequently, it is necessary to make the scope and requirements clear for the regulation of radiation environmental safety in the utilization of mineral resources as soon as possible.

2. INTERNATIONAL SURVEY OF THE REGULATION OF RADIATION ENVIRONMENTAL SAFETY IN THE UTILIZATION OF MINERAL RESOURCES

2.1. International Commission on Radiological Protection

In the system of radiation protection of humans recommended in ICRP Publication 103, The 2007 Recommendations of the International Commission on Radiological Protection [4], the Commission recognizes three types of exposure situations: planned exposure situations; emergency exposure situations; and existing exposure situations. For existing exposure situations, the Commission recommends that reference levels (restrictions on individual doses) are applied together with the principle of optimization of protection, and the exposure below the reference level cannot be ignored. According to ICRP Publication 104, Scope of Radiological Protection Control Measures [5], the regulation of industrial activities involving materials with activity concentrations not exceeding 1 Bq/g for uranium and thorium series radionuclides (10 Bq/g for ^{40}K) is not warranted.

2.2. IAEA

IAEA Safety Standards Series No. RS-G-1.7, Application of the Concepts of Exclusion, Exemption and Clearance [6], provides values of activity concentration for radionuclides of natural origin below which it is usually unnecessary to regulate, that is 1 Bq/g for uranium and thorium series radionuclides and 10 Bq/g for ^{40}K . This approach has been maintained, unchanged, in IAEA Safety Standards Series No. GSR Part 3 (Interim), Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards.¹

Safety Reports Series No. 49, Assessing the Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials [8], identifies the following industry sectors (in addition to the mining and processing of uranium

¹ INTERNATIONAL ATOMIC ENERGY AGENCY, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, IAEA Safety Standards Series No. GSR Part 3 (Interim), IAEA, Vienna (2011). [Editor's note] This has been superseded by Ref. [7].

ore) most likely to require regulatory consideration, roughly in descending order of priority:

- (1) Extraction of rare earth elements;
- (2) Production and use of thorium and its compounds;
- (3) Production of niobium and ferroniobium;
- (4) Mining of ores other than uranium ore;
- (5) Production of oil and gas;
- (6) Manufacture of titanium dioxide (TiO₂) pigments;
- (7) The phosphate industry;
- (8) Zircon and zirconia industries;
- (9) Production of tin, copper, aluminium, zinc, lead, and iron and steel;
- (10) Combustion of coal;
- (11) Water treatment.

2.3. European Union

The European Commission's draft Euratom Basic Safety Standards Directive [9] identifies the regulation system based on a graded approach, also taking the list of industrial sectors involving NORM into account:

- (1) Extraction of rare earths from monazite;
- (2) Production of thorium compounds and manufacture of thorium containing products;
- (3) Processing of niobium–tantalum ore;
- (4) Oil and gas production;
- (5) Geothermal energy production;
- (6) TiO₂ pigment production;
- (7) Thermal phosphorus production;
- (8) Zircon and zirconium industry;
- (9) Production of phosphate fertilizers;
- (10) Cement production, maintenance of clinker ovens;
- (11) Coal fired power plants, maintenance of boilers;
- (12) Phosphoric acid production;
- (13) Primary iron production;
- (14) Tin/lead/copper smelting;
- (15) Groundwater filtration facilities.

2.4. United States of America

In the United States of America, neither the Environmental Protection Agency (EPA), which has primary responsibility for setting federal radiation standards for environmental protection, nor any other Federal agency with responsibility for regulating radiation exposures has developed standards applicable to all exposure situations that involve NORM. The Conference of Radiation Control Program Directors has issued a model regulation, which has been adopted as a guide for the regulation of NORM in some individual states.

In the 1990s, the EPA investigated mining and mineral processing waste with relatively high natural radioactive levels; the waste investigated included the following types:

- (1) Uranium overburden;
- (2) Phosphate waste, including phosphogypsum and slag;
- (3) Phosphate fertilizers;
- (4) Coal combustion residues, comprising fly ash, bottom ash and slag;
- (5) Oil and gas scale and sludge;
- (6) Water treatment sludges and resins containing radium;
- (7) Metal mining and processing waste associated with rare earths, zircon, hafnium, titanium, tin and large volume industries such as copper and iron and steel;
- (8) Geothermal energy production wastes.

To sum up, the utilization of mineral resources is an important aspect contributing to a rise in the environmental radiation level. Among international bodies, the regulation of radiation environmental safety in the utilization of mineral resources is still being investigated. There are differences in regulatory requirements, such as exemption levels and the industry sectors needing regulatory consideration. Currently, the international community mostly adopts the IAEA international standards, which state that material containing radionuclides of natural origin at an activity concentration of less than 1 Bq/g for any radionuclide in the uranium and thorium decay chains is outside the scope of regulation, in line with the objective of making optimum use of regulatory resources. In order to judge whether a specific enterprise has a problem of radiation environmental safety, the international bodies and individual States tend to adopt an approach based on a list of industrial activities that may require regulation. The industries where there is a risk of radiation environmental safety are included in the list. Any facility which is included in the list needs to inform the regulatory body of its intended activities and, according to the details of the activities and the results

of a dose assessment, the regulatory body decides whether the intended activities can be exempted from regulation.

3. PROGRESS IN THE REGULATION OF RADIATION ENVIRONMENTAL SAFETY IN THE UTILIZATION OF MINERAL RESOURCES IN CHINA

On 4 February 2013, the Ministry of Environmental Protection of the People's Republic of China issued a Notice on the Regulatory Directory on the Radiation Environmental Safety in the Utilization of Mineral Resources (the First Batch), in which five industry sectors are listed in the regulatory directory and corresponding requirements for regulation are specified.

3.1. The basis for compiling the regulatory directory

In line with the experience of the international community, the regulatory directory on radiation environmental safety in the utilization of mineral resources is worked out according to the laws and regulations in China, for example the People's Republic of China Law on the Prevention and Control of Radioactive Pollution, the People's Republic of China Law on Environmental Impact Assessment [10], the purpose of which is to protect the environment and public health and promote the sustainable development of the utilization of mineral resources other than uranium (thorium) ores.

3.2. The principles adopted in compiling the regulatory directory

- (a) The industry sectors generally found to contain relatively high concentrations of natural radionuclides are included in the regulatory directory. In China, natural radionuclides in some industry sectors have been investigated and evaluated. Results show that the concentrations of uranium and thorium series radionuclides are commonly high in certain mineral resources (e.g. rare earth ore, niobium–tantalum and zircon), which are the first to be included in the regulatory directory.
- (b) The industry sectors included in the regulatory directory have been developed to a certain degree in China.
- (c) Other industry sectors such as the oil and gas industry, which are included in the lists of industries compiled by bodies such as the IAEA, are not yet included in the regulatory directory because investigation data have not yet been generated in China. But surveys of these industry sectors are

continuing and such industry sectors may be included in the regulatory directory once the investigations have been concluded.

- (d) Industry sectors which directly utilize uranium, thorium and their compounds for raw materials and additives (e.g. the production of welding electrode and gas mantles) are not included in the regulatory directory.

3.3. The regulatory directory

The regulatory directory on radiation environmental safety in the utilization of mineral resources in China is shown in Table 1. According to the demand on the management of radiation environmental safety, the Ministry of Environmental Protection of the People's Republic of China will adjust and publish new editions of this regulatory directory.

3.4. Regulatory requirements

For development projects of mineral resources which have been included in the Regulatory Directory on Radiation Environmental Safety in the Utilization of Mineral Resources (first batch) and in which the concentrations of uranium (thorium) series radionuclides in the materials (including the raw ore, intermediate product, tailings and other residues) exceed 1 Bq/g, the facilities must commission the relevant institution, which is within the environmental impact scope of the nuclear industry, to draw up a special report on the radiation environmental impact and a special report on the inspection and approval of the radiation environment measures taken.

The special report on the radiation environmental impact is to be contained in the environmental impact report of the project, to be drawn up simultaneously and to be submitted together with the environmental impact report. The design of the report is to be in accordance with The Classified Directory of Environmental Impact Report on the Administration of Construction Project, which is promulgated by the Ministry of Environmental Protection. The radiation monitoring on the stages of environmental impact and acceptance is to be commissioned to the monitoring institution having the appropriate qualification in the region.

TABLE 1. THE REGULATORY DIRECTORY ON RADIATION ENVIRONMENTAL SAFETY IN THE UTILIZATION OF MINERAL RESOURCES (FIRST BATCH)

Mineral	Industrial activities
1. Rare earths	Mining, beneficiation and processing of various rare earth ores, including monazite, bastnäsite, xenotime and ion adsorption clays
2. Niobium–tantalum	Mining, beneficiation and processing of minerals containing niobium and/or tantalum
3. Zircon and zirconia	Mining, beneficiation and processing of zircon sand and baddeleyite
4. Vanadium	Mining and processing of vanadium ore
5. Anthracite	Mining and utilization of anthracite

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ENVIRONMENTAL ASPECTS OF NORM

(Topical Session 3)

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Invited Paper

NORM LEVELS IN MINE PIT LAKES IN SOUTH-WESTERN SPAIN

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Abstract

Former mining activities in a pyritic area in south-western Spain have generated mine pits in which underground water and rainwater has accumulated. The accumulated waters have produced oxidation of the pyrite and, consequently, the pit water has become acidic, causing the dissolution of metals and radionuclides of natural origin. The paper discusses the activity concentration levels of uranium isotopes and other radionuclides in water samples and sediments collected from these mine pit lakes. Tributaries of the nearby Odiel River, when crossing the mining area, show low pH values and high concentrations of uranium isotopes due to acid mine drainage. Through the analysis of several isotope activity ratios, the presence of radionuclides in the pit lakes and the influence of these radionuclides on the surrounding area and the Odiel River are evaluated.

1. INTRODUCTION

The Iberian Pyrite Belt is a part of the South Zone of the Hercynian Iberian Massif in the south-western region of the Iberian Peninsula, with pyrite (FeS_2) being the main mineral associated with the volcano sedimentary complex. Mining activities in the area date back to the time of the Roman Empire. Intensive mining activities over the past 150 years generated mine pits in which underground and/or rainwater has accumulated. The accumulated waters have produced oxidation of FeS_2 and, consequently, the pH of the pit water has become acidic (approximately pH2), causing the dissolution of metals and radionuclides of natural origin.

The main objective of this work was to show and discuss the levels found for different radionuclides (mainly isotopes from the uranium series via alpha and gamma spectrometry) in water samples and sediments collected from several mine pit lakes located in the area. The sample collection was carried out in the summer of 2012, and simultaneously several physical and chemical parameters

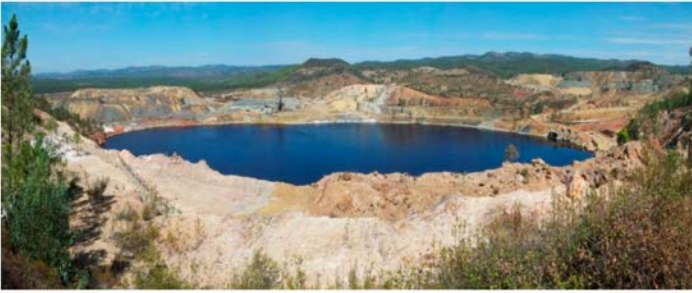
were measured, for use in the resulting discussion. Four pit lakes were selected for this study (see Fig. 1) from the dozens existing in the area, with some of their physicochemical characteristics summarized in Table 1.



Concepción



Confesionarios



San Telmo



La Zarza

FIG. 1. The four pit lakes investigated in this paper.

TABLE 1. CHARACTERISTICS OF THE FOUR PIT LAKES

	Area (km ²)	Volume (hm ³)	Maximum depth (m)	pH	Conductivity at the surface (mS)	Temp. at the surface (°C)
Concepción	1.20	0.4	30	2.71	1.122	24.8
Confesionarios	2.48	0.6	70	2.23	6.752	24.0
San Telmo	14.36	7.0	100	2.54	4.407	28.3
La Zarza	1.87	0.5	40	2.21	8.365	27.1

TABLE 2. SULPHATE AND HEAVY METAL CONCENTRATIONS IN WATER FROM THE FOUR PIT LAKES

	SO ₄ (g/L)	Fe (mg/L)	Al (mg/L)	Mn (mg/L)	Cu (mg/L)	Zn (mg/L)	As (mg/L)	Cd (mg/L)
Concepción	1.4	49	61	11	10	28	23	60
Confesionarios	6	800	290	6	2	7	85	85
San Telmo	3.8	160	121	41	21	89	80	210
La Zarza	16.1	3800	770	250	150	207	1330	275

Source: Ref. [1].

The pit lakes selected for this first study had areas in the range of 2–15 km² and maximum depths 30–100 m. The pH of the water was in the range of 2.3–2.7. In addition, the water of these pit lakes contained in solution a large amount of heavy metals, as shown in Table 2. The concentrations of iron, zinc, copper, arsenic and cadmium in solution are in all cases several orders of magnitude higher than those found in surface waters unaffected by human activities.

The waters of the pit lakes can interact with the environment, and a fraction can be transferred to surrounding aquifers and even to the main river nearby (Odiel River). The streams entering this river, when crossing the mining area, show low pH values and high concentrations of uranium isotopes due to acid mine drainage. The elevated amounts of heavy metals and radionuclides in the

waters entering the Odiel River through tributaries interacting with the mining areas, are maintained in solution along its course and are transported all the way to the estuary where mixing of the acidic water with seawater causes an abrupt change of pH and the precipitation of the contaminants transported in solution. The estuary thus acts as a repository for large amounts of heavy metals and radionuclides, which have caused the extinction of its biodiversity.

2. MATERIALS AND METHODS

To perform this study, superficial waters and superficial sediments along the shore of the pit lakes were collected in the four studied ecosystems: Concepción, Confesionarios, San Telmo and La Zarza. Measurements of the gamma dose rate at 1 m above the surface were performed along the perimeter of the pit lakes in order to evaluate possible increments above the background level in the area. Superficial waters and sediments were also collected in different tributaries of the Odiel River where they cross the mining areas 'infected' with dozens of pit lakes and tailings zones.

In all the waters and sediments collected, the concentrations of uranium isotopes (^{234}U , ^{235}U and ^{238}U), thorium isotopes (^{230}Th and ^{232}Th) and ^{210}Po were determined by the application of the alpha particle spectrometric technique using PIPS detectors. For the performance of these measurements, a radiochemical procedure that sequentially allows the isolation of polonium, thorium and uranium isotopes was applied [2].

The superficial sediments collected in the pit lakes were additionally analysed by low-background gamma spectrometry with hyperpure germanium detectors, in order to determine the concentrations of ^{210}Pb , ^{234}Th , ^{226}Ra and ^{40}K . The procedure employed for sample preparation and efficiency calibration for these measurements can be found in Ref. [3].

External gamma dose rate determinations were performed using a calibrated Berthold LB1236 gas dosimeter coupled to an electronic unit LB1230. This system works under a proportional regime and has a working range of 10^{-2} – 10^4 $\mu\text{Sv/h}$. It is sensitive to photons ranging from 30 keV to 1.2 MeV. The monitor was calibrated for dose rate and energy response at CIEMAT (Madrid, Spain) using ^{60}Co and ^{137}Cs sources, and is periodically subjected to verification checking. The energy dependence of the dosimeter response is around $\pm 20\%$ within the working energy range.

3. RESULTS AND DISCUSSION

The results obtained in the determination of activity concentrations of ^{234}U , ^{235}U , ^{238}U , ^{230}Th , ^{210}Po and ^{232}Th by alpha particle spectrometry in superficial waters collected in the four pit lakes are shown in Table 3. It is evident that some of these waters are particularly enriched in uranium, with activity concentrations exceeding 1 Bq/L, two orders of magnitude higher than the levels in surface waters unaffected by human activities. In addition, all the waters are characterized by a ^{234}U : ^{238}U activity ratio significantly greater than unity, clearly showing the greater tendency of ^{234}U to be dissolved from the original matrix in relation to its parent when in contact with acidic water. The levels of uranium found in these waters are, on the other hand, very well correlated with the conductivity values determined, indicating that the mechanisms affecting the presence of heavy metals and uranium in these waters are the same. In fact, the uranium concentrations are well correlated with the concentrations of dissolved iron that were also found to be elevated.

The influence of the acidification of the water was also evident in the activity concentrations of other radionuclides. The activity concentrations, particularly of ^{230}Th , were quite high in comparison with normal values found in surface waters unaffected by human activities, although the levels were not as elevated as those of uranium. Preferential dissolution of ^{230}Th in comparison with ^{232}Th was also observed, while there was little tendency for ^{210}Po levels to become elevated, even under these special conditions.

TABLE 3. ACTIVITY CONCENTRATIONS IN WATER FROM THE FOUR PIT LAKES

	Conductivity (mS)	Activity concentration (mBq/L)					
		U-234	U-235	U-238	Th-230	Po-210	Th-232
Concepción	1.122	39 ± 5	14 ± 3	1.1 ± 0.5	14 ± 3	<0.5	18 ± 3
Confesionarios	6.752	1100 ± 43	14 ± 5	247 ± 18	133 ± 8	13 ± 1	57 ± 4
San Telmo	4.407	500 ± 17	8.5 ± 1.8	219 ± 11	171 ± 10	<6	39 ± 6
La Zarza	8.365	1750 ± 30	58 ± 4	1110 ± 20	390 ± 20	33 ± 4	122 ± 12

The levels of uranium series radionuclides in the superficial sediments of the pit lakes, as determined by gamma spectrometry (see Table 4), were not elevated above those normally found in sediments unaffected by human activities. The activity concentrations found for ^{234}Th , indicative of the uranium present in these samples, were in general quite low, consistent with the tendency for uranium to dissolve under acidic conditions. Under the very low pH condition of the water, no precipitation of dissolved metals occurs, and there is no associated co-precipitation of radionuclides.

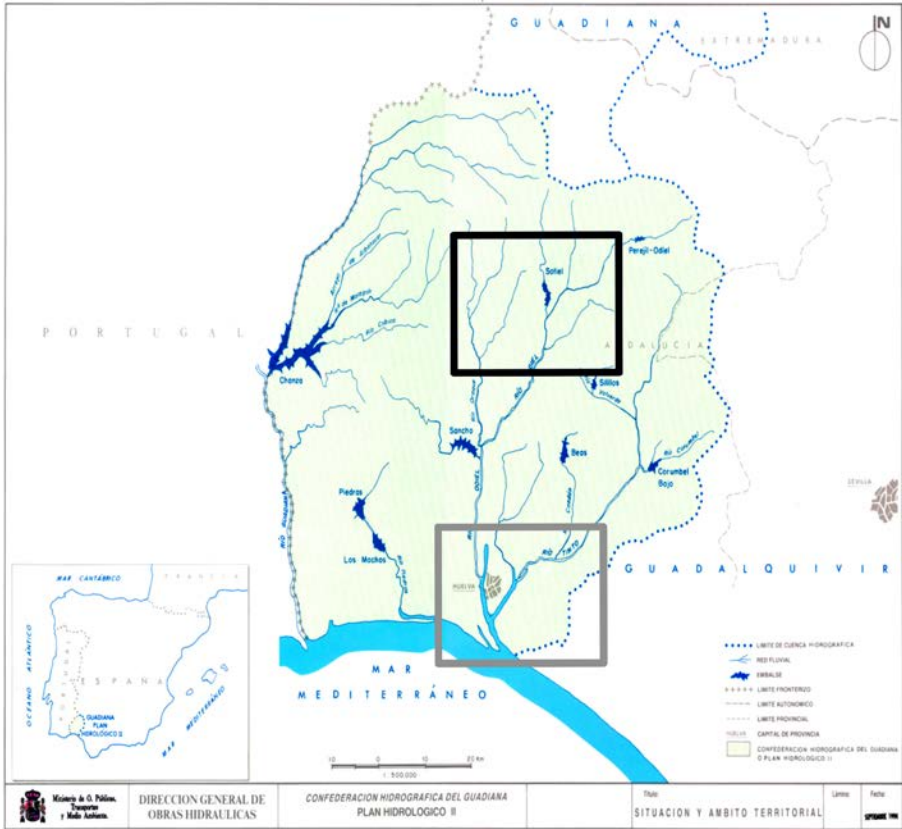
The radiometric results obtained for the lacustrine sediments are, on the other hand, in complete accordance with gamma dose rate measurements performed along the shoreline of the four pit lakes. The average values obtained (which are representatives of a set of at least six measurements in each case) were similar to those typically found in normal terrestrial areas unaffected by human activities:

- Concepción: 0.150 $\mu\text{Sv/h}$;
- Confesionarios: 0.206 $\mu\text{Sv/h}$;
- San Telmo: 0.170 $\mu\text{Sv/h}$;
- La Zarza: 0.185 $\mu\text{Sv/h}$.

The map in Fig. 2 shows the location of the majority of the pit lakes and associated tailings. It can be seen that the mining area is crossed by several small tributaries of the Odiel River, which have been affected by the mining activities. The acid drainage of the pit lakes and associated tailings are contributing to the strong acidification of the waters of the neighbouring tributaries and to the transport by these waters of elevated uranium concentrations. Table 5 shows the activity concentrations in these tributaries, indicating that the influence of the mining area on these tributaries is quite pronounced. The pH values are in the range of 2–3 and the ^{234}U activity concentrations are of the order of 1 Bq/L, similar to those in some of the pit lakes. In fact, the activity concentrations in the waters of the Odiel tributaries reflect the patterns found in the pit lake waters, with ^{234}U : ^{238}U activity ratios clearly greater than unity and with very low ^{210}Po : ^{234}U activity ratios, the latter confirming the extremely low tendency of ^{210}Po to dissolve, even under acidic conditions.

TABLE 4. ACTIVITY CONCENTRATIONS IN SEDIMENTS FROM THE FOUR PIT LAKES

	Activity concentration (Bq/g)						
	Pb-210	Th-234	Ra-226	Th-228	K-40	Cs-137	
Concepción	0.067 ± 0.007	0.007 ± 0.003	0.007 ± 0.003	0.014 ± 0.005	0.053 ± 0.010	<0.000 5	
Confesionarios	0.065 ± 0.006	0.007 ± 0.002	0.012 ± 0.006	0.031 ± 0.010	0.083 ± 0.005	0.000 6 ± 0.000 1	
San Telmo	0.087 ± 0.006	0.045 ± 0.007	0.049 ± 0.008	0.028 ± 0.005	0.195 ± 0.020	<0.000 3	
La Zarza	0.088 ± 0.009	0.008 ± 0.003	0.014 ± 0.004	0.013 ± 0.002	0.095 ± 0.015	<0.000 3	



Note: The area of pit lakes and associated tailings is framed in black, while the estuary of the Odiel River is framed in grey.

FIG. 2. The study area in south-western Spain.

Superficial sediments were collected in the Odiel River tributaries in the same locations as those of the water samples. The results are given in Table 6 and show, from the radioactive point of view, the same pattern as the sediments collected in the pit lakes. In particular, the uranium activity concentrations are quite low and the ^{210}Po : ^{234}U activity ratios are clearly greater than unity. The acidity of the water prevents the precipitation or deposition of the dissolved uranium and even seems to enhance the solubility of some of the uranium present originally in the sediment, as is reflected in the deficiency of uranium in the sediment samples in comparison with ^{210}Po .

NORM LEVELS IN MINE PIT LAKES IN SOUTH-WESTERN SPAIN

TABLE 5. ACTIVITY CONCENTRATIONS IN WATER FROM THE TRIBUTARIES OF THE ODIEL RIVER WHERE THEY CROSS THE MINING AREA

	pH	Activity concentration (mBq/L)			
		U-234	U-235	U-238	Po-210
1	2.76	1239 ± 28	27 ± 1	742 ± 17	1.7 ± 0.4
2	3.07	880 ± 19	23 ± 1	498 ± 11	4.2 ± 1.0
3	2.40	812 ± 16	23 ± 1	628 ± 13	1.8 ± 0.2
4	2.41	139 ± 5	3.7 ± 0.6	69 ± 3	1.2 ± 0.3
5	2.66	1449 ± 36	40 ± 2	886 ± 23	4.1 ± 0.4
6	2.48	844 ± 19	23 ± 1	505 ± 12	5.0 ± 0.9
7	2.72	127 ± 5	4 ± 1	80 ± 3	1.7 ± 0.4

TABLE 6. ACTIVITY CONCENTRATIONS IN THE SEDIMENTS FROM THE TRIBUTARIES OF THE ODIEL RIVER WHERE THEY CROSS THE MINING AREA

	Activity concentration (Bq/g)		
	U-234	U-238	Po-210
1	0.006 4 ± 0.000 4	0.005 0 ± 0.000 4	0.009 0 ± 0.000 4
2	0.005 1 ± 0.000 4	0.005 5 ± 0.000 4	0.008 8 ± 0.000 5
3	0.004 5 ± 0.000 3	0.003 7 ± 0.000 3	0.019 8 ± 0.001 1
4	0.012 4 ± 0.000 7	0.008 0 ± 0.000 5	0.021 7 ± 0.001 1
5	0.007 8 ± 0.000 4	0.005 3 ± 0.000 3	0.023 3 ± 0.001 4
6	0.004 5 ± 0.000 4	0.003 4 ± 0.000 3	0.030 6 ± 0.001 5
7	0.011 2 ± 0.000 6	0.008 0 ± 0.000 5	0.011 7 ± 0.000 6

The acidity in the tributaries affects, in turn, the Odiel River, resulting in a pH value of 3. The enhanced concentrations of uranium and heavy metals remain in solution until the Odiel River reaches the estuary where, on mixing with seawater, the river water becomes neutralized and the dissolved contaminants are precipitated. Thus, the estuary acts as a disposal area for the radionuclides and heavy metals transported from the mining areas 100 km upstream.

The ^{238}U activity concentrations in the superficial sediments collected at various locations in the Odiel estuary over a period of years are shown in Fig. 3. In most of the sediment samples, the concentrations are significantly elevated, sometimes up to 1 Bq/g or more, two orders of magnitude higher than the activity concentrations found in estuaries unaffected by human activities. During the final decades of the twentieth century, uranium contamination in the estuary was associated mainly with the releases of waste from a phosphoric acid plant. These releases were terminated in 1997 but, as is evident from Fig. 3, the enhanced uranium concentrations continued to be observed, while the concentrations of other radionuclides such as ^{226}Ra showed a marked decrease (see Fig. 4).

Uranium-238 activity concentrations in superficial sediment samples collected from the Odiel estuary in the summer of 2011 show that the levels of uranium are still of the same order as those measured in the earlier sampling campaigns. This reinforces the conclusion that the uranium contamination of the estuary has nowadays its origin in the mining activities performed a great distance away and far into the past. It is a clear example of how the radiological

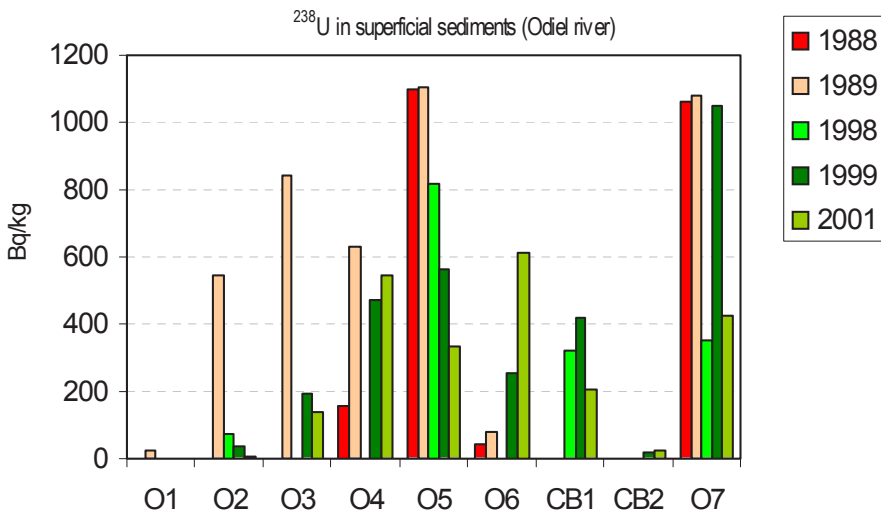


FIG. 3. Activity concentrations of ^{238}U in sediments from the Odiel River estuary.

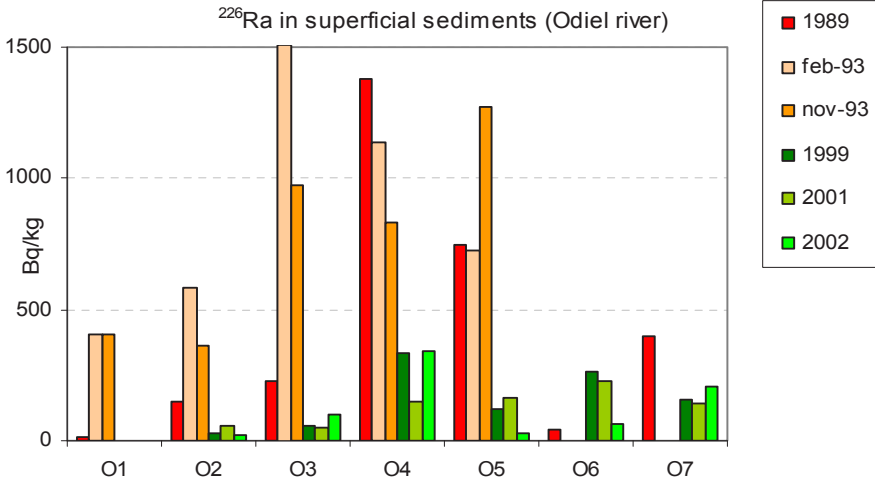


FIG. 4. Activity concentrations of ²²⁶Ra in sediments from the Odiel River estuary.

impact of some human activities can be readily observed in areas far away from the locations at which the radioactivity was released.

4. CONCLUSIONS

The intensive mining in the Pyrite Belt area in south-western Spain during the previous 150 years has left a legacy of dozens of pit lakes containing large amounts of acidic waters enriched in several heavy metals and radionuclides of natural origin.

Some of this acidic water interacts with the surrounding environment, and it is particularly noticeable how it becomes incorporated into the Odiel River via small tributaries that cross the mining area, and thus gives rise to contamination of one of the main rivers in the area.

The Odiel River transports the dissolved heavy metals and radionuclides all the way to the estuary. On mixing with seawater at the estuary, the river water is neutralized and the contaminants are precipitated. The Odiel estuary is therefore serving as a repository for the contaminants liberated by the former mining activities.

ACKNOWLEDGEMENTS

The financial support of the Spanish Nuclear Security Council through an I+D Project (2012–2014) is specially acknowledged. The authors are also very grateful to J. Díaz for his technical assistance in the laboratory.

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TRANSFER OF RADIOACTIVITY IN THE TREATMENT OF WASTEWATER BY PHOSPHATE

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Abstract

Radionuclides of natural, terrestrial origin are ubiquitous in the environment and contribute significantly to external and internal doses to the population. Of particular importance are the solid radionuclides belonging to the ^{238}U and ^{232}Th series which are present in soil and rocks. The aim of this work was to study the transfer of ^{238}U and ^{232}Th during the treatment of wastewater by infiltration percolation through a phosphate bed. To assess radiation dose due to the treated water which is used in agriculture and for the irrigation of gardens in urban areas, these radionuclides have been measured in wastewater, phosphate samples and treated water by using solid state nuclear track detectors.

1. INTRODUCTION

In Morocco, the mining areas are located in places with low water potential. The mobilization of non-conventional resources, namely the recycling of wastewater from the washing of phosphates, could meet the water demand. The need for water is growing and the recycling of wastewater is in line with an environmental culture which forms part of a vision for sustainable development.

The contribution to the recycling of urban wastewater was conducted using the process of infiltration percolation based on a well defined size of a phosphate bracket. The study has shown that the new bracket used has interesting performance in terms of removing the suspended particles and organic pollution by infiltration, percolation, adsorption, chemical reaction, biological pathway, predation and absorption mechanisms of plants or a purification process [1–3]. Treatment systems in the soil are able to achieve elimination levels comparable with those achieved with the best technologies for conventional wastewater treatment, with additional advantages such as the recovery and use of wastewater and nutrients for crop production [4, 5]. Studies in the field and in the laboratory led to the achievement of excellent processing performance of domestic wastewater by infiltration [6], with removal rates of COD, suspended solids and viruses of more than 80% and of phosphorus removal of almost 100% [7–11]. The radioactivity of phosphates has always been a concern because of uranium levels of 100–200 ppm, depending on the set [12]. A study of the distribution of radioactivity in the various components of the filtration bed, and especially that of uranium and thorium, is necessary to check whether the radioactivity exceeds international guidelines for treated water.

2. MATERIALS AND METHODS

2.1. Phosphate

The material used in this study was phosphate from Khouribga mine. It was washed with distilled water and dried in an oven at 105°C. A sample was characterized by X ray diffraction using a Siemens D500 powder diffractometer. A scanning electron microscope (SEM) (Zeiss DSM950) was used to determine the morphology of the material. The specific surface area was determined by the BET method using a Micromeritics ASAP2420. The results are summarized in Table 1. Analysis by DRX indicated that the natural phosphates include more than 200 mineral species, but the most common belong to the family of apatite. The mineralogy of the phosphate minerals of sedimentary deposits is close to that of fluorapatite [13].

2.2. Wastewater

The pretreated wastewater undergoes primary treatment for 1.5–2 days, which can significantly reduce the pollutant load. As the network of the city is unitary, the analysis performed on the sample provided the following results for the pollution parameters studied.

TABLE 1. CHARACTERIZATION OF THE PHOSPHATE

Type of analysis	Characteristic
SEM	Irregular or rounded grains
Surface area	14.8 m ² /g

Note: SEM — scanning electron microscope.

2.3. Pilot study of water treatment

Monitoring of the treatment by infiltration percolation was performed using a glass column 50 cm high and 10 cm in diameter. The filter material had a height of 15 cm and was placed on a filter cloth to prevent loss of fines. This was placed on 3 cm of gravel and everything was supported by a grid, allowing the free flow of the treated water.

The layer of filter material was covered by a 3 cm layer of gravel and webbing, to ensure the dispersion of the water supply to the entire bed and to avoid the formation of a preferential path [14].

2.4. Radioactivity

Analysis of the radioactivity was performed using a method described in detail in Ref. [15], based on the calculation of the detection efficiency of alpha particles emitted by uranium and thorium in the samples studied. CR-39 and LR-115 type II solid state nuclear track detectors (SSNTDs) were used. The technique involved placing the detectors in direct contact with the sample. It was based on the knowledge of the stopping power of alpha particles in the sample studied and the detection efficiency of the detector [16]. To measure the concentrations of ²³⁸U and ²³²Th in the phosphate samples, films were sealed in polyethylene bottles and then irradiated. After a month of radiation, chemical development was applied to each detector. The reading was performed using an automatic system consisting of a charge-coupled device camera connected to a microscope, with a table (X, Y and Z) controlled by software.

3. RESULTS AND DISCUSSION

The determination by alpha spectrometry of the radiation emitted by uranium and thorium in the system (gravel–phosphate–water) using SSNTDs

TABLE 2. RESULTS OF THE INFILTRATION PILOT STUDY

	$\rho_G^{LR} \times 10^5$ (tracks·cm ⁻² ·s ⁻¹)	$\rho_G^{CR} \times 10^5$ (tracks·cm ⁻² ·s ⁻¹)	U-238 concentration (ppm)	Th-232 concentration (ppm)
Before infiltration				
Untreated wastewater	2.96 ± 0.24	9.11 ± 0.73	0.78 ± 0.06	0.93 ± 0.07
Unused gravel	19.15 ± 1.53	61 ± 5	6.7 ± 0.54	17.90 ± 1.43
Unused phosphate	465 ± 37	1360 ± 108	157 ± 13	11.00 ± 0.97
After infiltration				
Treated wastewater	3.11 ± 0.28	9.55 ± 0.86	0.82 ± 0.07	0.95 ± 0.09
Used gravel	17.36 ± 1.56	55 ± 5	6.3 ± 0.57	17.40 ± 1.57
Used phosphate	449 ± 40	1313 ± 118	152 ± 11	10.50 ± 0.95

gave the concentration of radionuclides in the ^{238}U and ^{232}Th series. The results are shown in Table 2.

The concentration of uranium and thorium in the water increased after passing through the filter bed formed by the phosphate. This can be explained by the presence of calcite and apatite in these samples. In parallel with this, a decrease was noted in the concentrations of uranium and thorium in the phosphate and gravel used in the treatment, indicating a transfer of radioactivity from the phosphate to the water.

The World Health Organization guidelines for drinking water are 0.015 mg/L for uranium while for thorium, which is relatively insoluble in water, the concentration rarely exceeds 1 $\mu\text{g/L}$ (4 mBq/L). The radioactivity levels in the water treated by infiltration percolation on a bed of phosphate are tolerable compared with the recommendations of the International Commission on Radiological Protection [17–19].

4. CONCLUSION

This study shows that, during the treatment of wastewater using infiltration percolation on a bed of phosphate, radioactivity is transferred from the phosphate rock to the treated water. However, analyses of the concentrations of uranium and thorium in the treated water (0.82 ± 0.07 and 0.95 ± 0.09 ppm, respectively) and in the phosphate (152 ± 11 and 10.50 ± 0.95 ppm, respectively) show that the wastewater treated by the phosphate is not significantly contaminated by uranium and thorium. Therefore, it does not represent any hazard to the environment.

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^{222}Rn , ^{220}Rn CONCENTRATIONS AND MINER DOSES IN NON-URANIUM MINES IN CHINA

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Abstract

A preliminary survey of ^{222}Rn and ^{220}Rn concentrations in non-uranium mines in China was conducted in 17 different types of mines. In total, 44 mines in 12 provinces were surveyed. The results showed that among 25 metal mines ($n = 147$) the arithmetic mean and geometric mean concentrations were 1214 ± 2358 and 313 ± 5.5 Bq/m³ for ^{222}Rn , respectively (range 11–19 600 Bq/m³) and 268 ± 701 and 70 ± 4.4 Bq/m³ for ^{220}Rn , respectively (range <LLD–6554 Bq/m³). Among 18 non-metal mines ($n = 119$), the arithmetic mean and geometric mean concentrations were 101 ± 207 and 67 ± 3.4 Bq/m³ for ^{222}Rn , respectively (range 5–1784 Bq/m³) and 101 ± 207 and 67 ± 3.4 Bq/m³ for ^{220}Rn , respectively (range <LLD–436 Bq/m³). The ^{222}Rn equilibrium factor was 0.33 ± 0.15 in underground mines and 0.47 ± 0.18 in above ground buildings. The average ^{222}Rn concentrations in 15% of the sampled mines exceeded the workplace recommended limit of 1000 Bq/m³. The ^{222}Rn concentration at some individual points exceeded 10 000 Bq/m³. The arithmetic mean ^{222}Rn concentration in 14 coal mines in 6 provinces was 117 Bq/m³. Compared with the survey data from previous years, the ^{222}Rn concentrations in coal mines had reduced significantly. However, high ^{222}Rn exposure remains a problem in metal mines such as those for copper, tin, lead, zinc, gold and aluminum. The miners' radon exposures were estimated from the above results. The average annual effective dose for miners in metal mines was 7.75 mSv, with doses exceeding 20 mSv in four such mines. The average dose in a rare earth mine was 1.41 mSv, of which 53% was contributed by ^{220}Rn . The average dose in coal mines was 0.75 mSv, and in non-metal mines it was 0.38 mSv.

1. INTRODUCTION

It has been confirmed that lung cancer in uranium miners caused by inhalation of radon and its progeny is occupational cancer [1]. In 1988, radon was verified as a class A carcinogen by the International Cancer Research Centre, based on its carcinogenic effects [2]. High levels of radon were found in Yunnan tin mine which was associated with a significant increase of lung cancer incidence in the 1970s [3–5]. Later, some of the typical mines were measured

and it was found that radon levels were higher in non-ferrous metal mines and rare earth mines [6]. Due to the particular working environment in underground mines, radon and its progeny are the main source of miners' radiation exposure.

In recent years, with the rapid development of the mining industry, the number of mine workers has greatly increased in China. However, basic information on radiation levels in mines and on exposure levels of miners is insufficient. In this study, a preliminary investigation of radon levels was conducted in different types of non-uranium mines during the period 2006–2008, in order to estimate the doses received by miners.

2. BACKGROUND AND METHODS

2.1. The background to China's non-uranium mines

In China, 171 kinds of mineral have been discovered [7] and can be divided into four categories: metal ores, non-metal ores, energy related minerals and water and gas. Mines other than uranium mines are referred to as non-uranium mines, relating to 170 kinds of mineral. In 2006, 5.833 billion t of ore were mined in China. The amounts of coal and iron ore accounted for a large proportion of this — 1.962 billion and 424 million t, respectively, accounting for 33.6% and 7.3% of the total ore mined. This represented an increase of 11.14% over the figure for 2005. The total of major minerals and ores mined in China ranked third in the world.

According to statistics from the Ministry of Land and Resources [8], there were 126 370 mining related companies, excluding those involved in uranium and offshore oil and gas production, employing 7.98 million people by the end of 2006. Table 1 summarizes the quantities and ratios of workers in various types of facility.

2.2. Selection of facilities to be surveyed

The following factors were mainly considered. First, non-ferrous metals and underground mines were preferred. Second, in addition to mine type, size and the number of employees, the geographical distribution and radiation related factors were also considered. Third, due to the large amount of coal mines and iron mines (accounting for 33.6% and 7.3% of the total mining, respectively) the sample sizes of these two types of mines were appropriately increased. Fourth, some of the previously surveyed mines were chosen in order to study the change of radon concentration. In this survey, 44 facilities of various types were included:

TABLE 1. NUMBERS OF WORKERS, BY TYPE OF FACILITY

	Mineral	No. of workers	Proportion of total (%)
Metal mines	Ferrous metal ore	413 700	
	Non-ferrous metal ore	427 900	
	Precious metal ore	201 200	
	Rare earths ore	12 900	
	Metallurgical auxiliary minerals	97 800	
	Subtotal	1 153 500	14.4
Non-metal mines	Building materials and other non-metallic minerals	2 200 500	
	Raw chemical ore (phosphate, salt)	69 400	
	Subtotal	2 269 900	28.4
Energy mines	Non-uranium energy mineral	4 426 600	55.5
Water and gas extraction	Mineral water and groundwater	33 100	0.4
Other		99 900	1.3
Total		7 983 000	100

- (a) Twenty-five metal mines, involving 12 minerals, accounting for 56.8% of the total number of surveyed mines;
- (b) Eighteen non-metallic mines (including coal), involving four minerals, which accounted for 40.9% of surveyed mines;
- (c) One geothermal hot spring facility for tourism.

Of the facilities surveyed, 40 (90.9% of the total number surveyed) were underground mines and 4 were surface facilities, including 2 open pit mines, 1 ore processing plant and 1 hot spring. In addition, the sampling rate of the coal and iron mines was higher — 14 coal mines (31.8% of the facilities surveyed) and 8 iron mines (18.2% of the facilities surveyed) were included in the survey. The survey involved 12 provinces and more than 70 000 employees. The information is summarized in Table 2. The number of workers and the number of mines were estimated from data obtained from the Ministry of Land and Resources [8]. The geographic distribution of the facilities included in the survey is shown in Fig. 1.

TABLE 2. WORKERS AND FACILITIES COVERED BY THE SURVEY

	No. of workers			No. of facilities		
	Estimated total	Included in survey	Coverage (%)	Total	Included in survey	Coverage (%)
Metal mines	1 153 500	26 974	2.34		25	
Non-metallic mines	2 269 900	237	0.11		4	
Energy mines (coal mines)	4 426 600	41 237	0.93	35 751	14	0.039
Water or gas extraction	33 100	20	0.06		1	
Other	99 900					
Total	7 983 000	70 748	0.89	126 370	44	0.035

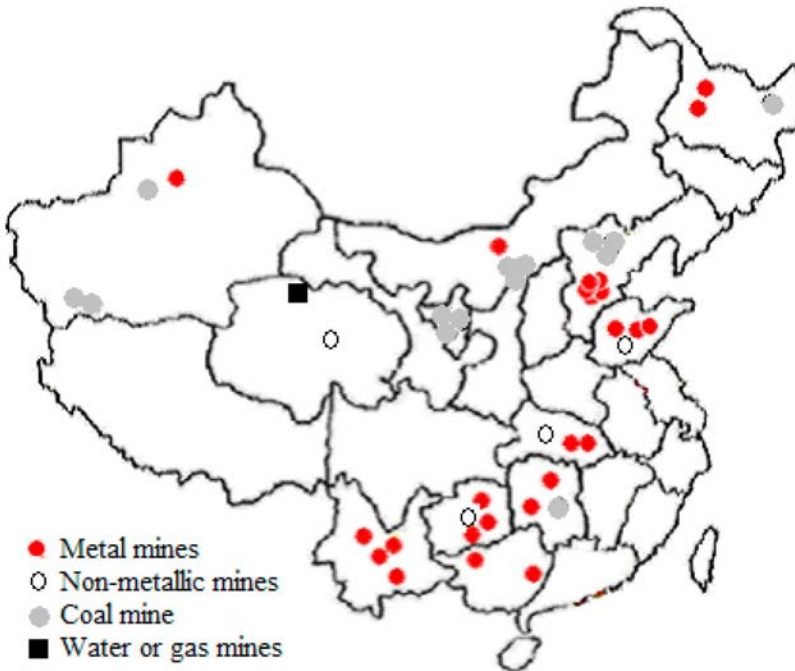


FIG. 1. Geographic distribution of the facilities included in the survey.

2.3. Selection of the measurement locations

The selection of the radon measurement locations focused mainly on underground work areas. Other work areas such as office areas and surface buildings (living areas or outdoor areas) were selected as control points. About 10–15 locations were selected for each facility, including 6–10 underground locations, 2–4 surface locations and 1–2 outdoor locations. More measurement locations were added for large mines or mines with multilevel extraction, depending on the specific situation.

Representative sites where workers remained for longer periods were selected for underground mines, such as the main roadways, stopes, power distribution rooms, tramcar hatchways, air inlets and air outlets. The location and height of the detection devices were determined according to the specific circumstances — a height of 1–1.8 m was representative of the breathing zone and was less susceptible to interference or loss during the measurement period. For surface locations, the work areas where workers spent most time, such as workshops, offices, duty rooms, as well as living areas such as dormitories or residences, were selected as measurement locations. Outdoor workplaces or living areas where people often remained were selected and the measurement height was increased as appropriate to prevent loss of detection devices.

Cumulative radon measurements in 44 mines of 17 different kinds from 12 provinces were completed by August 2008. Of 406 pairs of LD-P detectors deployed in 35 mines, 325 pairs gave effective results. The detector recovery efficiency was 80.1%. In 9 mines, 54 electret passive environmental radon monitors (E-PERMs) were deployed and the detector recovery efficiency was 100%.

2.4. Measurement devices and quality assurance

A ^{222}Rn and ^{220}Rn discriminative detector (R–T detector) [9] was used to measure cumulative ^{222}Rn and ^{220}Rn concentrations in mines. The detector was calibrated in a radon chamber at Nanhua University (NURL). The calibration factors were 3.15 ± 0.42 and 2.10 ± 0.18 tracks/cm² per kBq·h·m⁻³ for ^{222}Rn and ^{220}Rn , respectively. The lower limits on detection (LLD) were 6 and 35 Bq/m³ per 90 days, respectively. The saturation concentrations were 8000 and 15 000 Bq/m³ per 90 days, respectively. The detector was checked in an intercomparison with a Raduet detector of JCAC, Japan, and met the control limits [10]. The technical indicators of the detector meet the environmental radon measurement needs of mines. E-PERMs [11] were used for short term measurements (2–7 days). Calibration was done in a radon chamber at NURL; the calibration factors (CFs) and the relative percentage differences (RPDs) are shown in Table 3.

TABLE 3. E-PERM DETECTOR CALIBRATION RESULTS

Reference radon concentration (Bq/m ³)	No. of measurements	ΔV	Exposure time (h)	CF ^a	Measured radon concentration (Bq/m ³)	Standard deviation (Bq/m ³)	RPD ^b (%)
1028	5	15.2	6	2.18 ± 0.06	1000	23.3	1.6
640	5	15.2	10	2.16 ± 0.05	628	17.9	-3.4

^a CF — calibration factor.

^b RPD — relative percentage difference.

Continuous measuring devices, such as RTM2100, RAD7 and LCD-BWLM, were used for the rapid measurement of ^{222}Rn and ^{220}Rn concentrations, the potential alpha energy concentrations of ^{222}Rn and ^{220}Rn progeny and the equilibrium factor. All instruments were calibrated in the radon chamber at NURL. The calibration factors are shown in Table 4.

TABLE 4. CALIBRATION FACTORS OF CONTINUOUS MEASURING DEVICES

Type of device	Model	Manufacturer	F_{adjust}
^{222}Rn , ^{220}Rn monitor	RTM2100	SARAD, Germany	1.32 ± 0.06
Radon detector	RAD7	DurrIDGE, USA	1.01 ± 0.07
BWL monitor	LCD-BWLM	Tracerlab, Germany	1.20 ± 0.03

3. RESULTS

3.1. ^{222}Rn and ^{220}Rn levels in mines

R-T detectors and E-PERM detectors were used for cumulative radon measurements in underground workplaces (declines and roadways), surface workplaces (workshops and offices, among others) and living areas on mines. The exposure time of the R-T detector was 3 months or, for some mines, 6–12 months. For high radon areas, the exposure time was less than 90 days to avoid track saturation; alternatively, E-PERM detectors were used. The underground locations for which the exposure time was more than 90 days accounted for 84.5% of the total, while the corresponding figure for surface locations was 91.4%.

3.1.1. Metal mines

In the survey, 25 metal mines were selected, one of which was an open pit mine and the rest were underground mines. The concentrations of ^{222}Rn and ^{220}Rn in metal mines are shown in Table 5. For ^{222}Rn , the concentrations were in the range 11–19 600 Bq/m³, with an arithmetic mean and geometric mean of 1214 ± 2358 and 313 ± 5.5 Bq/m³, respectively. For ^{220}Rn , the concentrations ranged from below the LLD to 6554 Bq/m³, with an arithmetic mean and geometric mean

TABLE 5. ^{222}Rn AND ^{220}Rn CONCENTRATIONS IN METAL MINES

Mineral	Area	No. of samples	^{222}Rn concentration (Bq/m^3)			^{220}Rn concentration (Bq/m^3)		
			Range	Mean, standard deviation	Range	Mean, standard deviation	Range	Mean, standard deviation
Sn	Yunnan Guangxi	10	2 478–19 600	6 926 \pm 4 975	LLD–574	180 \pm 258		
		8	160–198	168 \pm 13	LLD–541	101 \pm 179		
Cu	Yunnan A Yunnan B Hubei	9	3 830–5 966	4 687 \pm 693	LLD–2 331	746 \pm 784		
		3	3 236–5 530	4077 \pm 1 264	—	—		
		6	146–1 374	765 \pm 515	LLD–1 355	528 \pm 497		
Pb, Zn	Yunnan Guangxi	3	238–3 236	2 128 \pm 1 645	—	—		
		11	72–936	683 \pm 305	LLD–1 049	207 \pm 319		
Au	Shandong A Shandong B Heilongjiang	7	1 005–2 771	1 660 \pm 651	LLD–472	149 \pm 157		
		6	321–767	454 \pm 162	—	—		
		3	109–426	298 \pm 181	<LLD	—		
W	Hunan	3	383–524	473 \pm 78	LLD–524	61 \pm 71		
Hg	Hunan	4	52–112	79 \pm 29	LLD–73	42 \pm 26		

TABLE 5. ^{222}Rn AND ^{220}Rn CONCENTRATIONS IN METAL MINES (cont.)

Mineral	Area	No. of samples	Rn-222 concentration (Bq/m ³)		Rn-220 concentration (Bq/m ³)	
			Range	Mean, standard deviation	Range	Mean, standard deviation
Fe	Shandong	5	83–510	275 ± 158	107–497	282 ± 147
	Heilongjiang	5	24–157	104 ± 49	33–797	232 ± 323
	Hubei	6	96–149	131 ± 49	LLD–219	66 ± 77
	Hebei A	6	16–385	111 ± 143	—	—
	Hebei B	5	52–138	103 ± 36	—	—
	Hebei C	4	20–56	33 ± 17	—	—
	Hebei D	4	11–198	79 ± 87	—	—
Hebei E	3	25–150	83.5 ± 63	—	—	
Al	Guizhou	8	209–1 958	1 216 ± 706	48–3 511	1 138 ± 1 208
Ni, Cu	Xinjiang	10	17–315	90 ± 82	LLD–121	47 ± 37
Mn	Guizhou	3	33–99	59 ± 35	182–567	358 ± 194
Sb	Guizhou	6	318–357	328 ± 15	<LLD	—
Rare earths	Inner Mongolia (open pit)	9	20–181	86 ± 69	69–6 554	962 ± 2 117
Overall		147	11–19 600	AM 1 214 ± 2 358 GM 313 ± 5.5	LLD–6 554	AM 268 ± 701 GM 70 ± 4.4

Note: AM — arithmetic mean; GM — geometric mean; LLD — lower limit on detection.

of 268 ± 701 and 70 ± 4.4 Bq/m³, respectively. The mean radon concentration in 6 of the 24 underground mines exceeded 1000 Bq/m³ (the workplace radon concentration limit (GB18871-2002)). One radon concentration measurement exceeded 10 000 Bq/m³.

3.1.2. *Non-metal mines*

Eighteen non-metal mines were surveyed, including 14 coal mines, 2 phosphate mines, 1 halogen mine and 1 clay mine. The results are shown in Table 6. The phosphate facility in Guizhou was a processing plant and the halogen mine in Sichuan was an open pit mine. The rest of the facilities were underground mines. The ²²²Rn concentrations in the non-metal mines were in the range 5–1784 Bq/m³ and the arithmetic mean and geometric mean were 98 ± 206 and 55 ± 2.5 Bq/m³, respectively. The ²²⁰Rn concentrations ranged from below the LLD to 463 Bq/m³ and the arithmetic mean and geometric mean were 60 ± 76 and 38 ± 2.4 Bq/m³, respectively. In previous literature, it was reported that the phosphate product had a high ²²⁶Ra concentration, but in this survey the ²²²Rn concentrations in the two large phosphate facilities were not highly elevated, the averages being 80 and 137 Bq/m³, respectively, and all measured concentrations were below 200 Bq/m³.

The mean ²²²Rn concentrations in the 14 coal mines were in the range 8–434 Bq/m³. The mean underground radon concentration in Ningxia Rukei coal mine was only 8 Bq/m³, similar to outdoor levels. The results of this study were lower than those reported in the past, owing to the improvement of ventilation in recent years.

A halogen salt mine was also surveyed. The barium sulphate (BaSO₄) by-product contained elevated levels of ²²⁶Ra. Brine was extracted from underground and piped to a sedimentation and purification plant. The measurements focused on BaSO₄ production and the storage workshop. The mean radon concentration which was based on ten measurement locations was only 18.0 ± 3.1 Bq/m³ and a radon pollution problem was not found.

3.1.3. *Results for ²²⁰Rn*

The measurement of ²²⁰Rn concentration was carried out in the 44 mines for comparison purposes. Detectors were deployed on or close to roadway walls. High ²²⁰Rn concentrations were detected in some mines, including the rare earths mine in Inner Mongolia and the Guizhou aluminum mine, both of which had mean values of about 1000 Bq/m³. The highest measured concentration was more

TABLE 6. ^{222}Rn AND ^{220}Rn CONCENTRATIONS IN NON-METAL MINES

Mineral	Area	No. of samples	Rn-222 concentration (Bq/m ³)			Rn-220 concentration (Bq/m ³)		
			Range	Mean, standard deviation	Range	Mean, standard deviation	Range	Mean, standard deviation
Phosphate	Guizhou (surface processing plant)	3	105–169	137 ± 32	—	—	—	—
	Hubei	13	60–159	80 ± 34	LLD–165	41 ± 41	41 ± 41	41 ± 41
Clay	Shandong	7	5–73	4 ± 22	LLD–98	42 ± 34	42 ± 34	42 ± 34
Halide salt	Sichuan	10	15–25	18.0 ± 3.1	LLD–98	43 ± 30	43 ± 30	43 ± 30
Coal	Ningxia A	7	33–189	108 ± 58	LLD–436	219 ± 183	219 ± 183	219 ± 183
	Ningxia B	5	5–12	8 ± 3	LLD–259	97 ± 94	97 ± 94	97 ± 94
	Ningxia C	6	20–195	84 ± 73	LLD–183	118 ± 56	118 ± 56	118 ± 56
	Inner Mongolia A	3	12–36	25 ± 12	LLD–91	44 ± 41	44 ± 41	44 ± 41
	Inner Mongolia B	4	19–34	27 ± 6	<LLD	—	—	—
	Inner Mongolia C	3	24–50	35 ± 14	LLD–62	34 ± 25	34 ± 25	34 ± 25
	Heilongjiang	4	10–103	35 ± 45	LLD–112	66 ± 53	66 ± 53	66 ± 53
	Xinjiang A	26	18–151	61 ± 31	LLD–355	56 ± 78	56 ± 78	56 ± 78
	Xinjiang B	5	65–99	80 ± 14	LLD–166	61 ± 64	61 ± 64	61 ± 64
	Xinjiang C	5	64–110	85 ± 25	LLD–97	36 ± 31	36 ± 31	36 ± 31
	Hebei A	4	26–80	59 ± 17	LLD–124	59 ± 57	59 ± 57	59 ± 57
	Hebei B	3	21–45	37 ± 11	LLD–47	29 ± 12	29 ± 12	29 ± 12
	Hebei C	2	29–43	36 ± 10	LLD–21	21 ± 1	21 ± 1	21 ± 1
Human		8	37–1784	434 ± 685	LLD–254	69 ± 93	69 ± 93	69 ± 93

TABLE 6. ^{222}Rn AND ^{220}Rn CONCENTRATIONS IN NON-METAL MINES (cont.)

Mineral	Area	No. of samples	Rn-222 concentration (Bq/m ³)		Rn-220 concentration (Bq/m ³)	
			Range	Mean, standard deviation	Range	Mean, standard deviation
Overall		118	5-1784	AM 98 ± 206 GM 55 ± 2.5	LLD-436	AM 60 ± 76 GM 38 ± 2.4

Note: AM — arithmetic mean; GM — geometric mean; LLD — lower limit on detection.

than 6000 Bq/m^3 in a rare earths workshop and was associated with the processing of ore samples with high ^{232}Th levels [12]. The ^{220}Rn problem is a new discovery in recent years. Information on ^{220}Rn levels in mines is very limited, therefore it is important to conduct further studies in mines with high ^{220}Rn levels.

3.2. High radon levels in underground mines

Table 7 shows the high radon levels measured in certain mines, which exceeded the control limit. Forty underground mines ($n = 243$) were measured and six of them had an average ^{222}Rn concentration exceeding 1000 Bq/m^3 . All six mines were metal mines, accounting for 15.0% of the measured mines. The ^{222}Rn concentrations exceeded 1000 Bq/m^3 at 40 locations, accounting for 16.5% of the total sampling locations in underground mines. At 24 locations, the concentration exceeded 3700 Bq/m^3 (the radon concentration limit for uranium mines EJ/T378-89), accounting for 9.9% of the total sampling locations in underground mines. There are more than 1 million workers in metal mines, and this survey involved about 27 000 mine workers, with a coverage rate of 2.3%. The radon problem in metal mines is worthy of attention.

3.3. Equilibrium factor

The equilibrium factors of ^{222}Rn , ^{220}Rn and their decay products in underground mine workplaces and surface living areas were determined using a continuous measuring device. Table 8 shows the results of those determinations. The equilibrium factors for indoor ^{222}Rn in surface workplaces and living areas were in the range 0.35–0.54 with a mean of 0.47 ± 0.18 ($n = 195$). This is close to the typical indoor value of 0.49 [13]. The equilibrium factors in underground mines were in the range 0.10–0.55 with a mean of 0.33 ± 0.15 ($n = 39$), slightly lower than the default value of 0.4 recommended for mines in ICRP Publication 65 [14]. The values determined in this study were also very close to the values measured previously in coal mines in China (mean: 0.34 ± 0.5 , range: 0.14–0.74) [15].

Table 9 shows the equilibrium factors determined for ^{220}Rn . The values determined in three mines were in the range 0.002–0.032, varying by an order of magnitude. The distribution of ^{220}Rn concentrations in underground mines was very uneven. This was a preliminary result. There are no other relevant reports and therefore no comparison can be made.

TABLE 7. LOCATIONS WITH HIGH RADON CONCENTRATIONS

Mineral	Area	Season	Measurement period (d)	Rn-222 concentration (Bq/m ³)			Proportion of measurements (%)	
				No. of samples	Range	Mean, standard deviation	>1000 Bq/m ³	>3700 Bq/m ³
Sn	Yunnan	Spring, summer	40	10	2478–19 600	6926 ± 4975	10	7
Cu	Yunnan A	Summer	1	3	3236–5530	4077 ± 1264	3	1
	Yunnan B	Whole year	351	9	3830–5966	4687 ± 693	9	8
	Hubei	Spring, summer	87	7	146–1374	765 ± 515	2	
Pb–Zn	Yunnan	Summer	1	3	238–3236	2128 ± 1645	2	
Au	Shandong A	Summer, autumn	135	7	1005–2771	1660 ± 651	7	
Al	Guizhou	Whole year	347	8	209–1958	1216 ± 706	5	
Coal	Hunan	Summer, autumn	93	8	37–1784	434 ± 685	2	
Total							40	16

TABLE 8. EQUILIBRIUM FACTOR FOR ^{222}Rn

Measurement location	Rn-222 concentration (Bq/m ³)	Equilibrium equivalent concentration (Bq/m ³)	Equilibrium factor
Surface workplaces, indoors			
Office, rare earth mine	142	75.1	0.53 ± 0.20
Office, iron mine	26	14.1	0.54 ± 0.17
Workshop, iron ore crushing	32	13.4	0.42 ± 0.15
Workshop, iron ore processing	150	62.2	0.41 ± 0.14
Workshop, halogen mine	17.2	7.9	0.50 ± 0.18
Workshop, iron ore crushing	46	23.3	0.51 ± 0.19
Hostel, tungsten mine	367	196	0.50 ± 0.16
Dormitory, copper mine	16.9	5.6	0.35 ± 0.16
Mean			0.47 ± 0.18
Underground workplaces			
Iron mine, Shandong	2121	486	0.23 ± 0.06
Iron mine, Hebei	36.5	5.7	0.15 ± 0.01
Copper mine, Hebei	57	5.9	0.10 ± 0.03
Copper mine, Yunnan	4980	1972	0.40 ± 0.12
Tin mine, Yunnan	7138	1848	0.26 ± 0.01
Tin mine, Guangxi	196	53.5	0.28 ± 0.05
Lead-tin mine, Yunnan	3073	1566	0.51 ± 0.04
Lead-tin mine, Guangxi	1393	571	0.41 ± 0.06
Gold mine, Shandong	669	368	0.55 ± 0.07
Mean			0.33 ± 0.15

TABLE 9. EQUILIBRIUM FACTOR FOR ^{220}Rn

Measurement location	No. of samples	Rn-220 concentration (Bq/m ³)	Equilibrium equivalent concentration (Bq/m ³)	Equilibrium factor
Underground				
Copper mine	4	254	0.7	0.003 ± 0.001
Iron mine	5	188	6.1	0.032 ± 0.022
Coal mine	4	4270	8.9	0.002 ± 0.002
Workshop				
Rare earth mine	5	271	1.9	0.007 ± 0.03

3.4. Dose estimation

Mineworkers in China work for 8 h/d which, assuming 250 working days per year, gives an annual working period of 2000 h. The mineworkers' annual effective dose due to the inhalation of ^{220}Rn , ^{222}Rn and its decay products during the working period can be estimated according to the dose conversion factor given by the United Nations Scientific Committee on the Effects of Atomic Radiation [16] and the measured equilibrium factors.

As shown in Table 10, mine workers in metal ore mines received the highest average dose, at 7.75 mSv/a, followed by rare earth miners, who received an average dose of 1.41 mSv/a, of which 53% was due to ^{220}Rn . The average dose received by coal miners was 0.76 mSv/a, which was lower than the level reported in recent literature [17]. This may be due to the fact that those coal mines were mainly state owned enterprises; they followed good work practices and had good underground ventilation. The lowest average dose was found in a non-metal mine (0.38 mSv/a). An average dose of 0.72 mSv/a was found in a phosphate mine. The doses in metal and phosphate mines were similar to those found in Polish mines (9.6 mSv/a for metal ore mining and 0.18 mSv/a for phosphate mining [18]).

TABLE 10. DOSE FROM INHALATION OF ^{220}Rn AND ^{222}Rn

	Rn isotope	Equilibrium factor	Av. concentration (Bq/m ³)			Annual av. effective dose (mSv)		
			Rn	Equilibrium equivalent	Rn	Rn	Progeny	Total
Metal mine (n = 144)	Rn-222	0.33	1214	401	0.41	7.21	7.75	
	Rn-220	0.002	337	0.67	0.07	0.05		
Non-metal mine (n = 119)	Rn-222	0.33	58	19	0.02	0.34	0.38	
	Rn-220	0.002	39	0.08	0.01	0.01		
Coal mine (n = 86)	Rn-222	0.34	113	38.4	0.04	0.69	0.76	
	Rn-220	0.002	68	0.14	0.01	0.01		
Phosphate mine (n = 16)	Rn-222	0.40	91	36.4	0.03	0.66	0.72	
	Rn-220	0.007	41	0.29	0.01	0.02		
Underground mine (n = 244)	Rn-222	0.33	773	255	0.26	4.59	5.58	
	Rn-220	0.002	1912	3.82	0.42	0.31		
Rare earth mine (n=9)	Rn-222	0.41	86	35.3	0.03	0.63	1.41	
	Rn-220	0.007	962	6.73	0.21	0.54		

4. DISCUSSION

4.1. Comparison with previous results

The results for ^{222}Rn were compared with the published literature as shown in Table 11. The measured results were basically the same as those reported previously for tin, copper, lead, zinc and gold mines, and indicated that radon is still a serious problem in China's non-ferrous metal mines. Radon concentrations in coal mines were significantly lower than in previous studies. The average for the 14 coal mines was 113 Bq/m³. In recent years, coal mines have generally improved their ventilation systems to prevent gas explosions. For example, only 55% of mines in Hunan Province had powered ventilation systems in the 1980s [19], whereas all mines in this survey have been using powered ventilation. Many mines have closed blind alleys and goafs, with the objective of decreasing underground ^{222}Rn concentrations.

The overall sampling rates in this survey were less than 1% (the number of sampled mineworkers was 0.9% of the total mine workers, and the number of mines sampled accounted for only 0.4% of all mines). The sampling rate of metal mines was 2.3%. The goal for future work is to expand the scale of measurements, especially for metal mines and thermal spas in order to obtain more comprehensive results.

4.2. Associated radioactivity problems

Thermal spas use water from underground [7]. A hot spring near a uranium mine was surveyed, and the ^{222}Rn concentration and gamma exposure rate were measured. There were three other mines with existing problems associated with radioactivity in this investigation. Table 12 shows ^{222}Rn , ^{220}Rn levels and gamma dose rates in all four facilities.

Barium sulphate, a by-product of a halogen mineral in Sichuan containing high levels of ^{226}Ra , had been found to cause radon contamination after being sold as a paint stabilizer [20]. Measurements in the BaSO₄ production workshops, storage workshop, finished product warehouse and the surrounding environment were conducted. The gamma exposure rate in the finished product warehouse was 4.07 μGy/h. The highest ^{222}Rn concentration in the BaSO₄ production and storage workshop was 24.5 Bq/m³ for a four month cumulative measurement. No increase of ^{222}Rn concentration in the workplace was found. This low exposure may be due to the open space design and good ventilation of the workshop, but it is important to track the ^{226}Ra level.

A coal seam in a coal mine in Xinjiang contained uranium ore. The mine operator paid great attention to the associated radioactivity problem, and the

TABLE 11. COMPARISON WITH PREVIOUSLY PUBLISHED LITERATURE

Mine type	Literature	Year	Measurement method	Rn-222 concentration (Bq/m ³)			Ratio of mean values
				No. of samples	Range	Arithmetic mean	
Tin	This paper	2006–2008	ATD	18	157–19 600	3 923	1.26
	Ref. [19]	1986–1994	Double membrane	5	1 576–5 180	3 120	
Copper	This paper	2006–2007	ATD	18	146–5 530	3 278	0.93
	Ref. [19]	1986–1994	Double membrane	10	145–15 625	3 537	
Tungsten	This paper	2006–2007	ATD	3	383–524	473	0.43
	Ref. [19]	1986–1994	Double membrane	11	112–3 904	1 092	
	Ref. [20]	1988	Balloon method		168–30 800	12 700	
Lead–zinc	This paper	2006–2008	ATD	14	72–3 236	993	0.97
	Ref. [19]	1986–1994	Double membrane	6	148–2 721	1 025	
	Ref. [21]	1998	Scintillation cell	9	1 097–3 933	2 376	
	Ref. [22]	1996	Double membrane	8	204–1 622	959	
Aluminum	This paper	2007–2008	ATD	8	340–1 958	1 216	0.36
	Ref. [19]	1986–1994	Double membrane	1	—	3 385	

TABLE 11. COMPARISON WITH PREVIOUSLY PUBLISHED LITERATURE (cont.)

Mine type	Literature	Year	Measurement method	Rn-222 concentration (Bq/m ³)			Ratio of mean values
				No. of samples	Range	Arithmetic mean	
Gold	This paper	2006-2008	ATD	16	71-2 771	946	0.95
	Ref. [23]	1998	Balloon method	21	51-1 134	1 000	
	Ref. [22]	1996	Double membrane	24	60-8 063	884	
Iron	This paper	2006-2008	ATD	38	11-510	120	0.03
	Ref. [24]	1997	ATD	141	109-13 340	3 778	
Coal	This paper	2004-2007	ATD	86	10-1 784	117	0.48
	Ref. [25]	2002-2004	ATD	718	40-3 187	245	
	Ref. [21]	1998	Scintillation cell	33	14-3 933	938	

TABLE 12. GAMMA DOSE RATES AND ^{222}Rn , ^{220}Rn LEVELS IN FOUR FACILITIES ASSOCIATED WITH RADIOACTIVITY

	Radionuclide	Gamma dose rate ($\mu\text{Gy/h}$)		Activity concentration (Bq/m^3)		
		No. of samples	Range	Radionuclide	No. of samples	Range
Halogen mine	Ra-226	0.036	0.097–4.07	Rn-222	11	15–25
Coal mine	U-238	0.028	0.065–0.395	Rn-222	5	120–295
Spa	U-238, Ra-226	0.059	0.205–28.718	Rn-222	5 (bath area) 5 (guest room)	580–12 460 254–860 000 ^a
Rare earths mine	Th-223	0.052	0.110–1.13	Rn-220	9	69–6 554

^a Estimated, based on saturation track.

management of the mining operation was based on the gamma dose rate at the working face. No mining was allowed when the gamma dose rate exceeded 70 $\mu\text{R/h}$. When the gamma dose rate was 50–70 $\mu\text{R/h}$, coal from the mine could only be used within the mine, but no ash could be used. When the gamma dose rate was less than 50 $\mu\text{R/h}$ there was no restriction on the sale and use of the coal. Underground mining in the same mine adopted a powered ventilation system with an air flow of 24 m^3/min . Although the gamma dose rate was high in some locations, the ^{222}Rn level was below the national control standard. The appropriate protective measures not only protected the workers' health but also prevented radioactive contamination. It would serve as a good example for other mines associated with radioactivity.

High levels of radon were measured at the Jiangzha hot springs resort in Sichuan. The highest level in the bathing area was more than 10 000 Bq/m^3 . The use of geothermal water has led to abnormal indoor radon and gamma exposure rates around hot springs, and doses received by the hot spring staff were close to 100 mSv/a [21]. There are about 3000 hot spring areas in China [22] and the amount of geothermal resource exploitation is 6.8 billion m^3/a [23]. The radon contamination associated with the development and utilization of geothermal resources needs further investigation.

The rare earth mine in Inner Mongolia has an average thorium oxide content of 0.04%. High levels of ^{220}Rn were measured in the beneficiation and alkali separation plants. There is no internationally adopted exposure limit for ^{220}Rn . The IAEA has an action level of 0.3 WL (80 Bq/m^3 equilibrium equivalent concentration) for ^{220}Rn decay products in the workplace [18]. It is important to measure ^{220}Rn decay products to determine the dose due to ^{220}Rn .

It has been reported that phosphate mines tend to be associated with radionuclides in the uranium decay series [24]. The ^{226}Ra content measured in two phosphate ores was much lower than in ore from Sichuan [25] and Zhejiang [26], and the radon level was acceptable.

4.3. Radon levels at ground level

The indoor and outdoor ^{222}Rn levels around a mine were surveyed and the results are shown in Table 13. The mean outdoor radon concentration was $30 \pm 38 \text{ Bq/m}^3$ ($n = 30$), with a range of 3–165 Bq/m^3 , which was 2.1 times the average outdoor radon concentration of China (14 Bq/m^3) [24]. The mean indoor radon concentrations for surface workplaces, office areas and living areas were $177 \pm 352 \text{ Bq/m}^3$ ($n = 25$), $197 \pm 406 \text{ Bq/m}^3$ ($n = 24$) and $59 \pm 52 \text{ Bq/m}^3$ ($n = 27$) respectively, higher than typical values of indoor radon (43.8 Bq/m^3) [27]. Five measurements of indoor radon concentration gave values of more than 1000 Bq/m^3 .

TABLE 13. RADON LEVELS AT GROUND LEVEL

Location	No. of samples	Range	Arithmetic mean, standard deviation	No. of measurements >400 Bq/m ³	No. of measurements >1000 Bq/m ³
Indoor					
Workplace	25	10–1390	177 ± 352	3	3
Office area	24	9–1394	197 ± 406	3	2
Living area	30	14–228	59 ± 52	—	—
Outdoor					
Workplace	30	3–165	30 ± 38	—	—

The environmental radon pollution in mines was mainly from two sources. One was radon diffusion from the mine to the ground level, resulting in local pollution, which was worse in summer months. The other source was building materials containing slag, such as the Yaogangxian Guest House at the tungsten mine in Hunan, which is built using slag cement. The mean radon concentrations in three guest rooms were 168–228 Bq/m³ over a 24 h measurement period. The use of slag is a significant concern in non-uranium mining.

5. CONCLUSION

Cumulative measurements of ^{222}Rn and ^{220}Rn concentrations were carried out in 25 metal mines, 18 non-metal mines and 1 hot spring in 12 provinces in China using a ^{222}Rn , ^{220}Rn discriminative detector. The results show that in 15% of the metal mines the ^{222}Rn concentration exceeded the workplace radon control limit. The miners' average annual effective dose was 7.75 mSv/a. High radon concentrations remain a serious problem for metal mines, especially non-ferrous metal mines. The issue of high ^{220}Rn levels in rare earth workplaces needs further study. The average annual effective dose for miners in non-metal mines was 0.38 mSv and was 0.76 mSv in coal mines. Coal miners' annual effective dose from inhalation of ^{222}Rn was significantly lower than in previous surveys. High ^{222}Rn concentrations were found at a hot spring near a uranium mine. Special attention needs to be paid to the radon issue when exploiting and using

geothermal resources. Elevated indoor radon levels caused by the improper use of slag as a building material is also to receive attention.

ACKNOWLEDGEMENTS

The authors acknowledge the following: the local Disease Control and Prevention Centre, Xinjiang Autonomous Region, Guangxi Zhuang Autonomous Region, Ningxia Hui Autonomous Region, Shandong Province, Guizhou, Yunnan, Sichuan, Hubei, Hunan, Heilongjiang Province, Dongchuan District of Yunnan, Xingtai City, Laiwu City, Ruoergai County and Baotou preventive health care centre; also Liu Fudong, China Institute of Atomic Energy for deploying and processing of the detectors and Linzhi Kai for helping with the gamma dose rate measurements. This survey was also supported by mines including Yunnan Dongchuan Tangdan copper mine, Hubei Tonglushan copper mine, Hunan Yaogangxian tungsten ore mine, Buya coal mines and other mines.

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Invited Paper

RESIDUAL ENVIRONMENTAL CONTAMINATION AT A FORMER FERRONIBIUM PRODUCTION SITE IN BELGIUM

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Abstract

From an aerial gamma survey, a site with residues from a former ferroniobium production facility was identified. In close collaboration with the current site operator, an inventory of the mineral residue piled on the ground was made and the material was characterized in terms of its radionuclide content ($<1\text{--}12$ and $<1\text{--}60$ Bq/g for radionuclides of the uranium and thorium decay series, respectively). The possibilities for total or partial use of this kind of heterogeneous material were evaluated and were mostly hindered by its chemical composition. Since it was documented that the residues had been used in the past for the levelling and filling of the site, a systematic gamma screening survey of the whole site was performed at ground level. Gamma dose rates of up to $6\ \mu\text{Sv/h}$ were detected at certain locations. Residues with elevated radionuclide concentrations were found to have been buried to depths of up to 1.5 m. In two measurement campaigns, the shallow and deep groundwater was screened for gross alpha and beta activity and for its chemical composition. Gross alpha and beta activity concentrations of up to 1000 and 6000 mBq/L, respectively, were found. At some locations, the pH reached a value of 12.9, as a result of past contamination by a certain calcium product. Short term and long term strategies for the cleanup process are currently under discussion.

1. INTRODUCTION

An airborne gamma survey of Belgium and Luxembourg conducted by the Geological Service of Belgium in 1994–1995 [1] revealed an anomaly (shown in red in Fig. 1) in the thorium spectrometric map of north-west Belgium that could not be explained on the basis of current industrial activities. In a report compiled in 2002 [2], it was mentioned that ferroniobium production had taken place at the location of the anomaly during 1965–1980 and that slag with ^{226}Ra and ^{232}Th activity concentrations of 4–5 and 60–70 Bq/g, respectively, was found on the site. This was confirmed by the current proprietor, who mentioned that about 6000 t of excavated material containing residues from the former production of

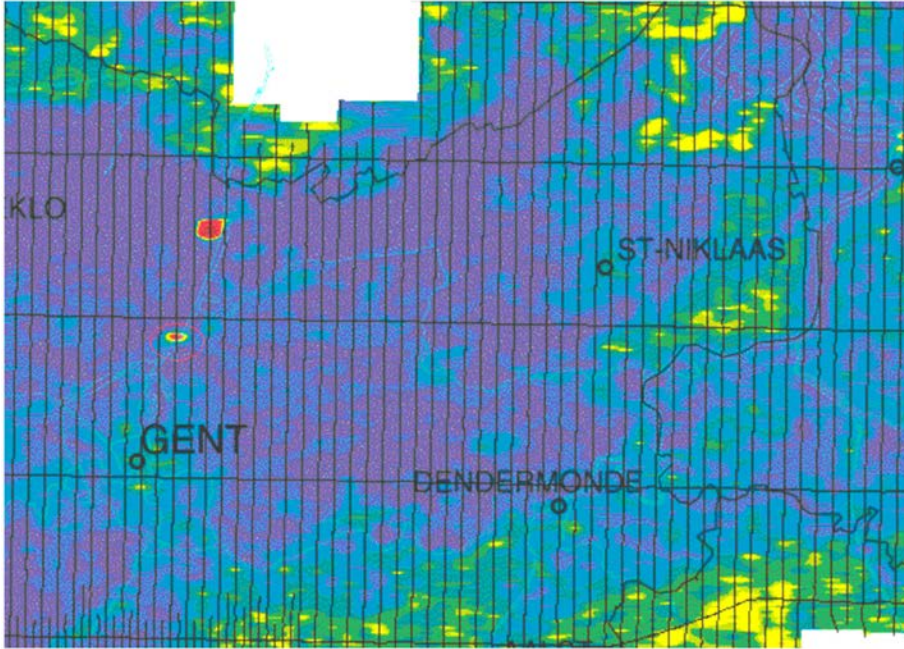


FIG. 1. Thorium anomaly in an airborne gamma survey of north-west Belgium.

ferroniobium from pyrochlore were stored on the site. The proprietor intended to characterize the material and to send it to a more appropriate storage site. Since the material was a mixture of various residues, all building debris, wood and iron were first separated. Of the remaining material, the more highly active fraction, comprising 18 t of slag residue, was transferred to 200 L drums. The rest was piled in heaps according to a rough classification based on diameter. The dose rate at 0.3 m from the drums was 1–7 $\mu\text{Sv/h}$, while the ^{226}Ra and ^{232}Th activity concentrations were 2–12 and 2–62 Bq/g, respectively, consistent with the ranges quoted in Ref. [2]. This paper describes the management of this excavated material and the detailed investigations carried out by the site operator in close collaboration with the Federal Agency for Nuclear Control.

2. DISPOSAL OF THE EXCAVATED MATERIAL

2.1. The more active fraction

At the time of the discovery of the residue, a final destination for radium and thorium containing NORM residues from former industrial activities had not been defined unequivocally.¹ It was therefore decided to evaluate the possibility of transporting the drums to a landfill site for Class 1 hazardous waste, on condition that no worker or member of the public should receive a dose exceeding 1 mSv/a. Dose calculations were made for various exposure scenarios involving workers at the former production site (storage of the residues in the drums and preparation of the material for transport), workers at the final destination (handling and storage of the drums), and members of the public (exposures of residents near both sites and exposure during transport between sites). The exposure pathways taken into account in the calculations are summarized in Table 1. The calculations indicated that, even with very conservative assumptions, the annual dose would never exceed a few tenths of a millisievert. Filling of the drums was found to be the most critical exposure scenario, for which the annual dose received by workers was calculated to be 0.18 mSv. Therefore, from a radiological point of view, the transfer of the residue to the Class 1 landfill site seemed to be fully justified. Leaching tests carried out on the residue showed that particular attention should be given to the presence of barium. Therefore, the material in the drums was subjected to physicochemical conditioning to solidify it before transport to the selected landfill site. Owing to the relatively high radium and thorium content of the residue, its transport was subject to control (ADR Class 7) and the vehicle had to be labelled appropriately.

2.2. The less active fraction

Initially, it was hoped that, for the less active fraction, some application in the building industry, such as road construction, could be found. However, the possibility of leaching of barium made such use problematic for the building industry. It was finally decided to dispose of this less active fraction at the same landfill site as that selected for the more active fraction.

¹ Since 25 March 2013, procedures for handling NORM residues have been defined in Belgian law [3].

TABLE 1. EXPOSURE PATHWAYS CONSIDERED IN THE DOSE CALCULATIONS

	Workers	Members of the public
Gamma radiation	X	
Ingestion of residue	X	
Inhalation of radon		X
Ingestion of contaminated food		X
Ingestion of contaminated drinking water		X

3. SITE INVESTIGATION

3.1. Mapping

Since there was no longer any documentation for the former processing activities and since the residues could have become spread all over the site, the first step in the site investigation was to compile a GIS map of the site using a portable sodium iodide detector coupled to precise GPS instrumentation. Areas with high count rates were identified (see Fig. 2) and 25 hotspots (with count rates exceeding five times background) were investigated and documented in detail. The composition of the slag residue found at the hotspots is summarized in Table 2.

3.2. Depth profiles and groundwater contamination

The depth of the slag residue was determined at each of the hotspots. It was found that the slag was deposited in layers, generally up to 0.75 m thick but occasionally up to 1.5 m thick. No slag was buried deeper than 1.5 m and drilling tests did not reveal any slag outside the hotspot areas.

Gross alpha activity concentrations were measured in 19 groundwater samples taken from both shallow locations (depth 1–4 m) and deep locations (depth 5–8 m). The results are shown in Table 3, indicating that the concentrations in the deep groundwater often exceeded 100 mBq/L, the screening value established in Council Directive 98/83/EC of 3 November 1998 on the quality of water intended for human consumption [4].

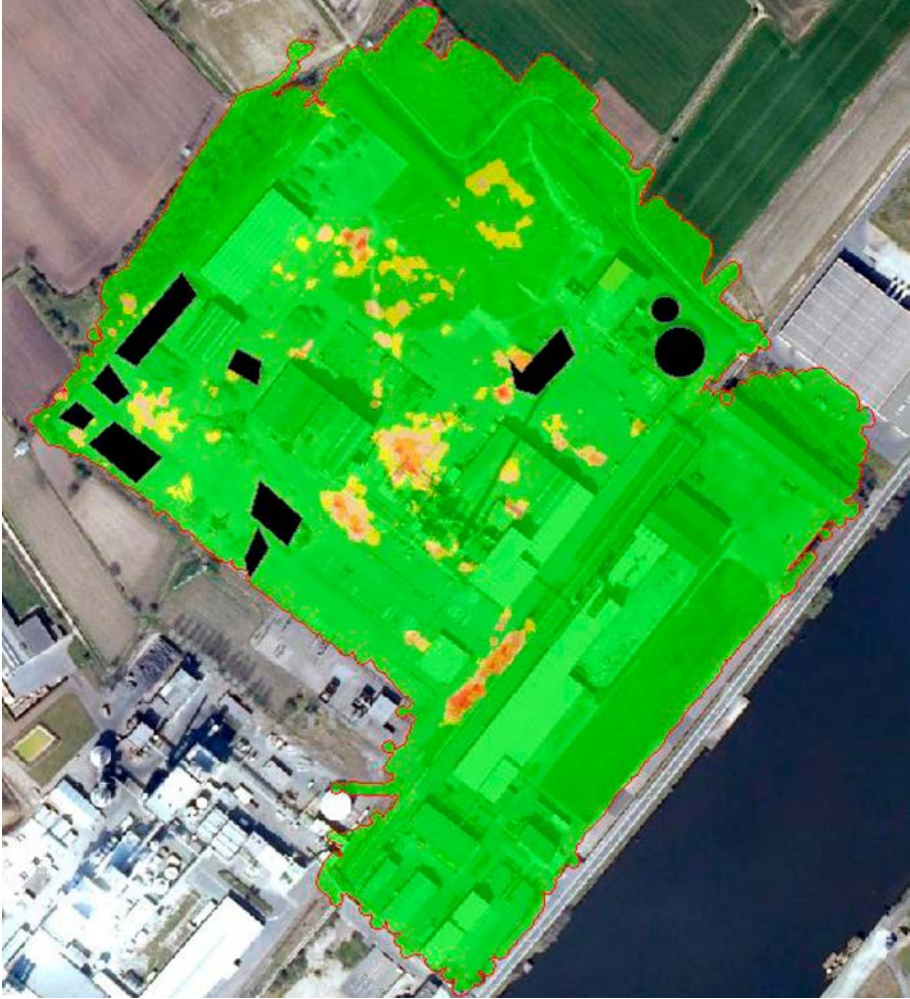


FIG. 2. GIS map of the site, showing the hotspot areas.

In one section of the site, most of the gross alpha activity concentrations exceeded 100 mBq/L. Follow-up groundwater measurements were made in this section. The results are shown in Tables 4 and 5. For water samples in which the gross alpha activity concentration exceeded 100 mBq/L, the radionuclide composition was determined by spectrometric analysis. However, such analyses did not provide an explanation for the high gross alpha activity concentrations — the activity concentrations of individual radionuclides did not correlate with the corresponding gross alpha activity concentration.

TABLE 2. COMPOSITION OF THE SLAG RESIDUE AT THE HOTSPOTS

	Activity concentration (Bq/g)		
	Min.	Av.	Max.
U-238	2.4	7.3	12.0
Th-230	2.5	8.6	12.0
Ra-226	2.3	7.5	12.5
Pb-210	1.6	4.6	7.0
U-235	0.1	0.3	0.5
Ac-227	0.1	0.3	0.5
Ra-228	1.9	32.2	62.0
Th-228	1.9	32.2	61.0

TABLE 3. GROSS ALPHA ACTIVITY CONCENTRATIONS IN THE GROUNDWATER

	Proportion of sampling locations (%)		
	Gross alpha activity concentration <50 mBq/L	Gross alpha activity concentration 50–100 mBq/L	Gross alpha activity concentration >100 mBq/L
Shallow (1–4 m)	66	17	17
Deep (5–8 m)	43	10	47

TABLE 4. RADIONUCLIDE CONCENTRATIONS IN SHALLOW GROUNDWATER AT LOCATIONS ASSOCIATED WITH HIGH GROSS ALPHA ACTIVITY

	Activity concentration (mBq/L)		
	Sample 1 pH = 8.4	Sample 2 pH = 8.0	Sample 3 pH = 8.1
Gross alpha	184 ± 22	930 ± 60	63 ± 29
Gross beta	1570 ± 90	5400 ± 200	1210 ± 140
U-238	87 ± 9	16.2 ± 2.4	
U-234	99 ± 10	18.8 ± 2.7	
Th-230	<0.5	11.0 ± 2.5	
Ra-226	15.7 ± 1.7	43 ± 3	
Th-232	<0.3	13.6 ± 2.9	
Th-228	0.9 ± 0.5	107 ± 16	
U-235	4.4 ± 0.9	0.7 ± 0.4	

In this follow-up measurement campaign, gross beta activity concentrations were also measured. The concentrations, shown in Tables 4 and 5, are all >1000 mBq/L and therefore need further detailed investigation. Unfortunately, the concentrations of ^{40}K and ^{228}Ra were not measured — these radionuclides probably contribute most of the gross beta activity.

In one deep groundwater sample, an extremely high pH of 12.9 was measured. The most plausible explanation for this anomaly is calcium contamination due to human activities.

The annual dose via the drinking water pathway was calculated from the activity concentration measurements. Assuming a daily intake of 2 L, the maximum annual dose was found to be 51 μSv . This value was associated with a sample having a gross alpha activity concentration of 570 mBq/L and a relatively high ^{226}Ra concentration of 250 mBq/L.

TABLE 5. RADIONUCLIDE CONCENTRATIONS IN DEEP GROUNDWATER AT LOCATIONS ASSOCIATED WITH HIGH GROSS ALPHA ACTIVITY

	Activity concentration (mBq/L)						
	Sample 1 pH = 7.7	Sample 2 pH = 7.7	Sample 3 pH = 8.4	Sample 5 pH = 7.7	Sample 6 pH = 8.0	Sample 7 pH = 12.9	
Gross alpha	550 ± 40	64 ± 13	124 ± 014	385 ± 027	66 ± 26	570 ± 60	
Gross beta	1840 ± 100	1420 ± 90	2000 ± 100	3050 ± 130	5800 ± 280	2300 ± 190	
U-238	15.1 ± 2.4		1.5 ± 0.5	9.9 ± 1.7		0.5 ± 0.4	
U-234	14.2 ± 2.3		1.3 ± 0.5	10.1 ± 1.7		1.0 ± 0.5	
Th-230	21 ± 3		1.3 ± 0.6	12.3 ± 2.7		1.3 ± 0.5	
Ra-226	18.2 ± 1.7		12.9 ± 2.0	25.2 ± 2.5		250 ± 20	
Th-232	21 ± 3		0.8 ± 0.4	8.5 ± 2.1		0.7 ± 0.5	
Th-228	98 ± 12		15.9 ± 2.5	81 ± 12		2.2 ± 0.8	
U-235	0.6 ± 0.4		<0.3	<0.5		<0.4	

3.3. Radon assessment

As part of the site investigation, radon concentrations were measured in the various buildings on site as well as in the open air. Neither indoor nor outdoor measurements identified any elevated concentrations — the outdoor concentrations were 5–10 Bq/m³ and the indoor concentrations were 5–40 Bq/m³.

3.4. Dose assessment for the critical working areas

With the help of the GIS map showing the hotspot areas, a dose assessment based on very conservative assumptions was performed for the three most critical occupational exposure situations: a worker filling the silo in the raw material hall, a worker supervising on-site transport activities and a worker sorting the slag material. The annual doses assessed for these situations, including doses received from the inhalation of radon as measured at indoor and outdoor locations, were 0.6, 0.8 and 0.3 mSv, respectively.

4. CONCLUSIONS

Although there was no immediate radiological risk to workers, members of the public or the environment due to the presence of the buried slag, the hotspots detected in the gamma survey of the site and the high activity concentrations, especially for thorium series radionuclides, indicated that radiation protection measures would have to be taken in any future excavation work. Should the use of the site be changed at some time in the future, the impact of the elevated activity concentrations in the groundwater would have to be evaluated. Therefore, the incorporation of a systematic cleanup of the site into the long term strategy should be considered. The material excavated has to be stored in a traceable way at the sites selected recently by the Federal Agency for Nuclear Control for the deposit of NORM residues.

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Invited Paper

APPLICATION OF ICRP RADIATION PROTECTION PRINCIPLES IN EXISTING EXPOSURE SITUATIONS WITH LARGE VOLUMES OF NORM: THE WISMUT CASE STUDY

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Abstract

An outstanding example of an existing exposure situation resulting from large scale uranium mining and processing is the Wismut legacy in eastern Germany. From 1946 to 1990, the Soviet–German stock company SDAG WISMUT produced 216 000 t of uranium and thus became the world’s fourth largest uranium producer at that time. Due to the mining of low grade ore, about 800 million t of NORM residues (also referred to as low level radioactive waste) were deposited at the sites and an area of about 10 000 km² became seriously affected and devastated. Mining and mineral processing took place in a densely populated area, whereby radioactive waste rock piles and tailings management facilities were placed close to residential areas. In such an existing exposure situation, the application of the International Commission on Radiological Protection key radiation protection principles has to meet site specific conditions and requires remedial measures which are far from standard solutions. This is illustrated in the paper. Approaches to decisions on justification of remedial activities are presented together with a description of the optimization process.

1. INTRODUCTION

Following the end of the Cold War, a number of States in Eastern Europe and Central Asia found themselves confronted with huge legacies left behind by uranium mining and processing operations. Uranium production in these countries had been conducted with complete disregard for the protection of humans and the environment. The legacies were enormous both with regard to the quantities of residues involved and the areas used for their storage. As a result of political changes in the early 1990s, many operators of former uranium mines either no longer existed as legal entities or were only contingently liable.

From a radiological perspective, therefore, the situation at hand constituted an existing exposure situation and its remediation required a modified application

of the basic radiation protection principles recommended by the International Commission on Radiological Protection (ICRP). These principles had been set out by the ICRP in Publication 6 [1], where the term “intervention” was assigned to the remediation of existing exposure situations. In the most recent ICRP recommendations (Publication 103 [2]), existing exposure situations are explicitly addressed along with planned exposure situations and emergency exposure situations. Even though uranium production residues are included within the definition of NORM, such residues are part of the nuclear fuel cycle and there is a tendency, therefore, to continue to categorize them as “low level radioactive waste”. The application of the ICRP basic principles to the management of those residues shows numerous parallels with other existing exposure situations involving legacies from non-uranium NORM industries.

This paper provides a review of how the basic principles of radiation protection were applied to the remediation of the legacies left behind by the former Soviet–German stock company SDAG Wismut. In terms of extent and severity of their environmental impact, the legacies from the Wismut operations constitute an exceptional example of an existing exposure situation involving large quantities of materials with elevated levels of radionuclides of natural origin. During the period 1946–1990, SDAG Wismut produced a total of 216 000 t of uranium in eastern Germany, ranking the company as the world’s fourth largest uranium producer at that time. Mining and processing of low grade uranium ores generated more than 800 million t of NORM residues (waste rock and chemical process tailings). Deposited at the mining and mineral processing sites, these residues had a dramatic environmental impact across an area of more than 10 000 km². Unlike the situation in other major uranium producing countries such as the United States of America, Canada and Australia, Wismut’s mining and processing sites were located in densely populated areas. As a result of this, waste rock piles and tailings ponds were established near residential areas, in some cases in the direct vicinity of homes. Since 1991, the newly founded national corporation Wismut GmbH has been conducting the rehabilitation of uranium mining legacies in eastern Germany.

2. APPLICATION OF THE ICRP PROTECTION PRINCIPLES TO THE WISMUT PROJECT

2.1. Justification

In terms of the principle of justification, any decision which leads to a modification of the exposure (including any decision on remedial action) should do more good than harm. Harm is understood to include negative social

implications as well as costs. With regard to the justification of remediation to be conducted at the former uranium mining sites in eastern Germany, the German Commission on Radiological Protection (SSK, Strahlenschutzkommission) made a landmark recommendation in 1992 [3] by specifying an annual effective dose of 1 mSv as the primary criterion to be applied to the use of former uranium mining related areas, buildings, and mine residue deposits. Where this level was exceeded, remedial action was to be taken into consideration from a radiation protection point of view. The 1 mSv criterion applies to exposure additional to natural background exposure. Initially, doses from indoor radon were not included.

The German Federal Ministry for the Environment, Nature Conservation, Building and Nuclear Security (BMUB, Bundesministerium für Umwelt, Naturschutz, Bau und Reaktorsicherheit) issued Calculation Basis Mining for the determination of effective doses from environmental radioactivity attributable to mining related activities [4]. The Calculation Basis evaluates “inhalation of radon” on an equal footing with other pathways in establishing the effective dose attributable to mining related activities and comparing it with the 1 mSv criterion. Outdoor radon concentrations are added to indoor levels on the basis of a reference person’s assumed dwelling period of 7 000 h. The 1 mSv criterion lies at the lower end of the range of criteria recommended by the ICRP below which an intervention in the event of radioactive residues in the human environment would “probably not be justified”. ICRP Publication 60, for instance, specified an annual dose criterion of 10 mSv.

The 1 mSv criterion for the justification of remediation discussed above can be regarded in today’s terms as being equivalent in some respects to the “reference level” introduced in ICRP Publication 103 for optimization in existing exposure situations. The ICRP recommends that such reference levels be set somewhere in the range 1–20 mSv. The remediation criterion of 1 mSv applied to the Wismut remediation project is thus at the lower bound of this range and is also below the reference level recommended for radon (10 mSv). The ICRP reference levels are for the most part significantly in excess of the 1 mSv criterion adopted by the SSK. Nevertheless, it should be noted that for the purpose of remediating the legacies left behind by SDAG Wismut, a dose criterion in excess of 1 mSv would not have received acceptance in Germany (and will not in the foreseeable future). Many factors contributed to the establishment of the 1 mSv criterion: the acknowledgement of a range of variations in the natural background radiation, the adoption of a 1 mSv criterion in the United States of America [5] and Canada [6], as well as the standards laid down in the Radiation Protection Ordinance of the former German Democratic Republic [7], which is still in effect for the Wismut remediation project. On the other hand, the United States of America, Canada and

many other States do not take inhalation of radon into account when applying the 1 mSv criterion.

Examples of two typical scenarios for exposure of the public in the vicinity of a nearby non-remediated mine residue deposit (Schlema-Alberoda site) and of a large scale tailings management area are described in Sections 2.1.1 and 2.1.2. The exposure in Example A is principally dominated by the inhalation of radon, while Example B illustrates an exposure scenario for the water pathway. These examples demonstrate that annual doses in excess of 1 mSv must either be attributed to the radon inhalation pathway (as a rule) or, in the case of the water pathway, are the result of conservative assessment approaches such as the consumption of baby food which was prepared with contaminated water. There are, however, many structures and land areas which do not have a radiological impact on the environment and would thus not result in annual effective doses to the general public in excess of 1 mSv. In spite of that, the remediation of such structures and land areas is justified, a priori, to comply with the requirements of German mining law, but also because of geotechnical risks and of environmental impacts due to heavy metals and other chemically toxic (i.e. non-radiological) contaminants. Stakeholder interests are also an issue. In many cases, these factors rather than radioactivity are the real driving forces behind the remedial actions.

2.1.1. Example A: Exposure in the vicinity of a non-remediated mine residue deposit

Figure 1 depicts the findings of an exposure pathway analysis established for an infant in the age group 2–7 years and for an adult reference person who live permanently in the immediate neighbourhood of a large non-covered mine residue deposit and use seepage water to irrigate their garden. The relevant exposure pathways to be considered in the present case include:

- (a) Exposure by ingestion of locally grown garden crops representing 25% of total consumption (GP);
- (b) External exposure by soil gamma radiation (Ext);
- (c) Exposure by inhalation of long lived alpha emitters in dust (LLA);
- (d) Exposure by inhalation of radon and its short lived decay products (Rn/DPr);
- (e) Exposure by direct ingestion of waste rock material (Dir-Ing).

In accordance with Ref. [4], an annual dwelling period of 7000 h near the mine residue deposit and a total annual sojourn of 250 h (small child) or 100 h (adult) were assumed.

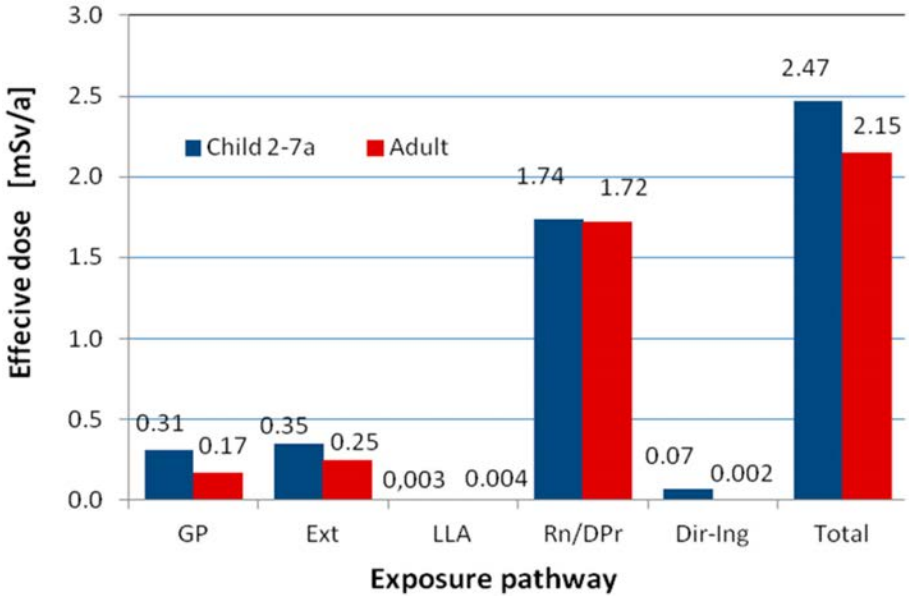


FIG. 1. Typical exposures from non-reclaimed uranium mining waste rock piles.

2.1.2. Example B: Exposure due to the use of water by seepage from a tailings management facility

This example considers the use of a watercourse for the irrigation of field and garden crops, livestock watering (each contributing 25% of locally grown food to annual consumption rate) and as drinking water (100%). In addition, consumption of fish (25%) is assumed. The radionuclide vector, which has to be determined in full, has a dominating influence on the calculation result. The vector considered in this case was identified as a small creek running across an area situated between two large tailings management facilities of Wismut. The striking feature here is the dominant contribution of uranium nuclides (see Table 1). A ^{238}U activity concentration of 5.2 Bq/L equates to 0.42 mg/L of elemental uranium.

TABLE 1. RADIONUCLIDE VECTOR FOR A WATERCOURSE IN THE SURROUNDINGS OF A TAILINGS MANAGEMENT FACILITY

	Activity concentration (Bq/L)
U-238	5.2
U-234	6.1
Th-230	0.17
Ra-226	0.02
Pb-210	0.025
Po-210	0.025
U-235	0.24
Pa-231	0.015
Ac-227	0.015

Figure 2 shows the results of the exposure pathway analysis for infants (age less than 1 year) and for adults. The results give details of the dose contributions by the exposure pathways drinking water (DW), fish consumption (Fi), consumption of mothers' milk and baby food (MM_BF), consumption of field and garden crops other than cereal products (FGP), and the consumption of dairy and meat products (DMP).

2.2. Dose limitation

During operations to remediate the legacies of uranium mining, Wismut follows common practice worldwide in that it complies fully with regulations on industrial safety, radiation protection and nature conservation (i.e. avoidance of unnecessary exposure, ventilation of underground mine workings, prevention of the spread of contamination and minimization of the radiological impacts of the remedial work). This leads to moderate radiation exposure scenarios both for the workers involved and for the general public. These remediation actions are in essence 'planned activities' that are subject to the requirements for planned exposure situations. Therefore, under the Wismut environmental reclamation

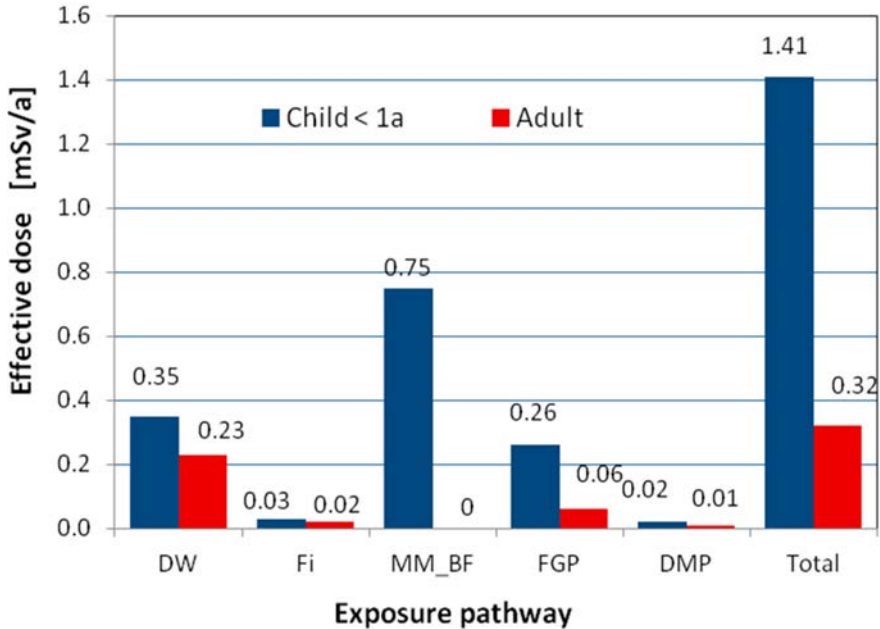


FIG. 2. Typical pattern of radiation exposure via the water pathway.

project, just as with other planned activities, dose limits rather than reference levels are applied to workers and the general public. These limits are:

- 1 mSv/a for the general public;
- 6 mSv/a for workers employed above ground in the remediation of contaminated land and waste rock piles, as well as in demolition and disassembly work;
- 20 mSv/a for workers employed underground or at tailings sites;
- 400 mSv as a lifetime occupational dose (with special provisions in case of such a limit being exceeded).

Data recorded at the Wismut site show that exposures of workers employed in the remediation of contaminated land and waste rock piles as well as in demolition and disassembly work were quite low. The materials being handled had low activity concentrations, usually not exceeding 1 Bq/g for the dominant radionuclide ^{226}Ra . These workers received annual doses of 0.5–2 mSv. Workers handling tailings (residues from the chemical processing of uranium ore) with ^{226}Ra activity concentrations of up to about 10 Bq/g could receive higher doses of up to 4 mSv (but noting that such workers are not permanently in direct contact with the tailings). Workers engaged in underground work in preparation

for mine flooding received the highest doses. Unless there was optimized ventilation of the mine workings, such workers would have received annual doses exceeding 10 mSv. The actual doses received in 2011 are shown in Fig. 3. The doses were determined assuming an average of 1400 working hours per year (maximum 1540 h), a fixed gamma dose rate of 0.2 $\mu\text{Sv/h}$, a dust activity concentration of 14 mBq/m^3 for each long lived alpha emitter in equilibrium (maximum 21 mBq/m^3) and a radon progeny potential alpha energy concentration of 3.5 MeV/cm^2 (maximum 11 MeV/cm^2).

During the past ten years of remedial action conducted by Wismut, annual doses received by members of the public (attributable to the remediation) did not exceed 1 mSv. Ingestion of contaminated garden and field crops turned out to be the most prominent exposure pathway because of dust generated by excavation and relocation of materials and its subsequent migration to horticultural and agricultural land. Dust abatement measures are in place to control this pathway of exposure.

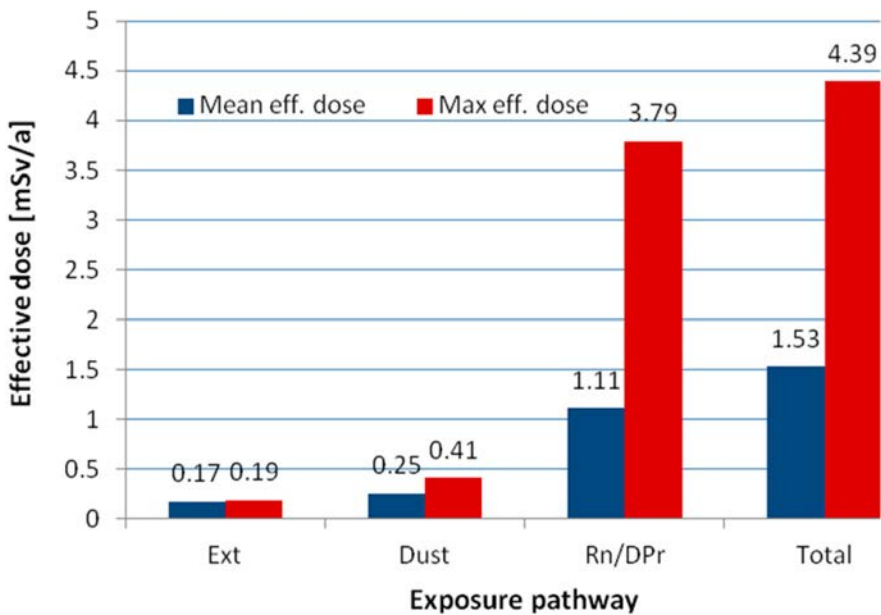


FIG. 3. Doses received by underground workers in 2011.

2.3. Optimization

ICRP Publications 60 and 103 give extensive coverage of the implementation of the optimization principle in large scale remediation projects. The probability of exposure, the number of exposed persons, and the amount of their individual doses are to be kept as low as reasonably achievable (ALARA principle), social and economic factors being taken into account [1, 2]. Issues to be addressed when remediating large uranium mining legacies include:

- (a) What cover thickness will be required, which materials will be needed and what cover layer design will be applied?
- (b) Which residual contamination will be acceptable in the effluents from water treatment plants?
- (c) To what extent do radioactive materials have to be excavated and removed in terms of the proposed remediation of contaminated land?

The optimization of actions to remediate mining legacies is certainly among the most demanding challenges to be faced, for the following reasons:

- (a) Dose reduction occurs within the range of variation of natural background radiation. This radiation is ubiquitous and can be measured with limited accuracy only. Because of that, the net benefit (dose reduction) achieved is difficult to ascertain.
- (b) Remediation costs often increase supralinearly with gains in net benefit (i.e. further dose reductions will inevitably be associated with increasingly cost intensive measures).
- (c) In addition to environmental aspects (not limited to radiological environmental impacts) optimization also has to take account of social and economic aspects, sustainability issues and in particular the interests of concerned parties such as residents, local communities, federal states and regulatory agencies.

The company Brenk Systemplanung GmbH is a consultant to the BMUB. In reviewing the fundamental decisions on rehabilitation measures to be implemented by Wismut, Brenk used an approach to the cost–benefit analysis which was based primarily on the determination of the collective dose and on the number of harmful events deduced from risk factors (e.g. cancer incidences, loss of life expectancy) [8]. The amount of harm was monetarized by comparison with the amount society would be prepared to pay in order to attain a certain degree of harm reduction (described by what is known as the alpha value). The identified

optimum rehabilitation option was the one that reduced the sum of remediation costs and harm equivalent costs to a minimum [8].

A cost–benefit analysis conducted along these lines does not go undisputed, primarily because of the uncertainties in the intermediate results (e.g. in the calculation of realistic collective doses) and in the assumptions to be made (e.g. the quantification of the alpha value, the integration period for the post-remedial condition (200 or 1000 years?) and the application of dose cut-off criteria). Although the ICRP, in Publication 103 [2], expresses the need for caution in calculating collective doses by summing small doses received by a large population, the procedure used at the time in Ref. [8] has proven its worth as a useful sensitivity analysis tool in arriving at fundamental decisions for large and complex rehabilitation objects. Cost relevant parameters were more easily identified and decision making became more transparent. At a later stage, when taking an extended approach, Brenk also introduced non-radiological risks into the cost–benefit analysis [9]. In preference to a pure cost–benefit analysis, Wismut successfully applied a multiattribute analysis which, in addition to costs, also considered ‘soft factors’ such as social factors, aspects of licensing and planning regulations, and acceptance issues, which in the broader sense means involvement of all concerned parties.

2.4. Protection of the environment

With regard to the protection of the environment, Wismut used to comply exclusively with the evaluation criteria as set out in ICRP Publication 60 and according to which the environment is adequately protected when humans are protected [1]. Recent approaches set out in ICRP Publications 103 [2] and 108 [10] focus in particular on the protection of biota by introducing the concept of reference animals and plants. The focus is on the protection of species against the negative impacts of radiation with regard to mortality, morbidity, reduced reproductive success and chromosomal damage.

Evaluations of the impacts of Wismut legacies on the environment use such approaches already with regard to aquatic biocoenosis in order to quantify the degree of surface water pollution by heavy metals (including uranium) and other toxic substances. However, these approaches deal with the chemotoxicological effect of these contaminants and not with radioactivity. Given the sheer size of the Wismut project, the application of state of the art radiological assessment tools in line with ICRP Publication 108 [10] promises to become an interesting field of activity for ambitious radiation protection experts in years to come.

3. CASE STUDY: RADON

As already outlined in Section 2.1, the inhalation of radon and its decay products can contribute significantly to the exposure of members of the public. Crucial parameters in this regard are the radon source terms (e.g. waste rock piles, near surface mine workings and upcast shafts), the location of sources in relation to population centres and local conditions of dispersion.

3.1. Radon from waste rock piles

When uranium mining was terminated, the radon situation prevailing at the Schlema-Alberoda site was a very complex one. As investigations had revealed, radon release from huge steep mine dumps was caused mainly by convective air currents within the dumps. These currents in turn resulted from temperature and pressure differences within the pile which were distinct from outside conditions. As a consequence, daily average radon concentrations measured at the 'toes' of large waste rock piles locally exceeded 2500 Bq/m³ during the summer. An exposure pathway analysis of annual average concentrations exceeding 1000 Bq/m³, performed along the lines of that described in Ref. [4], indicated an annual effective dose of more than 20 mSv, significantly above the 1 mSv criterion for the justification of remedial action.

Since relocation of the waste rock piles at the Schlema-Alberoda site was not feasible within reasonable technological and spending limits, Wismut implemented the concept of in situ remediation consisting of regrading, covering and vegetating. During the optimization process, a standard cover design of 1 m thickness was adopted on the basis of results obtained from thorough investigations into convective radon release (radon exhalation and gas permeability measurements, and tracer gas experiments). In the frame of optimization, collective doses via the atmospheric and aquatic pathways were considered and compared with costs associated with various cover designs (see Section 2.3). Over and above this, due regard was paid to the interests of residents in the immediate vicinity of the waste rock piles, in particular during regrading operations. Figure 4 illustrates the basic cover design to minimize radon exhalation from waste rock piles at the Schlema-Alberoda site. The positive remedial effect is demonstrated in Fig. 5.

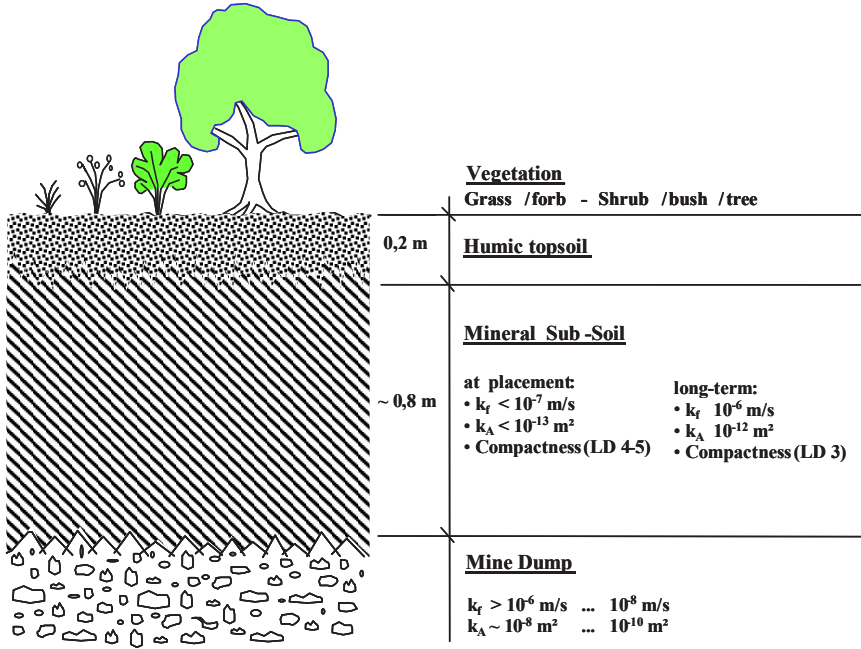


FIG. 4. Cover design to control convective radon release from steep waste rock piles.

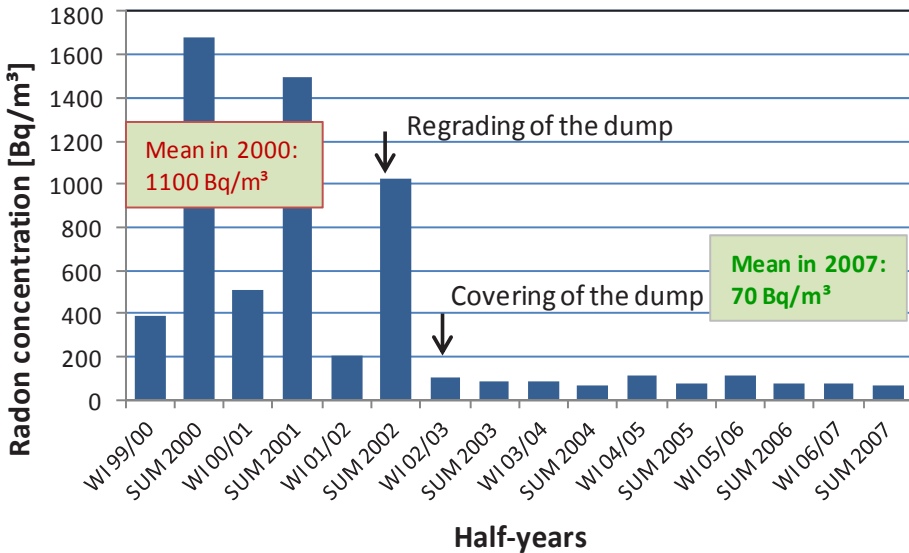


FIG. 5. Outdoor radon concentration at the toe of a waste rock pile prior to remediation, during intermediate regrading, and in the post-remedial condition.

Despite the positive example illustrated in Fig. 5, there are some less positive experiences to be aware of. Observations showed that a couple of years after completion of the 1 m thick standard cover, the radon attenuation effect of the cover tended to decline locally at some of the waste rock piles. The probable cause was deep-rooting plants and burrowing animals. Both processes, known as bio-intrusion, result in the formation of large void spaces within the cover which cause a new increase in radon exhalation. After a period of 5–10 years following the completion of cover construction, elevated radon concentrations were again observed locally at the toes of some waste rock piles at the Schlema site amounting to levels which, according to the Calculation Basis Mining [4], correspond to annual doses of about 2–3 mSv. Wismut is currently investigating various approaches to control such local effects. Here again, the ICRP principles of justification and optimization have to be applied in an iterative process. In doing so, the reference nature of the 1 mSv criterion and stakeholder interests have to be taken into account.

3.2. Control of radon release from underground mine workings

Not only mine dumps, but also underground mine workings are potential sources of radon release at the Schlema-Alberoda site. Once the mine is flooded, the final water level will be such that not all the near surface mine workings will be filled. For the time being, the mine workings continue to be extensively ventilated thus creating a permanent negative pressure of the mine air in relation to the outdoor atmospheric pressure. Experiments performed by Wismut have demonstrated that, should the ventilation be shut down, radon concentrations in nearby dwellings would increase rapidly. During these experiments, concentrations in excess of 20 000 Bq/m³ were observed in basements. Tracer gas tests have provided evidence that these concentration levels are due to convective air currents from the mine. The observed levels of radon concentration are such that remedial action is clearly justified. With a view to identifying the optimum remedial approach, Wismut investigated the following options:

- Individual remedial solutions for dwellings;
- Creation of a downward negative pressure by a decentralized ventilation scheme;
- Creation of a downward negative pressure by a centralized ventilation scheme.

Based on a multiattribute analysis, an optimized central ventilation design was identified as the long term solution for control of mine related radon. Factors

that led to this decision included the large number of dwellings concerned, their location, and legal issues requiring concerned homeowner consent.

4. CONCLUSIONS

The following conclusions may be drawn from more than 20 years of ongoing remediation of the Wismut legacy sites:

- (a) Application of ICRP basic radiation protection principles to the remediation of mining residues in existing exposure situations requires that, from the outset, criteria are set for the justification of remedial actions. Such actions must be feasible within reasonable technological and spending limits and acceptable to the regulating bodies, to the remediation company, to the public, as well as to other concerned parties.
- (b) The same applies to the approach to be chosen for identifying optimized remedial solutions. Optimization must be transparent and comprehensible. For this purpose, accepted optimization approaches are to be implemented as well as pertinent calculation guidelines and modelling.
- (c) Within the framework of justification and optimization, dose levels have the character of reference values. They have to be established on the basis of standardized and officially approved calculation guidelines. This will ensure that they are comparable and acceptable at the same time.
- (d) In the case of remediating the legacy sites of the former Wismut uranium mining operations, the ranges of reference levels as proposed by the ICRP were not fully explored. Instead, a remediation criterion was set at the lowest (and hence the most stringent) level of 1 mSv. While this approach has also followed in other countries for the remediation of mining related legacy sites, the inclusion of radon exposure in the 1 mSv remediation criterion level is not consistently applied in such countries.
- (e) When remedial measures fail to achieve target values or when conditions exceeding the target values re-emerge in the longer term, the appropriateness of the target value as a remediation criterion needs to be reassessed by repeating the justification and optimization process for existing exposure situations. Within this process, decisions on further remedial measures will in turn have to be made.
- (f) Protection of the environment in accordance with ICRP Publications 103 [2] and 108 [10] will be increasingly integrated into the assessment of remedial actions. By its sheer size, the Wismut remediation project will provide an ample field of activity for German radiation protection experts.

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THE DISTRIBUTION CHARACTERISTICS OF ^{222}Rn AND ^{220}Rn CONCENTRATIONS IN SOIL GAS IN ZHONGSHAN CITY, GUANGDONG PROVINCE, CHINA

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Abstract

A mapping survey of ^{222}Rn and ^{220}Rn concentrations in soil gas was conducted for the first time using a RAD7 portable radon monitor at 67 locations across an area of 1800 km² in Zhongshan City (ZSC), Guangdong Province. It was found that ^{222}Rn concentrations increased with increasing depth, whereas ^{220}Rn concentrations were almost unchanged at different depths. The sites with high ^{222}Rn values were located mainly in granite outcrops, while those with low values were located in a sedimentary region. The distribution patterns of ^{220}Rn coincide with the patterns of activity concentration of ^{232}Th in soil.

1. INTRODUCTION

Radon is one of the most hazardous radioactive elements in the indoor environment. Half of the natural radiation exposure of humans results from inhalation of the short lived decay products of radon (mainly ^{222}Rn). Inhalation of ^{222}Rn and its progeny gives rise to an average annual dose worldwide of 1.2 mSv [1]. The ratio of annual dose from ^{220}Rn and its progeny to that from ^{222}Rn and its progeny was reported to be 6% in the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) 1993 report [2] and 9% in the UNSCEAR 2000 report [1]. Radon survey results have shown that soil gas is a major source of radon in ground floor rooms of buildings in China and other countries.

It is known that Yangjing County (YC) is one of several high natural background radiation areas in China [3]. The dominant geological characteristics in such areas are the presence of elevated levels of monazite and uranium mineralization in soil and rocks. Research has shown that ^{220}Rn progeny concentration levels are relatively high and very variable in most areas of YC, Guangzhou City (GC) and Zhuhai City (ZC) [4, 5]. In YC, the average indoor

^{222}Rn and ^{220}Rn concentrations measured with passive CR-39 radon–thoron cups were 263 and 207 Bq/m³, respectively, with corresponding equilibrium equivalent concentrations (EECs) of 41.8 and 4.7 Bq/m³. In ZC, the ^{222}Rn and ^{220}Rn concentrations were 60.4 and 127.9 Bq/m³, respectively, with corresponding EECs of 52.9 and 4.0 Bq/m³ [6, 7]. In a survey of the soil in ZC conducted in 2003–2002 using a portable gamma spectrometer with an NaI(Tl) detector [8], the average activity concentrations of ^{238}U and ^{232}Th were found to be 0.0858 ± 0.0316 and 0.1598 ± 0.049 Bq/g, respectively. In the 1980s, a survey of the level and distribution of radionuclides in soil in China was performed nationwide. The survey results gave a geometric mean ^{232}Th activity concentration of 0.0847 Bq/g in Guangdong Province [9], almost 3.4 times higher than the average value worldwide (0.025 Bq/g).

Relatively high ^{232}Th activity concentrations, similar to those found in YC, had been observed in soil in ZC and Zhongshan City (ZSC). According to the results of a geological survey, these locations were associated with outcrop areas composed mainly of Middle and Late Jurassic and Cretaceous biotitic granite and granodiorite [10]. Therefore, during a radon mapping programme carried out in 2011, ^{222}Rn and ^{220}Rn activity concentrations in soil gas were measured in these areas.

2. INSTRUMENTS AND METHODS

The concentrations of ^{222}Rn and ^{220}Rn in soil gas were determined in ZSC by using a RAD7 radon monitor, which contains a solid state ion-implanted planar silicon detector and built-in pump with a flow rate of 1 L/min. It has desiccant (CaSO₄) tubes and inlet filters (pore size 1 μm) that block fine dust particles and radon daughters from entering the radon test chamber. The internal sample cell of the RAD7 is a 0.7 L conducting hemisphere with a 2200 V potential relative to the detector which is placed at the centre of the hemisphere. The RAD7 discriminates between ^{222}Rn and ^{220}Rn on the basis of alpha particle energies from the decay of ^{218}Po and ^{216}Po .

A thick walled stainless steel tube of 8 mm internal diameter and 110 cm long was driven into the ground to a depth of 80 cm at every sampling point. The sampling tube outlet was connected to the inlet of the RAD7 via a silicone pipe. Soil gas was transported into the internal sample cell by the built-in pump and was measured in sniff mode with a 3 min sampling time, while the pump ran continuously. In all measurements, the cycle time at each site was at least 30 min depending on soil permeability. Three sites were observed at every location. The final result was the average of the many measurements made at these three sites.

Soil samples were collected at depths ranging from 0 to 4 cm during the radon survey. The concentration of ^{232}Th were analysed in the laboratory using a high purity germanium gamma spectrometer, with a relative efficiency of 32% and an energy resolution of 1.76 keV at 1332 keV. For the determination of radionuclides and their activities, the GammaVision 32 software package was used.

3. OVERVIEW OF SURVEYED AREA

The surveyed area covered 1800 km², between longitudes 113°08' E and 113°37' E and latitudes 22°10' N and 22°46' N. A simplified geological map was extracted from a 1:500 000 digital geological map in the database of the China Geological Survey. Measurement locations are shown in Fig. 1. The surface deposits that formed since the Late Quaternary Period consist mainly of arene, medium sized arenilla and arenilla, or silty sand and silty clay–clay. The intrusive rocks include the Middle and Late Jurassic and Cretaceous biotitic-granite and granodiorite [10].

The surveyed area can be divided into three geomorphologic regions: the coastal mountain region; the hill and valley region; and the plain region. The mountain and hill regions are mainly covered with granite and the plain region with Quaternary sediments. The climate is oceanic monsoonal with an average annual temperature of 22.4°C and an average relative humidity of 79% in ZSC and ZC.

4. DISTRIBUTION OF ^{222}Rn AND ^{220}Rn CONCENTRATIONS

4.1. Depth variation

It is already known that ^{222}Rn concentrations in soil gas vary with depth as a result of radon exhalation at the soil surface, but it is not clear whether ^{220}Rn concentrations also vary in this manner. Three representative experimental sites were selected: CG05 (original weathered granite outcrops); NLCKC (all-weathered granite); and YMC (sandstone). The measured ^{222}Rn and ^{220}Rn concentrations are shown in Table 1, from which the following observations can be made:

- (a) The ^{222}Rn concentrations increase with increasing depth.
- (b) There is no obvious increase in ^{220}Rn concentration with depth.

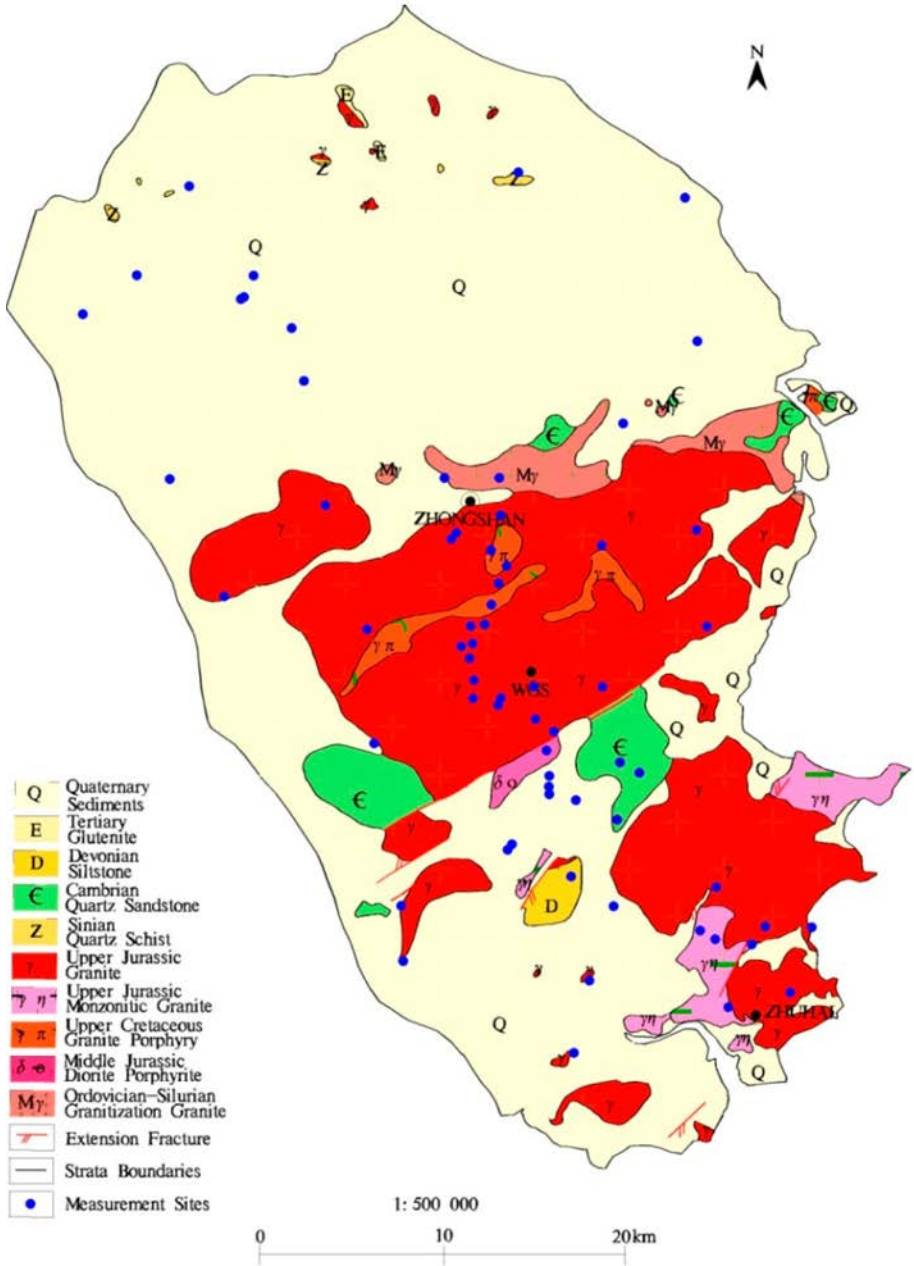


FIG. 1. Geological map of the surveyed area.

CHARACTERISTICS OF ^{222}Rn AND ^{220}Rn CONCENTRATIONS IN SOIL GAS

- (c) The ^{222}Rn and ^{220}Rn concentrations differ significantly between sites, especially between CG05 and the other two sites.
- (d) The reason for the high ^{222}Rn concentration at YMC needs to be investigated in detail because normally the concentration is not so high in a sandstone region.
- (e) Another surprising result is that the lowest value of ^{220}Rn concentration was recorded at a depth of 140 cm at YMC.

TABLE 1. VARIATION OF ^{222}Rn AND ^{220}Rn CONCENTRATIONS WITH DEPTH

Depth (cm)	Activity concentration (Bq/m ³)					
	CG05		NLCKC		YMC	
	Rn-222	Rn-220	Rn-222	Rn-220	Rn-222	Rn-220
20	18 500	257 000	15 200	153 500	38 900	115 300
40	37 200	261 500	22 500	172 000	80 000	139 000
60	57 100	229 300	32 700	186 000	132 200	154 000
80	53 800	263 500	—	—	193 200	177 300
100	65 800	267 200	47 500	145 800	227 500	165 200
120	—	—	50 400	167 000	229 900	127 300
140	70 200 ^a	232 400	65 000	182 700	181 500	68 900
160	78 800 ^b	289 300	67 800	172 300	266 500	164 200

Note: CG05 — original weathered granite outcrops; NLCKC — all-weathered granite; YMC — sandstone.

^a Depth 130 cm.

^b Depth 150 cm.

4.2. Profile characteristics

The profile of ²²²Rn and ²²⁰Rn concentrations along a typical geological section, the Wuguishan Mountain (WGS), is shown in Fig. 2. It is evident that the concentrations of ²²⁰Rn are higher than those of ²²²Rn except at Site CG17 where the surface deposits consist mainly of Late Quaternary sediments. The range of concentrations is about 2–3 times the average value.

4.3. Contour maps

A summary of the concentrations of ²²²Rn and ²²⁰Rn in the surveyed areas, as measured at 67 sites, is shown in Table 2.



FIG. 2. Profile of ²²²Rn and ²²⁰Rn concentrations along a typical geological section.

TABLE 2. SUMMARY OF ²²²Rn AND ²²⁰Rn CONCENTRATIONS AT 67 SITES

	Activity concentration (Bq/m ³)			
	Min.	Av.		Max.
		Sedimentary areas	Granitic areas	
Rn-222	270	37 500 ± 49 860	140 740 ± 201 780	1 199 000
Rn-220	6 650	23 300 ± 25 840	294 420 ± 81 360	461 000

Contour maps of ^{222}Rn and ^{220}Rn concentrations in ZSC are shown as in Figs 3 and 4, respectively. Generally speaking, the sites with high ^{222}Rn values are located in the granite outcrop regions, but the areas with high values are relatively small. Low concentrations of ^{222}Rn and ^{220}Rn are caused by the low porosity and permeability of the moist compact clay soil. However, high concentrations of ^{222}Rn and ^{220}Rn can be found when the surface soils are overlaid with fresh decomposed granite products, even though these sites are located in a Quaternary sediment region. Increased ^{222}Rn concentrations in soil gas occur when the deposit thickness of weathered granite products with a high ^{226}Ra content reaches 100 cm. A ^{222}Rn anomaly was found in a small weathered granite outcrop in the southern part of the surveyed area, with concentrations reaching 1 199 000 Bq/m³. The next highest concentration (486 000 Bq/m³) was recorded at a site in the western part of Wuguishan Mountain (WGS). The ^{220}Rn distribution patterns coincide with those of ^{232}Th activity concentrations in the soil [11].

5. CONCLUSIONS

A large scale survey of radon in soil gas has shown that the ^{222}Rn and ^{220}Rn concentrations are relatively high in the regions of granite outcrops in ZSC. The ^{220}Rn distribution patterns coincide with the patterns of ^{232}Th activity concentration in the soil. This may lead to higher concentrations of indoor ^{222}Rn and ^{220}Rn and their progeny. The activity concentrations of ^{226}Ra and ^{232}Th in the granitic rocks in ZSC are almost the same as those in YC and ZC. Therefore, the high ^{222}Rn and ^{220}Rn concentrations in soil gas in YC and ZC should be investigated.

ACKNOWLEDGEMENTS

This research was supported by the Natural Science Foundation of China (No. 40174096). The authors would like to express their thanks to Tian Gui and Peng Mali, graduate students in the authors' laboratory, for their hard work in the field.

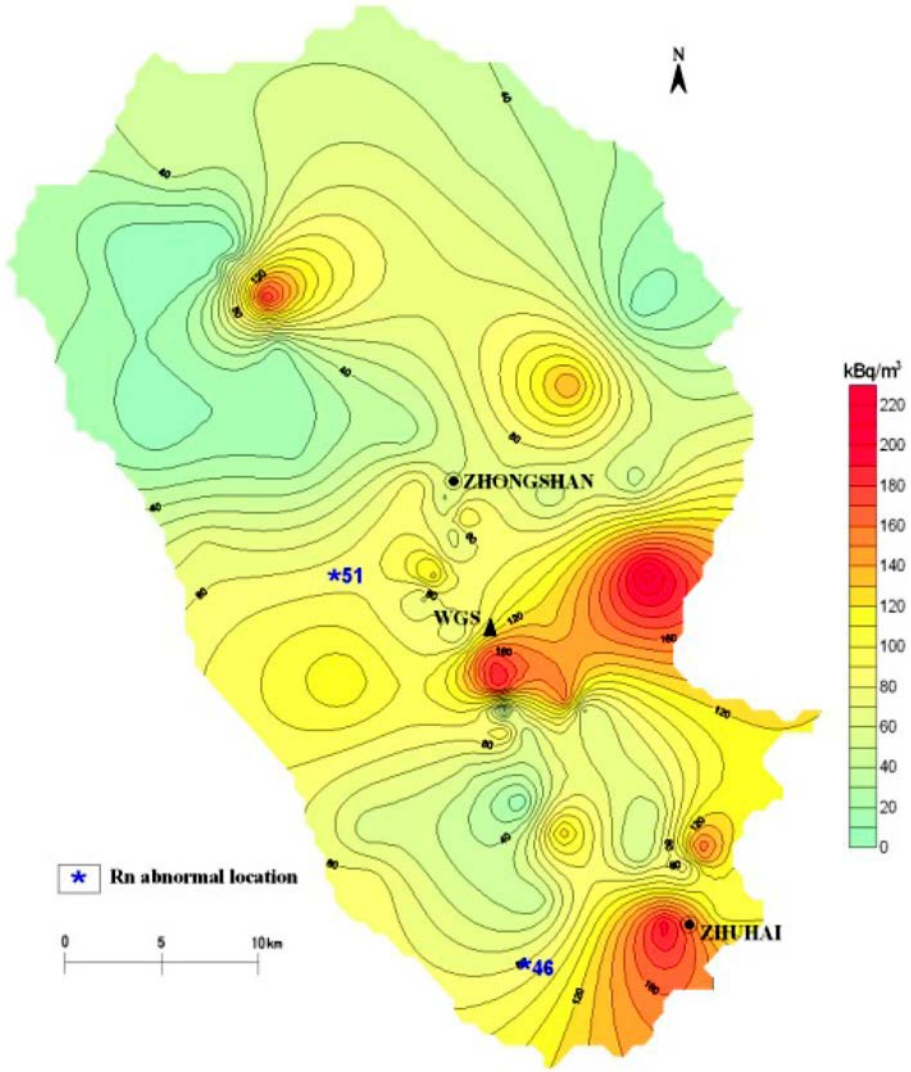


FIG. 3. Contour map of ^{222}Rn concentration in soil gas.

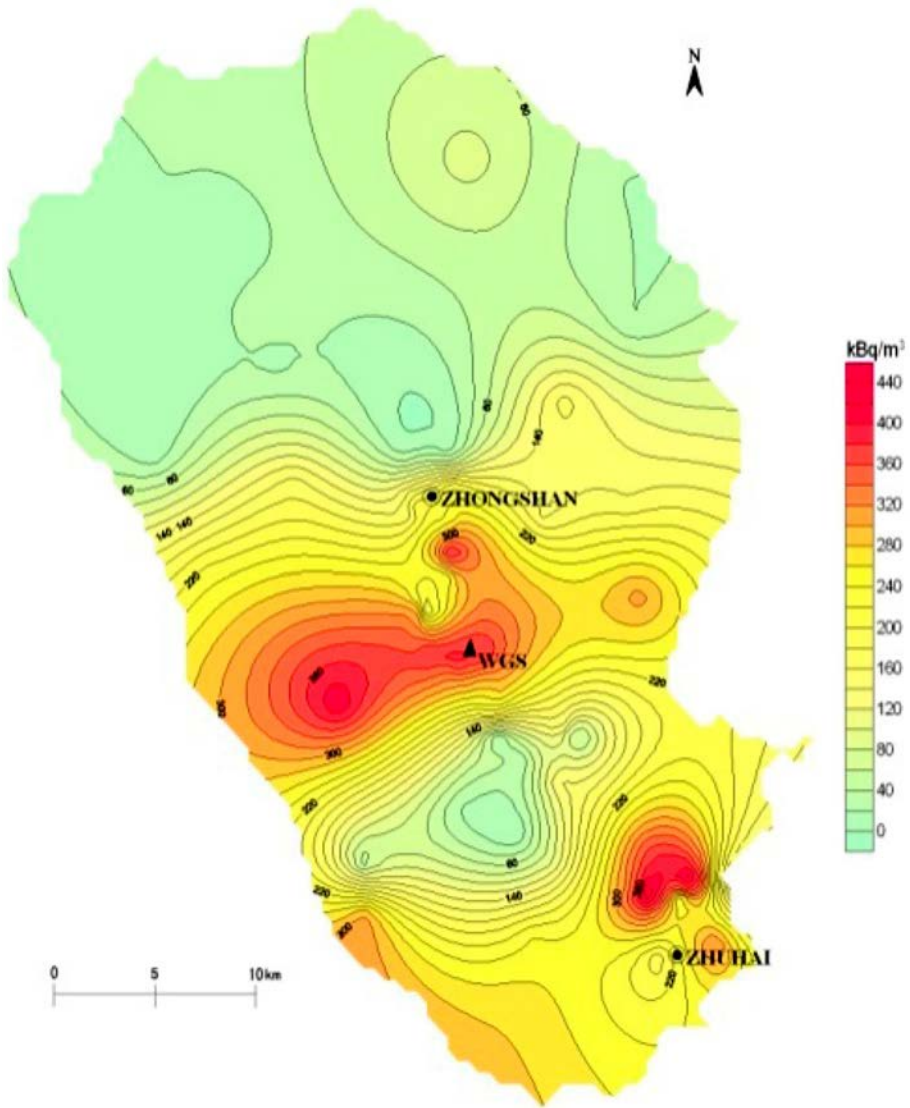


FIG. 4. Contour maps of ^{220}Rn concentration in soil gas.

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DOSIMETRIC EVALUATION OF THORON EXPOSURE IN THE THREE TYPICAL RURAL INDOOR ENVIRONMENTS OF CHINA

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Abstract

Brick houses, mud houses and caves are the three typical types of rural home in China, usually characterized by uncoated surfaces from where thoron gas is easily exhaled. In order to evaluate the dose from inhalation of thoron in these indoor environments, field measurements were carried out using portable devices for measuring the concentration and size distribution of thoron progeny. The dose conversion factor and annual doses from inhalation of thoron in these environments were calculated using dosimetric methods. When comparing the results with the those obtained in an urban indoor environment, it was found that the thoron progeny size distribution parameters for rural indoor environments (AMAD: 0.07652 μm ; GSD: 2.73) were much lower than those for urban indoor environments (AMAD: 0.115 μm ; GSD: 2.0), which makes the thoron exposure dose conversion coefficient for rural environments (307.37 nSv per $\text{Bq}\cdot\text{m}^{-3}\cdot\text{h}^{-1}$) much higher than that for urban indoor environments (113.44 nSv per $\text{Bq}\cdot\text{m}^{-3}\cdot\text{h}^{-1}$).

1. INTRODUCTION

Radon (^{222}Rn) has been widely investigated for many years because it contributes most of the dose to the human body from exposure to natural radiation [1]. Thoron (^{220}Rn) is a radioactive isotope of radon, but due to its short half-life (55.6 s) and the lack of measurement methods as well as measuring results, it has been ignored for a long time. With the improvement of thoron measuring methods and the availability of thoron related data, thoron exposure in some indoor environments has begun to attract attention [2–3].

Brick houses, mud houses and caves are the three typical types of rural home in China, usually characterized by uncoated surfaces from where thoron gas is easily exhaled. Thoron gas concentrations can reach quite high levels, especially in some high background regions such as Yangjiang, Guangdong Province [4], and it is appropriate that thoron exposure in rural homes should be evaluated. In indoor environments, the state of equilibrium between thoron gas and its progeny is highly variable in both space and time. Therefore, for the purposes of dose evaluation, direct measurement of thoron progeny seems to be the most reasonable approach [5].

The dose conversion factor which characterizes the dose to the respiratory tract per unit exposure to thoron progeny changes with the unattached fraction of thoron progeny and their size distribution [6]. Since the unattached fractions of ^{212}Bi and ^{212}Pb , which contribute nearly all the dose from thoron progeny, can usually be ignored in typical indoor environments, the assessment of dose from thoron inhalation can be determined from measurements of the size distribution of the attached thoron progeny and the thoron progeny concentrations.

Doses from the inhalation of thoron were assessed from thoron progeny size distribution and concentration measurements made in three typical rural indoor environments and one kind of urban indoor environment for comparison. The dose conversion factors and annual effective doses were calculated using dosimetric methods.

2. MATERIALS AND METHODS

2.1. Measuring devices

Two portable integrating devices were used for making the field measurements for dose assessment. One was a portable ‘progeny integrating sampling unit’ for measuring the equilibrium equivalent concentration (EEC) of ^{222}Rn and ^{220}Rn progeny [7]. The other was a newly developed device for

measuring the ^{212}Pb particle size distribution [8]. The two devices are illustrated in Fig. 1.

The device for measuring the EECs of ^{222}Rn and ^{220}Rn progeny is a cylinder made of stainless steel with four air inlets (3 mm diameter) on its wall and one outlet at the top. Four discs of CR-39¹ are set at the sites and ^{222}Rn and ^{220}Rn progeny are collected on the surface of a 0.8 μm pore membrane filter². The thoron progeny are distinguished from the radon progeny by discriminating between the alpha particles emitted by the ^{222}Rn and ^{220}Rn progeny using Al Mylar absorbers (mainly the 8.78 MeV alpha particle from ^{212}Po) and ignoring the unattached ^{216}Po in the air. The flow rate is 0.8 L/min, the standard sampling period is 24 h and the lower limits on detection for the EECs of ^{222}Rn and ^{220}Rn are 0.57 Bq/m³ and 0.07 Bq/m³, respectively. Because it is not yet possible to calibrate ^{222}Rn and ^{220}Rn progeny concentrations in different environments, this device has been used in conjunction with two sets of working level monitors, WLM-plus 200³ and WLx⁴, which can determine the individual concentrations of ^{222}Rn or ^{220}Rn progeny and had been compared in the Environmental Measurements Laboratory of the United States Department of Energy.

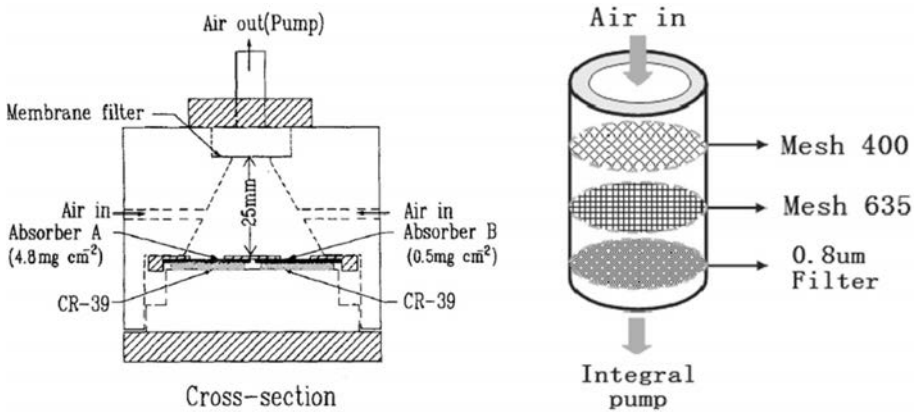


FIG. 1. Devices for measuring the concentration and size distribution of thoron progeny.

- 1 Fukuvi Chemical Industry Co. Ltd, Japan.
- 2 Advantec MFS, Inc., United States of America.
- 3 Tracerlab Instruments, Germany.
- 4 Pylon Electronics, Inc., Canada.

The device for measuring the ^{212}Pb particle size distribution consists of three holders with a diameter of 2 cm, containing, respectively, a 400 mesh screen, a 635 mesh screen and a back-up filter. With a flow rate of 3 L/min, aerosol particles with different sizes are collected separately on different screens by Brownian diffusion with different penetration rates. After waiting for 6 h after sampling (the ^{212}Bi and radon progeny are then almost completely decayed), the alpha particles emitted from the progeny of ^{212}Pb on the different screens and the filter are recorded separately by three CR-39 discs. In order to reconstruct the aerosol particle size distributions, the Monte Carlo method referred to as the ThB Size Distribution Calculation was used to calculate the particle size collection efficiencies. The uncertainties in the activity median aerodynamic diameter (AMAD) and geometric standard deviation (GSD) are designed to be 0.005 μm and 0.1 μm , respectively. Ignoring the different ^{212}Pb and ^{212}Bi size distributions and unattached ^{216}Po , the measured values can be used to determine the size distribution of the thoron progeny. This device was compared with the results of a screen diffusion battery with the relationship between the count median diameter and AMAD.

2.2. Dose evaluation

The LUDEP 2.07 computer code was used for thoron dose evaluation. LUDEP (LUng Dose Evaluation Program) is a computer program for personal computers developed by a Task Group of Committee 2 of the International Commission on Radiation Protection (ICRP) for developing the model of the human respiratory tract described in ICRP Publication 66 [9]. LUDEP enables the user to calculate doses and dose rates from intakes of radionuclides to regions of the respiratory tract and to other body organs for a wide range of user defined conditions. LUDEP 2.07 includes databases containing the ICRP Publication 30 biokinetic models and special treatment of radioactive decay chains, which is also able to calculate bioassay functions [10, 11]. The deposition fractions and dose conversion factors of user defined radionuclide size distributions can be calculated in LUDEP 2.07. The use of LUDEP 2.07 for thoron dose evaluation has been reported in Ref. [12].

In the calculation of dose, the unattached fraction of thoron progeny was ignored, as there is almost no unattached ^{212}Pb and ^{212}Bi in normal indoor environments. Therefore, it was assumed that all thoron progeny contributing to the dose conversion factors were attached. While the half-life of ^{212}Bi (1.01 h) is much shorter than that of ^{212}Pb (10.6 h), the assumption that ^{212}Bi has the same particle size distribution as that of ^{212}Pb is reasonable. A ratio of ^{212}Pb to ^{212}Bi of

1 to 0.25 is used in this calculation [13]. The half-time for absorption of ^{212}Pb and ^{212}Bi into the bloodstream was taken to be 10 h [14].

3. RESULTS AND DISCUSSION

3.1. Measurement results for three typical rural indoor environments

For the three typical types of rural home (brick houses, mud houses and caves), field measurements were carried out over the past year in the Pinggu District of Beijing (latitude 40.35°N , longitude 117.17°E), Yangjiang in Guangdong Province (latitude 21.83°N , longitude 111.54°E) and Datong in Shanxi Province (latitude 39.81°N , longitude 113.56°E). Yangjiang is an area of high natural background radiation where the ^{232}Th concentration in soil is about 16 times the national average value, and where a high thoron concentration has been found [4]. The bedrooms were chosen for measurement and the windows were kept half closed and the doors closed, as was usual. Additional aerosol sources were avoided to keep the environment stable. For comparison, two brick houses in an urban indoor environment were chosen — the sampling time for these houses was 48 h instead of the normal 24 h in order to achieve a reasonable lower limit on detection. The results are shown in Table 1.

TABLE 1. RESULTS OF INDOOR THORON PROGENY MEASUREMENTS

	Type of construction	No. of samples	Size distribution		EEC (Bq/m^3)
			AMAD (μm)	GSD	
Pinggu	Brick	7	0.11 (0.09–0.13)	2.5 (2.3–2.7)	0.95 (0.27–1.35)
Yangjiang	Mud	8	0.0638 (0.03–0.13)	2.7 (1.9–3.3)	25.25 (14.03–53.32)
Datong	Cave	5	0.05 (0.04–0.06)	3.1 (3.1–3.6)	2.33 (1.63–4.41)
Urban, for comparison	Brick	2	0.155 (0.15–0.16)	2.0 (1.7–2.2)	0.14 (0.06–0.22)

Note: AMAD — activity median aerodynamic diameter; EEC — equilibrium equivalent concentration; GSD — geometric standard deviation.

For both the size distribution and the concentration, there are quite large differences between the three rural homes and the urban homes. The size distributions in the rural homes reflect significantly smaller particle sizes than those for the urban home, while the concentrations are significantly higher. The results for the size distribution are inconsistent with the results of previous surveys [8], possibly because of the good ventilation conditions in the present study. The thoron concentrations are higher than found previously, mainly because, in the present study, the background levels were high and the uncoated walls allowed more thoron exhalation.

The size distributions and concentrations in the rural homes are significantly different from those in the urban homes, although the differences are less when comparing only brick homes. The thoron progeny concentrations in brick homes, regardless of whether rural or urban, are significantly lower than those in mud homes or caves, while the sizes of the thoron progeny particles in brick homes are distinctly larger. This could be due to differences in ventilation and thoron exhalation rates. The average thoron progeny concentration in the mud homes is unusually high — 25.25 Bq/m^3 compared with a global average value of 0.5 Bq/m^3 (see annex E of Ref. [15]).

3.2. Dose assessment

The following inputs were chosen for the LUDEP 2.07 calculations:

- (a) A shape factor of 1.1;
- (b) A particle density of 1.4 g/cm^3 ;
- (c) A breathing rate of $0.78 \text{ m}^3/\text{h}$;
- (d) An occupancy factor of 0.8;
- (e) The ^{212}Pb and ^{212}Bi biological dynamics models Pb (D).MOD and Bi (D).MOD, respectively;
- (f) A weight factor of 0.025 for the extrathoracic region.

The calculated dose conversion factors and annual average doses are given in Table 2.

TABLE 2. RESULTS OF THE DOSE ASSESSMENT

	Dose conversion factor (nSv per Bq m ⁻³ h ⁻¹)					Annual average effective dose (mSv)
	Extrathoracic region	Bronchial region	Bronchiolar region	Alveolar interstitial	Total	
Pinggu	3.24	57.53	95.11	3.52	159.40	0.26
Yangjiang	4.51	86.05	140.50	4.67	235.73	10.12
Datong	23.12	315.83	286.95	3.25	629.14	2.49
Urban environment	2.39	41.36	67.10	2.58	113.44	0.03

Comparing the dose conversion factors for the different environments, it is evident that the dose conversion factor for the cave houses is the largest because of the very small aerosol size. For all three rural environments, the dose conversion factors are higher than for the urban environment, especially when considering mud homes and caves. However, the highest dose (for mud homes in Yangjiang) is largely due to the exceptionally high thoron concentration. The annual dose for the urban homes is only 0.03 mSv, significantly lower than the doses for the rural environment because of both the small dose conversion factor and the low thoron progeny concentration. The dose conversion factor for brick houses in the urban environment is quite close to the result published in Ref. [16]. The results of this study suggest that the dose conversion factors traditionally used for thoron progeny are applicable only to the urban indoor environment. The bronchial and bronchiolar regions of the lung contribute most to the total dose conversion factor, and the contributions from the extrathoracic region and alveolar interstitial could, on a comparative basis, be ignored.

4. CONCLUSION

Much research has been carried out to determine the relationship between thoron progeny and environmental parameters and between the dose conversion factor and the thoron progeny particle size. However, relatively few field measurements have been carried out, especially in rural areas where, in countries such as China, most people live.

The work described in this paper evaluated thoron exposures in three typical types of rural home in China: brick houses, mud houses and caves. For comparison, measurements were also made in urban homes. Thoron progeny size distributions and thoron progeny EECs were determined. From these measurements, doses were assessed. The results show that rural indoor environments are associated with much larger dose conversion factors than those for the urban indoor environment, due to the smaller thoron progeny particle sizes. The smallest particle sizes were found in caves which, as a result, were associated with the largest dose conversion factor. The annual average effective dose due to thoron exposure is influenced by the dose conversion factor as well as the thoron EEC. The average annual dose assessed for mud homes in Yangjiang was 10.12 mSv, which suggests that further attention might be needed.

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BRAZILIAN NORM INDUSTRIES: LESSONS AND CHALLENGES

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Abstract

Several industries operating in Brazil are dealing with NORM, especially those related to the mining and beneficiation of tin, niobium and phosphate, and to oil exploration. The Brazilian National Nuclear Energy Commission has been investigating the larger industries in order to assess the extent of exposure of workers and members of the public from NORM. The paper presents the characteristics of Brazil's larger NORM industries, as well as the methodologies used to evaluate the radiological impact associated with their operation. The results of radionuclide analyses of environmental samples collected on site at these facilities demonstrate the importance of developing guidelines, especially for soil remediation. Finally, the Brazilian regulations, the main advances, and the challenges facing NORM industries are briefly discussed.

1. INTRODUCTION

Exposure to natural sources is responsible for about 90% of the total dose received by individuals every year [1]. Although activity concentrations vary considerably with the type of environment and the location, exposure to natural sources in the environment is, with the exception of radon, not normally amenable to control. Natural resources extracted from the ground such as coal, other mineral ores, oil and natural gas contain varying concentrations of radionuclides of natural origin. When these resources are extracted and processed, their natural state can be modified, which may result in the enhancement of the original radioactivity content of the material. Such enhancements may be observed in the residues created by the process and in the products and by-products and can sometimes be high enough to pose a risk to both humans and the environment if they are not controlled properly. Materials of this kind are referred to as NORM. Long lived radioactive elements such as uranium, thorium and potassium and any of their decay products such as radium and radon are contained in NORM. These elements have always been present in the Earth's crust and atmosphere, and are found naturally concentrated in some minerals. NORM can be distinguished from artificial sources such as those produced by nuclear power and those

used in nuclear medicine (where the radioactive properties of the material may be what make it useful) but, from the perspective of radiation doses received by individuals, such a distinction is arbitrary in that the same international standards of radiation protection apply. Public exposure from NORM industries is mainly a result of processing, recycling, storage and disposal of residues. In contrast with nuclear industries, the presence of radioactivity in NORM industries is often incidental to the use to which radioactive material is being put. Another consideration is that many NORM residues can be recycled or used as by-products (e.g. phosphogypsum and coal ash) unlike the residues generated by nuclear industries, which have to be disposed of as waste. Scenarios involving significant exposure of members of the public can easily be envisaged, leading to exemption levels being exceeded [2]. The main issues associated with NORM industries are, according to Ref. [3]:

- (a) Effects of processing: Processing of NORM leads to the production of products (commodities) and waste. In many cases, the original radionuclide composition or concentration is changed in the products and/or waste as a result of processing. This can complicate the process of assessing the effects of NORM.
- (b) General public and industry awareness of NORM: Many industries, particularly the mining industries other than uranium and mineral sand mining, may have operated until recently without realizing that their operations could give rise to NORM in their products and/or waste.
- (c) Application of radiation protection requirements: In the nuclear power industry, the potential consequences of a serious accident can be catastrophic in the short term and highly deleterious to human health and the environment in the long term. Therefore, stringent design and operational controls are applied, not only to mitigate radiological impacts in the event of an accident, but to reduce the probability of an accident. In NORM industries, there is no potential for a nuclear or radiological emergency. Hence, any proposed control measures in NORM industries need to be based only on controlling radiation exposure.

In Brazil, the National Nuclear Energy Commission (CNEN, Comissão Nacional de Energia Nuclear) is the regulatory authority in the nuclear field. The CNEN is in charge of the licensing and control of nuclear installations in the country. CNEN issues standards and carries out inspections to assess compliance with legal requirements. At present, NORM industries are not regulated by the CNEN. However, it is recognized that no other governmental institution in the country is prepared to assess the radiological consequences of the operation of such industries [4]. As a result, the CNEN has been assessing and preparing

reports regarding the consequences of the operation of NORM industries (mining and mineral processing industries) since the 1980s in response to requests from other state and federal agencies. This paper presents the characteristics of Brazilian NORM industries, as well as the methodologies used to evaluate the radiological impacts associated with their operation.

2. BRAZILIAN NORM INDUSTRIES

In the extensive territory of Brazil (8.5 million km²), the mineral deposits associated with NORM industries can be divided into the following groups [5]:

- (a) Pegmatite and carbonatite niobium–tantalum deposits (columbite–tantalite);
- (b) Deposits of tin, niobium–tantalum and zircon in granitic intrusions;
- (c) Deposits of mineral sand containing zircon, titanium and thorium (monazite);
- (d) Phosphate deposits;
- (e) Cupriferous deposits of the Carajás mineral province.

According to Ref. [5], there are at least four known Cretaceous carbonatite complexes that have niobium deposits in Brazil. Two of these deposits are being mined and are responsible for 70% of world niobium production. The minerals belong to the pyrochlore group and have been exposed to intense weathering, which is typically the case in facilities using open pit mines. One of the deposits located in the state of Minas Gerais has reserves of around 460 million t with an average yield of 2.5–3.0% niobium pentoxide (Nb₂O₅); the deposit contains up to 0.008% triuranium octaoxide (U₃O₈) and 0.135% thorium oxide (ThO₂), which become concentrated throughout the mineral and metallurgical processing of niobium [6]. In another mine located in the state of Goiás, the reserve is 10.2 million t with an average yield of 0.9% Nb₂O₅. The pyrochlore ore contains up to 0.03% U₃O₈ and 0.05% ThO₂. Some areas of these carbonatites show among the highest levels of natural radioactivity in Brazil.

The main sources of columbite and tantalite ore are in the pegmatite provinces in the states of Bahia and Minas Gerais and in the north-east of the country (in the states of Rio Grande do Norte and Paraíba). This ore is extracted by prospectors and is exported after being concentrated. Operations are intermittent, depending on the international price of tantalum and weather conditions that are unfavourable for farming (in the dry season). The concentrated ore may contain up to 1% U₃O₈ and 1.8% ThO₂, but normally the values are much lower. Small volumes are also produced in the region of São João Del Rey in Minas Gerais,

where tantalum is found with microlite containing uranium and is processed by hydrofluoric leaching.

Deposits of tin, niobium–tantalum and zircon, related to two granitic intrusions of the middle Proterozoic period (around 1 700 million years old), are located in the state of Amazonas in the northern part of the Amazon craton. The majority of the associated uranium and thorium is contained within the crystalline structure of these minerals, with the exception of cassiterite. The ore has average levels of 0.0176% tin, 0.808% zirconium oxide, 0.223% Nb₂O₅ and 0.29% tantalum pentoxide and contains uranium and thorium reserves of around 10 000 t [6] at 0.03% uranium and 0.135% thorium.

In the coastal regions of north and north-east Brazil, from the state of Rio de Janeiro to the Amazon valley, there are large Tertiary sedimentary deposits parallel to the coast grouped under the name ‘the Barreiras Formation’. These sedimentary deposits are made up of sand, red clay and ferriferous concretions that came from erosion of gneiss and granite rock from the Brazilian shield. They contain minerals such as zircon, ilmenite, rutile and monazite. The zircon has average levels of 0.03% U₃O₈ and 0.016% ThO₂.

The main source of raw materials for phosphate fertilizer production is apatite ore. Phosphate rock, comprising mainly apatite, is treated with sulphuric acid and water (the ‘wet process’) and converted into phosphoric acid, giving rise to a calcium sulphate residue called phosphogypsum (CaSO₄ nH₂O) [7]. Some of the phosphate deposits are in the same carbonatite structures as the pyrochlore (niobium) deposits mentioned above. This phosphate ore contains up to 14% phosphorus pentoxide with average uranium and thorium contents of up to 0.0185% U₃O₈ and 0.0480% ThO₂. The ²²⁶Ra concentrations are up to about 3 Bq/g [6]. The most important producers of phosphate fertilizer are located in Rio Grande, RS; Cubatão and Cajati, SP; and Uberaba, MG.

The mineral province of Carajás, located in the state of Para, in northern Brazil, is one of the largest iron ore reserves in the world. It also has important deposits of manganese, nickel, copper and gold. One of the interesting aspects of this mineral province is the occurrence of several stratigraphically and tectonically related copper–gold deposits known as the copper–gold belt. The deposits are made up of breccias with sharp dips, embedded in Archeozoic volcanic sedimentary rocks with a low metamorphic grade. These polytomic breccias are classified into three types by hydrothermal matrix: chlorite, iron and magnetite. The uranium has a very irregular distribution and is apparently associated with the magnetite breccia. The ore mineral is a chalcopyrite. Some of its anomalous concentrations are: copper, 0.5–11%; gold, 0.5–15 ppm; and uranium, 28–380 ppm. In the Salobo area, the U₃O₈ concentrations can reach 500 ppm [8].

In addition to these deposits, the occurrence of radionuclides of natural origin in the extraction of oil and natural gas must be addressed. When oil is extracted from beneath the soil, solids and water are produced with it. Under certain conditions, barium and calcium salts (sulphates and carbonates) containing ^{226}Ra and ^{228}Ra are precipitated. Over time, these precipitates form thick layers of scale inside the production tubulars and other equipment and must be removed and disposed of safely [9].

Table 1 shows the radionuclide activity concentrations for some typical materials associated with mining and oil extraction in Brazil. The Table shows that in monazite, niobium–tantalum slag and pyrochlore slag, the activity concentrations of the ^{232}Th series radionuclides are dominant, while for zircon and oil extraction scale, the ^{238}U series radionuclides are dominant.

TABLE 1. TYPICAL NORM ACTIVITY CONCENTRATIONS

	Activity concentration (Bq/g)	
	U-238 series radionuclides	Th-232 series radionuclides
Monazite	6–40	6–300
Ta–Nb slag	800–1200	1800–2200
Pyrochlore slag	17–140	190–800
Zircon	8.0–500	1.2–6.0
Phosphogypsum	7–728 (Ra-226)	—
Oil extraction scale	0.1–4000 (Ra-226, Pb-210)	—

Source: See Ref. [5].

The main radiological issues associated with NORM industries in Brazil are:

- (a) Generation of NORM residues, especially during chemical or pyrometallurgical processing, leading to an increase in radionuclide concentrations of two orders of magnitude above the original concentration in the ore;
- (b) Inadequate criteria for the control of NORM waste disposal;

- (c) Acid mine (rock) drainage, mainly in coal and gold mines;
- (d) Internal exposure of workers via airborne dust inhalation;
- (e) External exposure to gamma radiation from pyrochlore slag.

The regulatory bodies do not clearly define the criteria for allowing the disposal of uranium and thorium containing tailings in industrial landfill facilities designed for hazardous waste. As a result, in the absence of a clearly designated disposal approach, the material remains piled on the ground for indefinite periods.

3. STANDARDS AND REGULATIONS

All Brazilian non-uranium NORM industries are obliged to follow the CNEN Standard NE-4.01 [10] (facilities for the mining and processing of uranium ore are excluded since they are considered to be nuclear installations). According to this standard, the annual flow of activity generated by each operational step (in becquerels per year), including the annual activity of the waste, must be estimated based on the activity concentrations of samples from the various process steps and the masses of material involved over the course of a year. On the basis of this information, facilities are divided into three categories:

- Category I: Facilities with NORM activity concentrations exceeding 500 Bq/g or with doses to members of the public or workers exceeding 1 mSv/a at any point of the operational process.
- Category II: Facilities with NORM activity concentrations of 10–500 Bq/g.
- Category III: Facilities with NORM activity concentrations below 10 Bq/g and with doses below 1 mSv/a.

According to the category in which the facility is classified, it may have to comply with different levels of requirements that may even approach those required in the licensing of uranium production facilities. The idea is to exempt those facilities falling into category III. However, every facility submits the following preliminary information to the CNEN:

- (a) A simplified operational flow chart;
- (b) Levels of the relevant radionuclides in the ore, residues, wastes and effluents;
- (c) A mass balance;
- (d) The solubilities of radionuclides in the waste;

- (e) A description of the waste management strategies (e.g. dams, impoundments and piles);
- (f) A simplified description of the surrounding environment.

It must be pointed out that the CNEN has no legal authority to control these industries. Because of this, the application of this standard is not compulsory. On the other hand, the CNEN is responsible for the radiological procedures of these industries. Consequently, depending on the situation, the CNEN contacts the Environmental Protection Agency, which obligates the industries to comply with the requirements contained in the standard to control mining activities in the country with respect to radiological issues [11].

Because Brazil is so large and has such a large number of NORM facilities, the CNEN has mainly inspected facilities classified as categories I and II according to CNEN Standard 4.01. During each visit, not only are the features related to radiation protection in the workplace evaluated, but also environmental issues in order to ensure that emissions and waste disposal at the facility do not expose the public to radiation levels above those established by law. For this purpose, an environmental radiation monitoring programme was established that took into consideration the specific impacts of the production process on the neighbouring environment for each installation. Samples are taken of the soil, surface water, sediment, aerosols, groundwater and plants. All samples are analysed to determine the concentrations of ^{238}U , ^{232}Th , ^{226}Ra , ^{228}Ra and ^{210}Pb .

The exemption level for regulatory control established in CNEN Standard NE-6.02 [12] is in general 100 Bq/g, or 500 Bq/g for natural solid substances. Moreover, CNEN Standard NE-6.05 [13] states that waste can be disposed of directly into a landfill if the radionuclide activity concentration is below 74 Bq/g. It must be pointed out that this value was established for small amounts of material from medical, industrial, and research uses. It is not intended to apply to large volumes such as those generated in the mining and mineral processing industry. If the concentrations of NORM waste are below the values mentioned above, its final disposal can only be exempted from regulatory control by the CNEN if it is certain that it will not give rise to a dose to a member of the public that exceeds the criterion of 1 mSv/a established by CNEN-NE-3.01 [14]. To demonstrate this, a radiological risk assessment must be carried out for different scenarios of exposure.

The use of phosphogypsum in agriculture and as a building material has been studied [7, 15, 16]. These studies contributed to the development of a specific standard for the use of phosphogypsum in agriculture and the cement industry. A reference level of 1 Bq/g for the activity concentrations of ^{226}Ra and ^{228}Ra was chosen. A regulatory standard is currently being developed for the use of phosphogypsum in construction.

4. CONTAMINATED SITES

Stockpiling of solid radioactive waste in the open air should take into account the soil, water and air as potentially significant pathways of human exposure. Clearly, the radiological impacts associated with NORM industries do not only come directly from uranium and thorium in the ore. They can also be the result of mineral processing, the occurrence of acid mine drainage, and other important geochemical processes that may concentrate radionuclides in specific matrices. Relevant pathways of exposure to radiation originating from waste could also be influenced by the type of disposal, the local environment, the land use and the demographics of the people in the region [11].

In January 1996, the Companhia Ambiental do Estado de São Paulo (CETESB) embarked on a research and development project in order to establish quality reference values and intervention values for soil and groundwater in the state of São Paulo. The project only took organic contaminants, volatile organic material and metals into consideration and did not deal with radiological issues. In December 2005, a new list was published, made up of 84 substances and defining three reference values for soil and groundwater [17]. The three values are:

- (1) Quality reference value: This is the concentration of a specific substance in the soil or groundwater, which defines a soil as clean or the natural quality of groundwater.
- (2) Prevention value: This is the concentration of a specific substance above which prejudicial changes to soil or groundwater can take place. This value indicates the quality of a soil to be able to carry out its main functions, protecting ecological receptors and groundwater quality. Its determination for soil was based on trials with ecological receptors.
- (3) Intervention value: This is the concentration of a specific substance in the soil or groundwater above which there are potential direct or indirect risks to human health, considering a generic exposure scenario. For the soil, they were established for agricultural, residential and industrial maximum protection area exposure scenarios.

An area being studied is classified as a contaminated area under investigation when there are signs pointing to the presence of soil and groundwater contaminants in concentrations above the intervention levels. This shows that action is needed to recover the risk receptors. According to Regulatory Position 3.01/007 [18], generic intervention levels are reference levels established by CNEN a priori that are to be considered in the planning phase and used in the justification and optimization process for intervention situations. An existing annual dose of 10 mSv is to be used as a generic reference

value for an intervention action in situations in which there is chronic exposure of the public. Estimate or measurement of an existing reference dose value over 10 mSv/year always needs to have an evaluation to determine whether protection or remediation measures need to be implemented. The CNEN does not establish an upper intervention level above which protection or remediation measures are mandatory, but always considers intervention justified when the existing dose is above 50 mSv/year.

Therefore, prevention intervention values established by the CNEN are based on effective dose and not risk to human health. Nevertheless, it is always much easier for regulatory bodies to have levels to compare with instead of working with dose values that require environmental assessments and calculations to be carried out. The levels may be given in terms of activity concentration of the radioactive materials involved in a particular activity and can be either generic or specified for particular types of activity.

In order to facilitate the decision making process, establish priorities and reduce costs, radionuclide orientation values for soil quality have been applied which aid in the initial step of the investigation and remediation of areas contaminated by NORM. In this case, values estimated from dose calculations from different scenarios proposed in Ref. [19] have been used. The intervention value is the concentration of a given radionuclide in the soil that will give rise to a dose of 50 mSv/a to a member of the public. This study also defined the prevention value as the concentration of a given radionuclide in the soil that will give rise to a dose of 10 mSv/a to a member of the public. Intervention and prevention levels are shown in Table 2.

This practice has shown that many of the areas investigated have contamination levels above those established for industrial areas, mainly around waste stockpile areas. Clearly, the existence of a contaminated area can generate problems such as negative health impacts, compromising the quality of water resources, restricting soil usage, and damaging public and private property, in addition to environmental damage. The presence of contamination in certain areas could hinder the process of obtaining new licences or renewing existing licences if the situation is not adequately taken up with the environmental agency.

5. CONCLUSIONS

The lack of specific regulations for NORM waste management often leads to situations that result in uranium and thorium contamination of areas without the population, the government, or even the company knowing about it. Clearly, there is a need to move towards a more harmonized approach in order to minimize problems like this that appear to be occurring on an ever increasing basis.

TABLE 2. INTERVENTION VALUES AND PREVENTION VALUES FOR SOIL CONTAMINATION

	Activity concentration (Bq/g)					
	Intervention value (dose = 50 mSv/a)			Prevention value (dose = 10 mSv/a)		
	Agriculture	Residential	Industrial	Agriculture	Residential	Industrial
U-238	130	360	470	26	72	94
Th-232	190	190	65	38	38	13
Ra-226	4.5	9	60	0.9	1.8	12
Ra-228	0.75	1.7	31	0.15	0.34	6.2
Pb-210	8.5	19	170	1.7	3.8	34

Source: See Ref. [19].

Obviously these facilities must not only take technical aspects into account, but social and environmental considerations as well. Many industrial sites in Brazil are small mining companies which are responsible for generating much of a region's jobs. The implementation of environmental education programmes, seminars and courses mainly for technicians of the environmental agencies and the public has proven to be an effective way to improve awareness and, ultimately, to minimize the radiological impacts associated with industrial activities involving NORM. There would be an obvious benefit in encouraging these small NORM operations to use the best available techniques for reducing the amounts and concentrations of NORM waste.

It is also important to adopt radionuclide reference values for soil quality based on the concept of intervention values and prevention values [17]. The use of these values, taking account of the various current and future exposure scenarios facilitates decision making regarding the remediation approach to be adopted for a contaminated area. An important advantage of this strategy is the avoidance of the need for future remedial action in the simplest situations in which there are no adverse human health effects that justify further investigation or remedial action.

Whenever possible, the use of NORM residues is to be encouraged. An example is the use of phosphogypsum in agricultural and construction applications. The benefits of this approach include the preservation of natural

resources, the reduction of air and water pollution, the reduction of the amount of NORM waste requiring disposal and creation of jobs.

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ASSESSMENT OF URANIUM IN DRINKING WATER IN KITWE, CHAMBESHI AND CHINGOLA IN THE COPPERBELT REGION OF ZAMBIA

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Abstract

The paper discusses uranium in drinking water in three towns located in the uraniumiferous Copperbelt region of Zambia. The mining towns of Kitwe, Chambeshi and Chingola in the Copperbelt region have two main sources of drinking water: underground mine water and surface water from the Kafue River system. Chambeshi abstracts its water from underground while Chingola abstracts water from both underground and surface water sources. Kitwe abstracts water from the Kafue River which drains the entire Copperbelt region. In Zambia, World Health Organization (WHO) guidelines for drinking water are followed, although routine monitoring for radioactivity is not mandatory. The overall objective of this study was to gather baseline data on uranium contamination in drinking water and compare with the WHO guidelines and also to establish the source with the highest levels of uranium. An alpha spectrometer was used to analyse the samples and the results from this survey indicate that the average total uranium activity concentrations were in the range of 78.50 mBq/L in Kitwe to 600.78 mBq/L in Chambeshi. The committed effective doses for adults were in the range of 3.58 μ Sv/a in Kitwe to 20.28 μ Sv/a in Chambeshi. From both water sources, the committed effective doses were below the WHO guideline reference dose level of 100 μ Sv/a. The uranium activity concentration for Chambeshi was above the recommended screening level for alpha emitters in drinking water of 500 mBq/L. The average ^{235}U : ^{238}U ratio was 0.046, the natural abundance ratio for uranium.

1. INTRODUCTION

Long lived uranium and radium isotopes are responsible for natural radioactivity in both groundwaters and surface waters [1]. During the decay process, alpha and beta radiation is emitted. Natural uranium consists of ^{234}U , ^{235}U and ^{238}U , each with its own particular mass abundance and half-life [1]:

- Uranium-234: 0.005 55%, $T_{1/2} = 2.44 \times 10^5$ a;
- Uranium-235: 0.72%, $T_{1/2} = 7.04 \times 10^8$ a;
- Uranium-238: 99.28%, $T_{1/2} = 4.47 \times 10^9$ a.

Uranium is widespread throughout the environment and most of the uranium in water originates from uranium leached from rocks and soils [2]. Uranium ingestion (water and food) and inhalation can lead to cancer and kidney damage [2].

The industrial activities of copper and cobalt mining in the Copperbelt region started in the 1930s, and the geological framework of this region is part of the Katanga supergroup containing uranium mineralization [3]. Besides heavy metals, radionuclides are also present in mine wastewater discharged to surface water courses from mining activities [4]. Therefore, concern about radioactivity in the drinking water has led to an increased demand for data. Drinking water quality studies for heavy metals and their health effects in the Copperbelt region due to mining activities have been performed many times [5], but, as far as is known, none has focused on the assessment of radiological risks due to drinking water contamination by uranium from mining activities.

The wastewaters from mining and metallurgical activities discharged to mine residue deposits end up in the Kafue River without proper treatment to remove radionuclides [6]. The Kafue River drains the entire Copperbelt region (see Fig. 1) and, supplemented by underground mine water, is the chief source of drinking water for the residents of the region.

The mining towns of Kitwe, Chambeshi and Chingola are located in the region where radon gas measurements in the underground mines showed that the radon levels were over 1000 Bq/m^3 [7]. These towns have two sources of drinking water: underground mine water and the Kafue River. In order to better evaluate the radiological risks affecting the residents of this region, a deeper knowledge of the uranium contamination in the water is required, aimed at strengthening consumers' security concerning drinking water quality according to World Health Organization (WHO) guidelines (see chapter 9 of Ref. [8]), which have been adopted by Zambia. The WHO reference dose level for adults is $100 \mu\text{Sv}$ from one year's drinking water consumption (730 L) and for practical purposes, the recommended screening levels for drinking water below which no

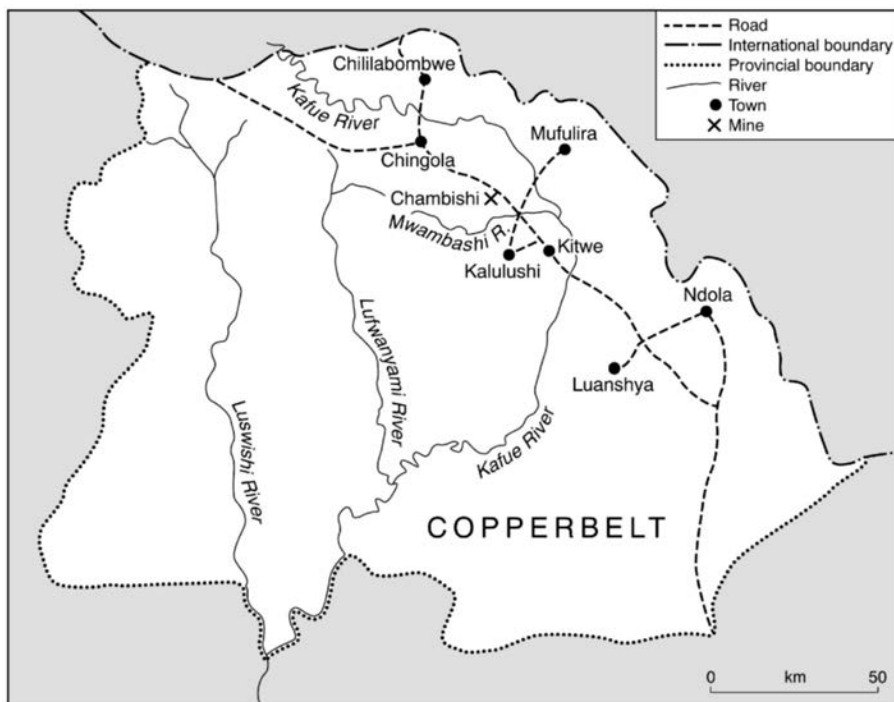


FIG. 1. The copperbelt region.

further action is required are 500 mBq/L for gross alpha activity and 1000 mBq/L for gross beta activity. The WHO recommendations do not differentiate between radionuclides of natural and artificial origin. The uranium isotopes ^{234}U , ^{235}U and ^{238}U are predominantly alpha emitters and the recommended screening level is therefore 500 mBq/L. The aim of this work was to gather baseline data related to the uranium contamination of drinking water and compare with the WHO screening levels and also to establish the source with the highest concentrations of uranium.

2. METHODS AND INSTRUMENTATION

Samples of tap water were collected on the same day from the outlets of water treatment plants in Kitwe, Chambeshi and Chingola. Six samples were collected at each sampling location at monthly intervals between August 2009 and October 2010. The samples were preserved with 50% nitric acid to a pH of 2 and transported to Lusaka in coolboxes. They were stored in a refrigerator

at 4°C before being shipped to the South African Nuclear Energy Corporation for analysis. The analysis involved filtering of 2.5 L of sample and evaporating to near dryness. The residue was dissolved in a small amount of mineral acid. The solution was loaded into a liquid extraction chromatography column where uranium was selectively absorbed. The column was then rinsed with acid to remove interfering elements and uranium was eluted and co-precipitated with lanthanide fluoride and collected on a special filter. Uranium-232 was added as tracer for chemical recovery determination. The samples were measured using a Canberra Alpha Analyst Spectrometer for 24 h to determine the alpha activity spectrum.

3. RESULTS

The activity concentrations of ^{234}U , ^{235}U and ^{238}U in the drinking water samples are shown in Tables 1–6 and the summed activity concentrations for all three uranium isotopes are shown in Table 7. The ratio of ^{235}U concentration to the ^{238}U concentration was found to be 0.046 (the natural abundance ratio) in every sample. The uncertainty for each analysis was reported as one standard deviation (or a coverage factor, K , of 1). The uncertainty was calculated mainly from the counting statistics. It is not the standard deviation obtained from replicate measurements but the standard deviation associated with the tracer activity, the addition of yield tracer to the sample and the counting statistics of the sample and blank.

The committed effective doses for adults via the consumption of drinking water supplied by the two utility companies were assessed from the activity concentration data. The effective dose is a product of the dose coefficient, the activity concentration and the amount of water consumed annually [9]. The dose coefficients for ^{234}U , ^{235}U and ^{238}U recommended by the International Commission on Radiological Protection — 4.9×10^{-8} , 4.7×10^{-8} and 4.5×10^{-8} Sv/Bq, respectively — were applied [9]. The consumption of water was assumed to be 730 L/a, as given in the WHO guidelines (see chapter 9 of Ref. [8], and Ref. [10]). Based on the sum of the mean isotopic activity concentrations for each sampling site for the six month sampling period (see Tables 1–6), the annual committed effective doses to the adult population for each sampling site were assessed and are shown in Table 8. The summed activity concentrations and annual effective doses are also presented graphically in Figs 2 and 3, respectively.

TABLE 1. URANIUM IN DRINKING WATER FROM UNDERGROUND, CHINGOLA

Sampling date	Activity concentration (mBq/L)							
	U-234		U-235		U-238		U-234/U-238	
	Value	LLD	Value	LLD	Value	LLD	Total	
2009-08-11	60.1 ± 6.9	7.7	2.73 ± 0.30	0.24	59.3 ± 6.6	5.3	122.13	1.01
2009-09-15	60.9 ± 5.2	6.7	2.21 ± 0.13	0.13	48.1 ± 4.3	2.8	91.21	1.27
2009-10-13	55.7 ± 6.0	4.5	1.65 ± 0.22	0.21	35.8 ± 4.8	4.5	93.15	1.56
2009-11-10	96.0 ± 10.2	2.8	3.52 ± 0.41	0.13	76.5 ± 8.9	2.8	176.02	1.26
2010-02-04	42.5 ± 3.9	0.95	1.66 ± 0.16	0.04	36.0 ± 3.6	0.95	80.46	1.19
2010-10-28	39.5 ± 4.0	1.1	1.60 ± 0.17	0.05	34.9 ± 3.7	1.1	76.00	1.13
Mean	59.03 ± 6.0	3.96	2.23 ± 0.23	0.13	48.43 ± 5.32	2.90	106.50	1.24

Note: LLD — lower limit on detection.

TABLE 2. URANIUM IN DRINKING WATER FROM UNDERGROUND, CHAMBESHI

Sampling date	Activity concentration (mBq/L)								Total
	U-234		U-235		U-238		U-234/U-238		
	Value	LLD	Value	LLD	Value	LLD	Value	LLD	
2009-08-11	412 ± 19	9.4	12.2 ± 0.7	0.30	266 ± 15	6.4	690.20	1.55	
2009-09-15	582 ± 28	17	15.7 ± 0.9	0.15	340 ± 21	3.4	937.70	1.71	
2009-10-13	343 ± 13	3.3	9.51 ± 0.45	0.15	207 ± 10	3.3	559.51	1.66	
2009-11-10	452 ± 16	5.3	13.0 ± 0.6	0.24	282 ± 13	5.3	747.00	1.60	
2010-02-04	337 ± 12	1.1	9.53 ± 0.41	0.05	207 ± 9	1.1	553.53	1.63	
2010-10-28	56.6 ± 5.1	1.2	2.65 ± 0.23	0.06	57.5 ± 5.1	1.2	116.75	0.98	
Mean	363.77 ± 15.52	6.22	10.43 ± 0.55	0.16	226.58 ± 12.18	3.45	600.78	1.52	

Note: LLD — lower limit on detection.

TABLE 3. URANIUM IN DRINKING WATER FROM THE KAFUE RIVER, CHINGOLA

Sampling date	Activity concentration (mBq/L)								Total	U-234/U-238
	U-234		U-235		U-238					
	Value	LLD	Value	LLD	Value	LLD	Value	LLD		
2009-08-11	85.9 ± 6.2	3.2	3.81 ± 0.28	0.18	82.9 ± 6.0	4.0	172.61	4.0	1.04	
2009-09-15	87.8 ± 7.4	8.0	2.90 ± 0.28	0.19	62.9 ± 6.0	4.2	153.60	4.2	1.40	
2009-10-13	183 ± 11.0	8.6	6.94 ± 0.45	0.21	151 ± 10	4.5	340.94	4.5	1.21	
2009-11-10	144 ± 7.0	1.0	5.68 ± 0.31	0.05	123 ± 7.0	1.0	272.68	1.0	1.17	
2010-02-04	28.6 ± 3.2	0.95	0.92 ± 0.12	0.044	20.0 ± 2.7	0.95	49.52	0.95	1.43	
2010-10-28	73.6 ± 8.6	2.6	2.90 ± 0.36	0.12	63.0 ± 7.8	2.6	139.50	2.6	1.17	
Mean	100.48 ± 7.2	4.06	3.86 ± 0.30	0.13	83.80 ± 6.58	2.88	188.14	2.88	1.24	

Note: LLD — lower limit on detection.

TABLE 4. URANIUM IN DRINKING WATER FROM MWAMBASHI STREAM (A TRIBUTARY OF THE KAFUE RIVER), GARNETONE TOWNSHIP, KITWE

Sampling date	Activity concentration (mBq/L)							
	U-234		U-235		U-238		U-234/U-238	
	Value	LLD	Value	LLD	Value	LLD	Value	LLD
2009-08-11	39.8 ± 4.7	6.8	1.30 ± 0.17	0.15	28.3 ± 3.7	3.3	69.40	1.41
2009-09-15	147.0 ± 17	12	2.19 ± 0.45	0.54	47.6 ± 9.7	12	121.19	3.09
2009-10-13	78.9 ± 7.6	6.3	1.73 ± 0.25	0.34	37.6 ± 5.3	7.4	88.23	2.07
2009-11-10	91.3 ± 9.8	13	1.82 ± 0.28	0.31	39.6 ± 6.2	6.8	84.98	2.31
2010-02-04	15.8 ± 2.4	0.97	0.36 ± 0.08	0.05	7.91 ± 1.69	0.97	24.07	1.00
2010-10-28	55.7 ± 5.1	1.2	1.21 ± 0.16	0.06	26.2 ± 3.4	1.2	83.11	1.60
Mean	71.42 ± 7.77	6.71	1.44 ± 0.23	0.24	31.20 ± 5.0	5.28	78.0	1.91

Note: LLD — lower limit on detection.

TABLE 5. URANIUM IN WATER FROM THE KAFUE RIVER, BULANGILO TOWNSHIP, KITWE

Sampling date	Activity concentration (mBq/L)										Total	U-234/U-238
	U-234		U-235		U-238							
	Value	LLD	Value	LLD	Value	LLD	Value	LLD	Value	LLD		
2009-08-11	61.9 ± 8.4	9.4	2.99 ± 0.38	0.34	64.9 ± 8.2	7.5	129.79					0.95
2009-09-15	86.4 ± 10.3	14	3.73 ± 0.42	0.29	81.1 ± 9.2	6.4	171.23					1.07
2009-10-13	128.0 ± 8.0	3.5	4.28 ± 0.30	0.06	92.9 ± 6.6	1.3	225.18					1.38
2009-11-10	97.1 ± 6.5	1.1	4.31 ± 0.29	0.05	93.6 ± 6.2	1.1	195.01					1.04
2010-02-04	21.6 ± 3.5	1.5	1.07 ± 0.16	0.07	23.2 ± 3.5	1.5	45.87					0.93
2010-10-28	79.0 ± 6.1	1.2	3.08 ± 0.26	0.06	66.9 ± 5.5	1.2	83.11					1.18
Mean	79.0 ± 7.13	5.12	2.53 ± 0.30	0.15	70.43 ± 6.53	3.17	141.70					1.09

Note: LLD — lower limit on detection.

TABLE 6. URANIUM IN DRINKING WATER FROM THE KAFUE RIVER, NKANA EAST TOWNSHIP, KITWE

Sampling date	Activity concentration (mBq/L)								Total	
	U-234		U-235		U-238		U-234/U-238			
	Value	LLD	Value	LLD	Value	LLD	Value	LLD		
2009-08-11	51.4 ± 7.2	7.0	1.70 ± 0.29	0.47	37.0 ± 6.4	10.0			90.10	1.39
2009-09-15	103.0 ± 11	8.5	2.50 ± 0.36	0.14	54.4 ± 7.8	3.0			159.90	1.89
2009-10-13	110.0 ± 10	7.4	4.02 ± 0.38	0.10	87.4 ± 8.3	2.2			201.42	1.26
2009-11-10	111.0 ± 9	1.9	3.81 ± 0.35	0.10	82.7 ± 7.7	1.9			197.51	1.34
2010-02-04	10.9 ± 2.3	1.3	0.52 ± 0.11	0.05	11.4 ± 2.3	1.3			22.82	0.96
2010-10-28	87.6 ± 7.7	1.7	3.46 ± 0.32	0.08	75.0 ± 6.9	1.7			166.06	1.17
Mean	60.48 ± 7.87	4.63	2.67 ± 0.30	0.16	57.98 ± 6.57	3.35			139.64	1.34

Note: LLD — lower limit on detection.

TABLE 7. TOTAL URANIUM IN DRINKING WATER FROM THE SIX SAMPLING LOCATIONS

Sampling date	Total activity concentration (mBq/L)					
	Chingola, underground water	Chingola, surface water	Chambeshi, underground water	Kitwe (Garnetone), surface water	Kitwe (Bulangililo), surface water	Kitwe (Nkana East), surface water
2009-08-11	122.13	172.61	690.20	69.40	129.79	90.10
2009-09-15	91.21	153.60	937.70	121.19	171.23	159.90
2009-10-13	93.15	340.94	559.51	88.23	225.18	201.42
2009-11-10	176.02	272.68	747.00	84.98	195.01	197.51
2010-02-04	80.46	49.52	553.53	24.07	45.87	22.82
2010-10-28	76.00	139.50	116.75	83.11	148.98	166.06
Mean	77.16	188.14	600.78	78.50	152.68	139.64

TABLE 8. DOSES VIA THE DRINKING WATER PATHWAY

	Activity concentration (mBq/L)			Annual effective dose (μ Sv/a)			Total
	U-234	U-235	U-238	U-234	U-235	U-238	
Chingola, underground water	59.03	2.23	48.43	2.11	0.08	1.59	3.78
Chambeshi, underground water	363.77	10.43	226.58	12.48	0.36	7.44	20.28
Chingola, surface water	100.48	3.86	83.80	3.59	0.13	2.75	6.47
Kitwe (Garnetone), surface water	71.42	1.44	31.20	2.55	0.01	1.02	3.58
Kitwe (Bulangililo), surface water	79.0	2.53	70.43	2.83	0.01	2.31	5.15
Kitwe (Nkana East), surface water	60.48	2.67	57.98	2.16	0.01	1.91	4.08

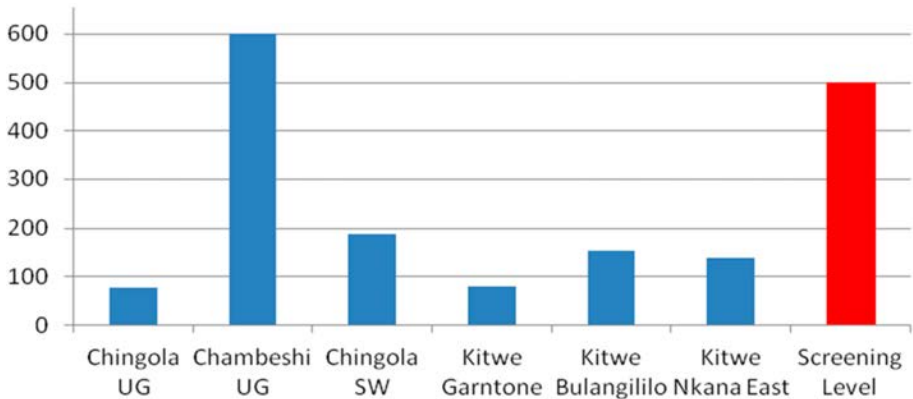


FIG. 2. Total uranium in drinking water (mBq/L).

ASSESSMENT OF URANIUM IN DRINKING WATER IN ZAMBIA

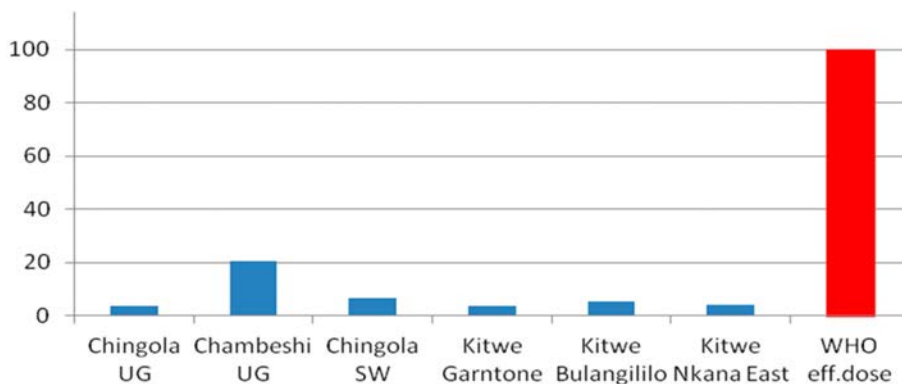


FIG. 3. Dose from the drinking water pathway ($\mu\text{Sv/a}$).

4. DISCUSSION

The results of this survey show that the activity concentrations of uranium isotopes in drinking water in the three towns can vary over wide ranges. The ranges of ^{238}U concentrations were as follows:

- Chambeshi, underground: 57.5–340.0 mBq/L;
- Chingola, underground: 34.9–76.5 mBq/L;
- Chingola, surface: 20–82.9 mBq/L;
- Kitwe, surface (Garnetone): 7.91–47.6 mBq/L;
- Kitwe, surface (Bulangililo): 23.2–93.6 mBq/L;
- Kitwe, surface (Nkana East): 11.4–87.4 mBq/L.

There was a tendency for the ^{234}U concentrations to be higher than the ^{238}U concentrations, usually by a moderate amount but occasionally up to as much as three times higher. The ^{235}U concentrations were consistently lower than the corresponding ^{238}U concentrations by a factor of 22, consistent with the natural abundance ratio. The committed effective dose were in the range of 3.58 $\mu\text{Sv/a}$ for Kitwe (Garnetone) to 20.28 $\mu\text{Sv/a}$ for Chambeshi.

This is the first time this kind of investigation has been carried out in Zambia. The uranium activity concentrations observed in this work were higher than reported for drinking water in the United States of America [11]. This was expected, considering the nature of the uranium mineralogy in Zambia. The uranium activity concentrations for underground water were higher than those in surface water. This finding is similar to the situation in the United States of America where, in some states, concentrations in groundwater have been found to

be up to four times higher than in surface water [12]. The differences in uranium concentrations observed across the various sampling sites can be attributed to the uranium concentrations in the aquifer rock and the chemical composition of water, including pH, redox potential, oxygen content, carbon dioxide content and complexing agents [12].

5. CONCLUSIONS

It can be concluded from the limited amount of measurement data gathered in this study that the uranium detected in all the water samples originated from natural sources with a ^{235}U : ^{238}U activity concentration ratio of 0.046, the natural abundance ratio. There was disequilibrium between ^{234}U and ^{238}U in all the samples. The activity concentrations of ^{234}U from both underground and surface waters were higher than those of ^{238}U and the ^{234}U : ^{238}U activity concentration ratios for surface water and groundwater were similar. This indicates that the rate of leaching and displacement of ^{234}U from the host minerals was greater than that of ^{238}U .

The water at Chambeshi had the highest total uranium activity concentration of 600.78 mBq/L, higher than the WHO recommended level of 500 mBq/L for alpha emitters. This water was associated with the highest annual committed effective dose of 20.28 μSv . Although the maximum committed effective dose was below the WHO guideline value of 100 μSv , the nature of the uranium mineralogy in this mining area of Zambia suggests that the screening of drinking water for uranium and other radionuclides is to be encouraged.

ACKNOWLEDGEMENTS

The authors would like to thank the National Scientific and Technology Council and the National Institute for Scientific and Industrial Research of the Republic of Zambia for the grant to carry out this study.

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CONTAMINATION AT THE LEGACY SITE OF A RARE EARTHS PLANT: A CASE STUDY

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Abstract

At a former large rare earths plant known as YL, a serious pollution problem was caused by obsolete technology and ineffective management during past production activities. When the plant closed (without decommissioning) because of bankruptcy, large amounts of NORM waste containing ^{226}Ra and ^{232}Th were left behind. The site became a legacy site and a potential threat to the surrounding environment. A decommissioning project including radiation measurements and remedial actions was initiated and, finally, the site was successfully decommissioned and released for unrestricted future use. During the course of the site survey, the contamination status was determined from the results of radiation measurements. Owing to the presence of NORM, particular problems were encountered during the decommissioning process. These problems, and the solutions found, are described. The experience gained at this site could be helpful for future decommissioning activities.

1. INTRODUCTION

Over the past few years, many decommissioning activities have been carried out in China, but the decommissioning of a former rare earths plant known as YL was the first time that particular problems with NORM were faced. YL used to be a large scale rare earths processing plant. It was built in the 1960s near a large river and covered an area of 160 000 m². Initially, a large amount of monazite was used for rare earth production and serious pollution problems were caused by obsolete technology and ineffective management. In 2003, owing to bankruptcy, the plant was closed without being decommissioned and became a legacy site. During past production activities, about 10 000 m³ of radioactive waste containing ^{226}Ra and ^{232}Th were left at the site and posed a threat to the surrounding environment [1]. In order to eliminate the threat and protect the environment, a decommissioning project was launched by the Ministry of Environmental Protection in 2009. A radiological site survey, decommissioning monitoring and a termination survey were carried out successfully. After removal of the radioactive material, decontamination of the structures and equipment and

cleanup the contaminated area, the site was released at the end of April 2010 for unrestricted use.

Because of the presence of NORM, particular problems were encountered during the decommissioning process, caused by a lack of technical standards and operational guidelines, a lack of effective management and protection measures in the past and NORM waste disposal issues. The contamination status of the site was determined from the results of site survey measurements and the particular problems associated with such a legacy site contaminated by NORM were identified and solved. The same problems may not necessarily be encountered at all decommissioning sites but the experience gained at this site could be helpful for most other future decommissioning efforts.

2. DECOMMISSIONING PROJECT

2.1. Objectives

The objectives of the decommissioning project were defined as follows:

- (a) Decontamination of on-site buildings and structures;
- (b) Dismantling and decontamination of production equipment and recycling of scrap metal;
- (c) Cleanup and restoration of contaminated on-site areas;
- (d) Removal of radioactive materials including radioactive waste and non-metallic components;
- (e) Release of the site from regulatory control for unrestricted future use;
- (f) Protection of the decommissioning workers and the public to meet the relevant regulations and standards.

2.2. Decommissioning programme

The main steps of the decommissioning programme are summarized in Fig. 1.

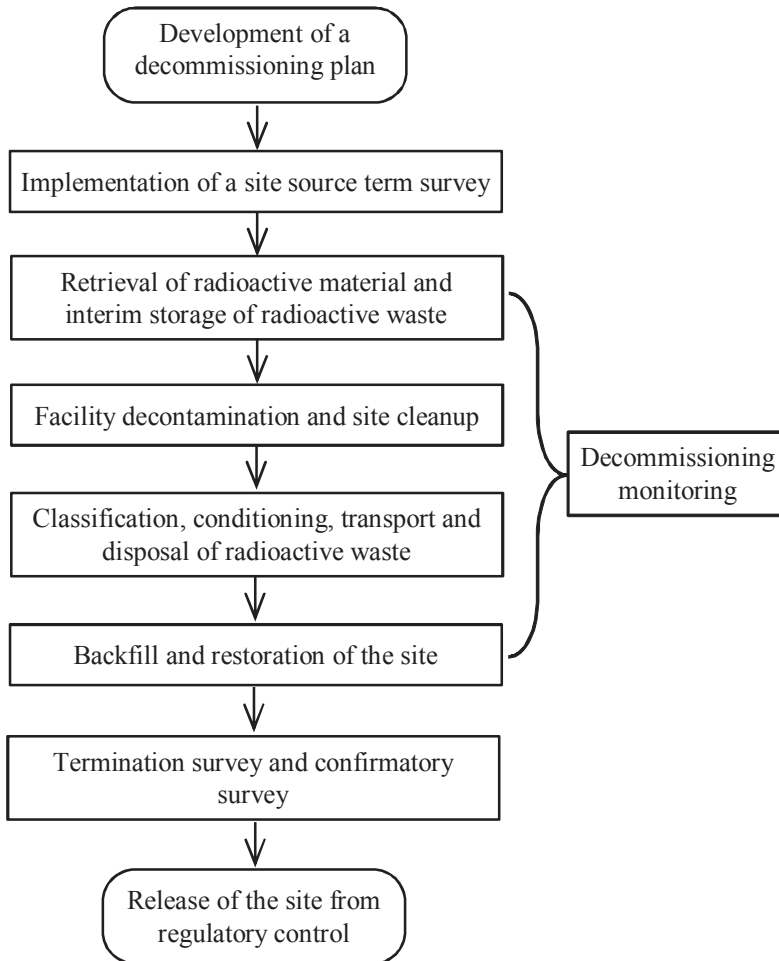


FIG. 1. Flow diagram for the decommissioning project.

2.3. Scope and content of the decommissioning plan

When implementing the source term survey, areas of the site were classified into three categories according to the level of contamination: areas with a potential for high levels of contamination, areas with a potential for low levels of contamination and areas with no contamination. The plant site was divided into 15 blocks. Ten blocks in the production area (the eastern half of the plant site) and

three parts of the office area were identified as potentially highly contaminated areas requiring remediation, including in particular:

- Workshops 1–8;
- Semifinished library;
- Waste repository;
- Areas surrounding these facilities;
- Part of a wall foundation in the west of the plant;
- Waste piles in the south of the site [2].

2.4. Radiation measurements

The decommissioning project involved a source term survey of the site, monitoring during decommissioning activities and, finally, a termination survey. Field measurements and laboratory analyses were conducted as part of all the above activities but the methods varied. Generally, gamma dose rates, surface contamination and radon concentrations were included in the field measurements. Samples of soil, slag and water, as well as smear samples from contaminated surfaces, were gathered from various locations. Concentrations of ^{232}Th and ^{226}Ra , gross alpha activity and gross beta radioactivity were determined in the laboratory.

2.5. Decommissioning criteria

The decommissioning criteria included action levels for gamma dose rate, a classification scheme for radioactive waste, clearance levels for materials and acceptable levels of residual radionuclides in soil. The criteria are listed in Table 1.

3. CONTAMINATION EVALUATION

3.1. Contamination status

The contamination of the plant was expressed in terms of the contaminated areas and the soil depths in need of decontamination. A contamination map of areas of surface soil is shown in Fig. 2. The areas are delineated by the colours red, yellow and green according to the dose rate (and white for locations that could not be accessed). Furthermore, the hot spots in the confirmed contamination areas (red areas) were also indicated on the contamination map.

CONTAMINATION AT THE LEGACY SITE OF A RARE EARTHS PLANT

TABLE 1. DECOMMISSIONING CRITERIA

	Description	Value
Action levels of gamma dose rate, preliminary identification	Non-contaminated	<0.5 $\mu\text{Sv/h}$
	Suspected contamination	0.5–1 $\mu\text{Sv/h}$
	Confirmed contamination	>1 $\mu\text{Sv/h}$
Classification of radioactive waste	Exempt waste	<1 Bq/g
	Very low level waste	1–20 Bq/g
	Low level waste	20–1 000 Bq/g
	Intermediate level waste	1 000–40 000 000 Bq/g
Clearance of materials	Alpha surface contamination	<0.08 Bq/cm ²
	Beta surface contamination	<0.8 Bq/cm ²
	Recycling and use of scrap metal, alpha surface contamination	<20 Bq/cm ²
	Residual radionuclides in soil	²²⁶ Ra and ²³² Th concentrations

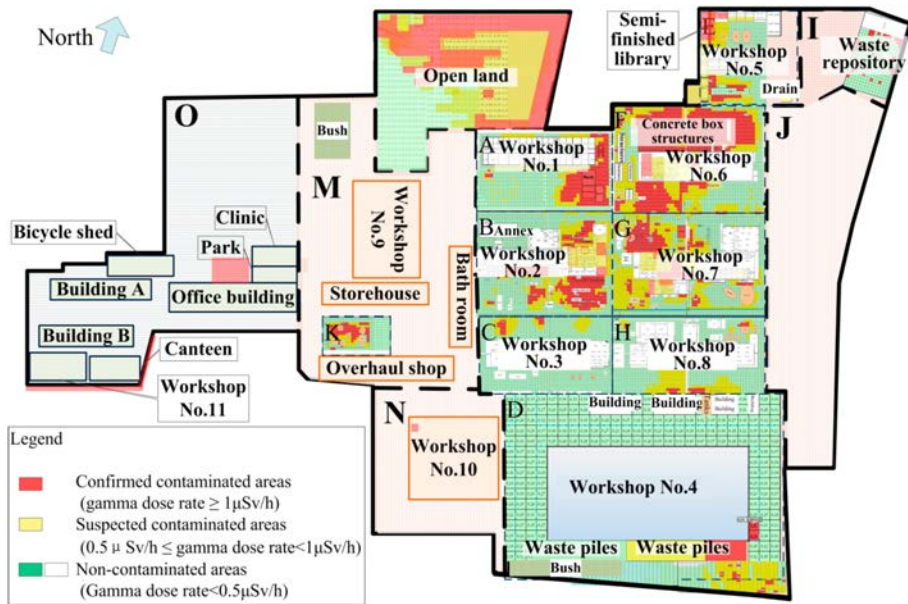


FIG. 2. Contamination map of ground surface areas.

According to the results of the source term survey, a total surface area of more than 21 000 m² was contaminated by NORM. The subsurface contaminated areas were identified during the decommissioning process. A total area of about 30 600 m² was found to be contaminated and the depths in need of cleanup were up to 3 m, with an average of about 1.5 m. The maximum dose rates measured at the site were 800 μSv/h in the facilities and 110 μSv/h in open areas. The samples taken at the site had a wide range of activity concentrations. Generally, the radiation level varied with the different stages of rare earths production. The more the process approached the finished product stage, the higher the radiation level. The radiation level also depended on the different production technologies used. The more advanced the production technology, the lower the radiation level. The contamination areas including subsurface soil are shown in Fig. 3.

3.2. Generation of radioactive waste

3.2.1. Amounts

The waste generated from the decommissioning project was categorized according to the activity concentration of samples taken from the waste. During the decommissioning, about 35 000 m³ of very low level waste (VLLW), 2961 m³ of low level waste (LLW) and 99 m³ of intermediate level waste (ILW) were generated [2].

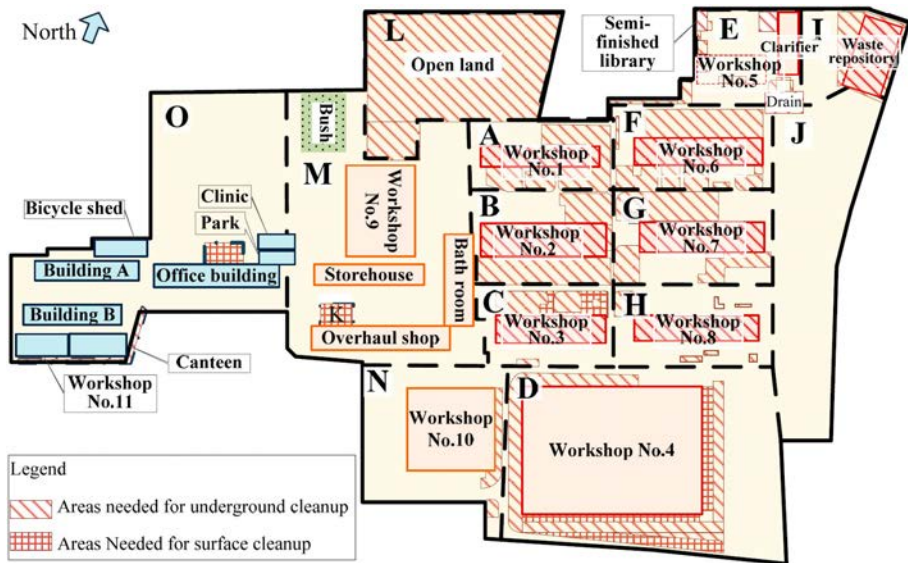


FIG. 3. Contamination map including areas of subsurface contamination.

3.2.2. *Disposals*

The following options for disposal were considered:

- (a) EW was used for backfill on site or for other unrestricted uses.
- (b) VLLW was sent for disposal in a municipal solid waste landfill facility with limited regulatory control.
- (c) LLW was sent for disposal in an engineered near surface facility.
- (d) ILW was sent for disposal in an engineered facility located in deep stable geological formations.
- (e) Scrap metal would be sent back for melting after decontamination.

4. PROBLEMS AND SOLUTIONS

Since this was the first NORM contaminated site in China to be decommissioned and remediated, there was no prior experience to learn from and the difficulties had to be solved as they arose. The particular circumstances of the decommissioning project led to the following problems being encountered.

4.1. Case 1

The source term was complicated for reasons such as:

- (a) The NORM waste was used in buildings and for road bases.
- (b) Areas containing layers of contaminated material had been covered with concrete.
- (c) Cone shaped deposits of contamination had been buried due to subsequent backfill.
- (d) NORM waste was located beneath buildings or structures that had been constructed subsequently (see Fig. 4).

The problem and its solution can be summarized as follows:

- Problem 1: The complicated distribution of the contamination made it very difficult to get accurate results in the source term survey.

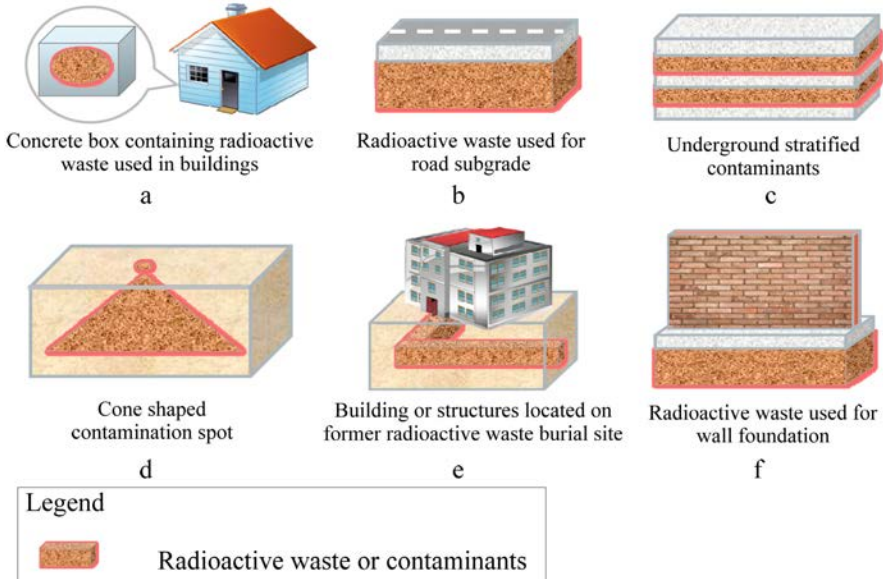


FIG. 4. Configurations of source terms identified at the plant site.

- Solution: Devoting more effort to the gathering of information. Information can be gathered by document review, personal interviews and site reconnaissance [3]. Generally, a more conservative estimate of the contamination status can be made in the preliminary site survey, and the source term survey should ideally be conducted throughout the decommissioning process.

4.2. Case 2

The sampling method adopted in the source term survey was based on an assumption that the vertical distribution of contamination was consistent with what was observed at the surface. The sampling points were mainly selected at hotspots as shown in Fig. 5. Sometimes, however, the contamination status of the subsurface soil was quite different from that at the surface, an example being the cone shaped contamination referred to in Case 1, where direct measurements and sampling at the surface did not work effectively. Such a sampling approach resulted in an underestimation of the subsurface contamination. The problem and its solution can be summarized as follows:

- Problem 2: It is very difficult to ensure that soil sampling locations and direct surface measurements are representative of the subsurface contamination.

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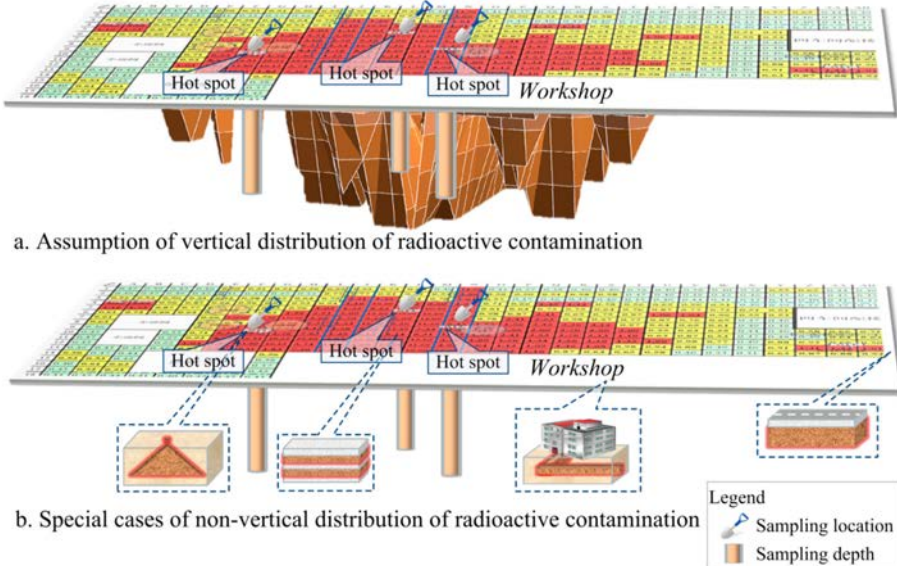


FIG. 5. Spatial distribution of radioactive contamination.

- Solution: Devoting more effort to the techniques and methods for sampling at special places, such as concrete covered ground, cobblestone riverbeds and deep subsurface contamination. Sometimes, underground radium contamination can be identified by the measurement of radon.

4.3. Case 3

The criteria for radioactive waste classification are expressed in the national regulations in terms of activity concentration and half-life [4]. However, it might be unrealistic to have to make large numbers of activity concentration measurements in the limited timescale of the decommissioning project. The problem and its solution can be summarized as follows:

- Problem 3: The requirement for classifying the NORM waste precisely is at odds with the need to ensure the progress of the project.
- Solution: Developing a quick classification method based primarily on field measurements rather than on laboratory analysis. The radioactive waste can be classified by measuring the gamma radiation level with portable instrumentation. This will require that a set of factors for converting dose rate measurements (in microsieverts per hour or counts per second) to activity concentrations (in becquerels per gram) needs to be derived.

4.4. Case 4

Because most NORM facilities were not associated with radioactivity in the past, the management and supervision of radiation safety were not taken seriously. Therefore, some special problems which differ from other decontamination and decommissioning activities were encountered in the decommissioning project. These problems were solved successfully based on expert judgment with regard to specific conditions. However, these may be encountered in other cases and may not be solved in the same way. These problems can be summarized as follows:

- Problem 4: The range of NORM industries and the NORM residues generated is not defined clearly, leading to situations in which radioactivity may be encountered unexpectedly during decommissioning and remediation of mineral processing facilities.
- Problem 5: There is a lack of information on historical production activities in mineral processing facilities and on management regulations and operational technical standards relevant to NORM.
- Problem 6: The disposal methods for NORM waste are not specified clearly.
- Problem 7: The decommissioning of NORM facilities has not yet attracted widespread attention.

The following suggestions are made:

- Suggestion 1: Improve the review and regulatory mechanism and establish management regulations and operational technical standards for NORM.
- Suggestion 2: Perform radioactive source term surveys with due account taken of special problems of the NORM industry and conduct research activities focusing on the evaluation and control of NORM contamination.
- Suggestion 3: Strengthen the training of radiation protection personnel and workers with respect to basic knowledge and operational skills related to control of exposure to NORM.
- Suggestion 4: Ensuring timely processing and disposal of radioactive waste, with due regard to radioactive waste minimization.

5. CONCLUSIONS

Through this case study of the decommissioning of a NORM facility, some common and special problems have been mentioned and consideration has been given to solutions and suggestions. It should be noted that the above mentioned problems are only the tip of the iceberg in a decommissioning project. Even if the

problems have been solved, more efficient solutions may also exist. However, these solutions and suggestions would be helpful to future decommissioning activities. Finally, it is expected that more attention could be given to NORM contamination problems.

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^{222}Rn AT A GROUNDWATER TREATMENT PLANT

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Abstract

Groundwater is a source of drinking water, usually of good quality, but compared with surface water the concentrations of radionuclides of natural origin are higher. A drinking water treatment plant for treating groundwater was monitored. The raw water is aerated, filtered through gravity sand filters and then aerated again. Radium-226 extracted from the raw water is partially retained in the filtration sand. Decay of the accumulated ^{226}Ra generates gaseous ^{222}Rn which is released into the treated water and into the air of the plant hall, especially during the washing of the filters. Radon-222 can pose a health risk to the operating personnel or to the public. The study evaluated the following factors: ^{222}Ra and ^{226}Ra concentrations in the raw and treated water, the amount of ^{226}Ra accumulated in the filtration sand and the ^{222}Rn concentration in the air of the plant.

1. INTRODUCTION

The quality of groundwater used as a source of drinking water can vary widely. Groundwater is generally a drinking water source of good quality but the concentrations of radionuclides of natural origin are higher than those in surface water. During the water treatment process, radionuclides are removed from the water together with ferric and manganic oxides [1–6]. After the raw water has been aerated, it is filtered in gravity filters or pressure filters filled with sand, or more recently with other filtration media. The use of this process stretches back to times when little was known about natural radioactivity in water and the associated risks from its ingestion [7].

The plant chosen for this study processes groundwater from a high quality underground source and distributes it in bottles as table water. There are six gravity filters in the plant, filled with filtration sand, naturally covered with manganic and ferric oxides. The filtration hall is shown in Fig. 1. Radium-226 retained in the filtration medium decays to produce gaseous ^{222}Rn . During water treatment, ^{222}Rn is released into the treated water. At the filter washing stage, it is released from the water into the air of the plant hall, where it can represent a potential health risk for the operating personnel.



FIG. 1. The filtration hall with open filters.

2. METHODS

Large volume samples of the raw water and treated water, as well as samples of sand from the filters, were collected on a regular basis. In the laboratory, the water samples were evaporated and the sand samples were dried at 105°C. The concentrations of ^{226}Ra in the dried water and sand samples were determined by gamma spectrometric analysis using a Canberra-Packard S 100 instrument with a high purity germanium detector, ADC, MCA, according to the standard ČSN ISO 10703 (75 7630) [8]. The ^{222}Rn concentrations in the raw water, and in the treated water after final aeration, were determined using the emanation method described in ČSN 75 7624, Water quality: Determination of radon-222. The ^{222}Rn concentration in the air inside the building was measured using an automatic radon concentration monitor Radim 3. The ^{222}Rn concentration was determined via the ^{218}Po activity, collected on the surface of a semiconductor detector with an electric field. The minimum detectable activity was 30 Bq/m³ for a measurement period of 1 h, determined with a statistical error of 20%. The instrument was powered by a rechargeable battery with an operating life of 400 h.

In addition to measuring the ^{226}Ra activity concentration in the filtration sand, the gamma dose rate was at the same time measured at 15 locations on the surface of each of the drained filters, using an NB 3201 monitor containing a plastic scintillator. The dose rate was derived from the anode current of the photomultiplier. The monitor was capable of measuring dose rates in the range 0.01–30 000 nGy/s.

The measuring instruments are regularly verified by the Czech Metrological Institute, in accordance with legal requirements.

Monitoring at this plant started in 1996 and is ongoing. The results presented in this paper relate to the period 1996–2011.

2.1. Removal of radionuclides from the water

The groundwater from the artesian borehole used as the raw water supply has a relatively stable ^{226}Ra and ^{222}Rn content, with average concentrations of 0.186 and 5.83 Bq/L, respectively. At the beginning of the monitoring period, the ^{226}Ra concentrations were more variable, as a result of disturbance of the filtration sand, in some cases by its exchange. The ^{222}Rn and ^{226}Ra concentrations in the treated water are also stable over the long term, with average concentrations of 0.072 and 5.65 Bq/L, respectively. The values during the period 1996–2011 are shown in Figs 2 and 3. The long term results show that ^{226}Ra is removed

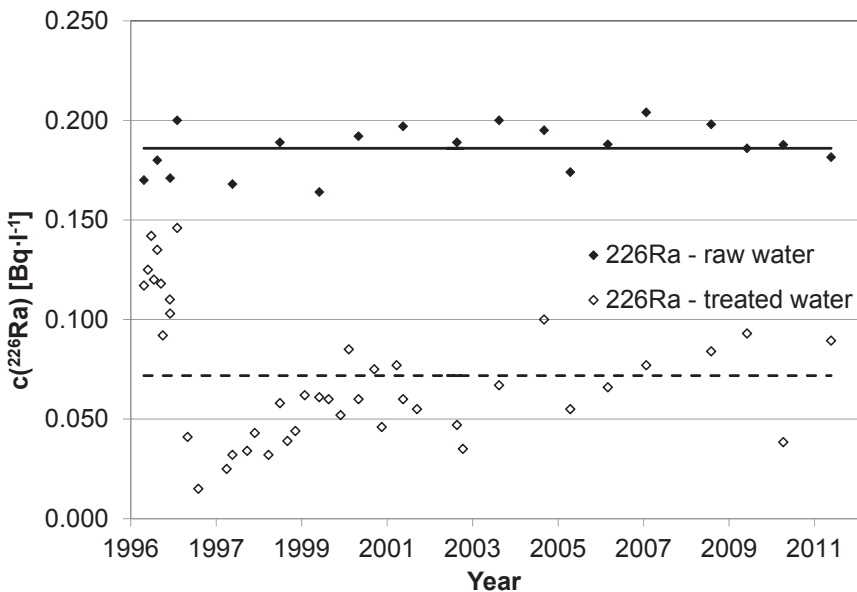


FIG. 2. Radium-226 concentrations in the raw and treated water.

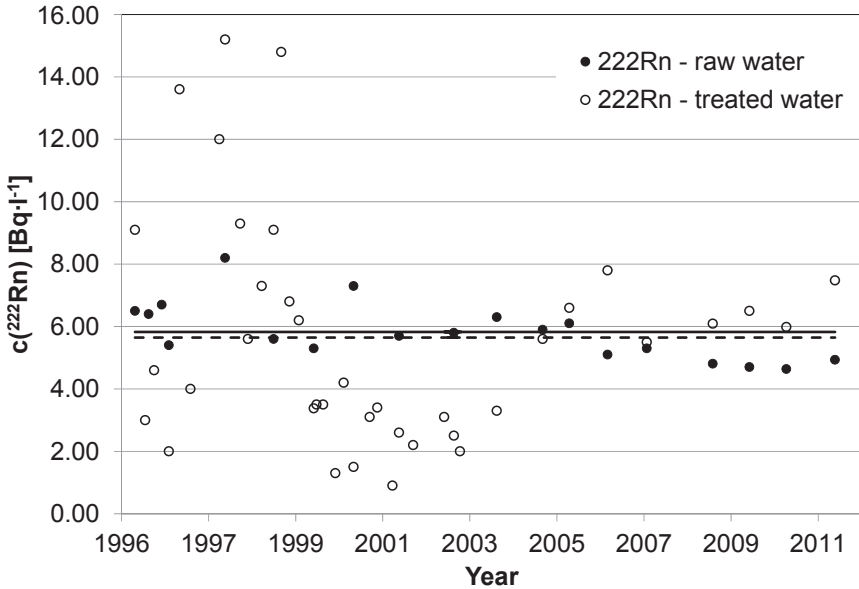


FIG. 3. Radon-222 concentrations in the raw and treated water.

from the water with approximately 60% efficiency. On the other hand, the overall efficiency of ^{222}Rn removal is only about 3%. Although, ^{222}Rn is initially expected to be removed by aeration with an efficiency of over 90 %, it appears that, during the subsequent filtration, the water is enriched again with ^{222}Rn originating from the ^{226}Ra accumulated in the filtration sand. In this plant, another aeration step follows the filtration, in order to lower the ^{222}Rn contamination originating from filters.

2.2. Contamination of the treated water by ^{222}Rn from the filter sand

The degree of secondary ^{222}Rn contamination of the treated water depends on the ^{226}Ra activity concentration in the filter sand, the filter loading, the residence time and the emanation coefficient. Assuming that all the ^{222}Rn released from the sand enters the filtered water, the ^{222}Rn concentration in the treated water can be calculated from the following equation:

$$c_{\text{Rn}} = \frac{a_{\text{Ra}} \cdot \frac{\lambda_{\text{Rn}}}{\lambda_{\text{Rn}} - \lambda_{\text{Ra}}} \cdot (e^{\lambda_{\text{Ra}} t} - e^{-\lambda_{\text{Rn}} t})}{L \cdot t_{\text{det}}} + c_{0\text{Rn}} \cdot e^{-\lambda_{\text{Rn}} t} \quad (1)$$

where

- c_{Rn} is the ^{222}Rn activity concentration in the water after filtration (Bq/L);
 a_{Ra} is the ^{226}Ra activity concentration in the sand (Bq/kg);
 λ_{Ra} is the ^{226}Ra decay constant ($4.94 \times 10^{-8} \text{ h}^{-1}$);
 λ_{Rn} is the ^{222}Rn decay constant ($7.55 \times 10^{-3} \text{ h}^{-1}$);
 t is the time (h);
 L is the filter loading ($\text{kg}^{-1} \cdot \text{h}^{-1}$);
 t_{det} is the medium retention time of water in the filter (h);

and $c_{0\text{Rn}}$ is the ^{222}Rn activity concentration in the water before filtration (Bq/L).

The first part of Eq. (1) characterizes the generation of ^{222}Rn by the decay of ^{226}Ra , retained in the filtration sand, and its simultaneous decay. The second part of the equation describes the spontaneous decay of the ^{222}Rn entering the gravity filter with the raw water. Since $\lambda_{^{222}\text{Rn}} \gg \lambda_{^{226}\text{Ra}}$, Eq. (1) can be simplified to:

$$c_{\text{Rn}} = \frac{c_{\text{Ra}} \cdot (1 - e^{-\lambda_{\text{Rn}} \cdot t})}{L \cdot t_{\text{det}}} + c_{0\text{Rn}} \cdot e^{-\lambda_{\text{Rn}} \cdot t} \quad (2)$$

Using Eq. (2), the ^{222}Rn concentrations were calculated for the 24 h period following the filter washing. Figure 4 compares the measured ^{222}Rn concentrations in the filtered water with the values calculated from Eq. (2). At first, the ^{222}Rn concentration in the filtered water grows with time. The longer water is in contact with the filtration sand, saturated with ^{226}Ra , the higher the ^{222}Rn concentrations in the water leaving the filter. As the medium retention time is reached, the ^{222}Rn concentration settles at a stable value, as the contact time is never longer than the retention time.

As Fig. 4 illustrates, the theoretically calculated equilibrium values are higher than the measured values. The average value of the measured equilibrium ^{222}Rn concentrations was 47.4 Bq/L, while the corresponding calculated value was 67.0 Bq/L. This provides a method for calculating the emanation coefficient of the filtration sand (i.e. the fraction of ^{222}Rn , from the decay of ^{226}Ra in the sand) released into the water. The emanation coefficient K_e is defined as the ratio of the ^{222}Rn concentration in the aqueous phase to the ^{222}Rn concentration corresponding to the ^{226}Ra content in the filtration sand. Neglecting the transfer

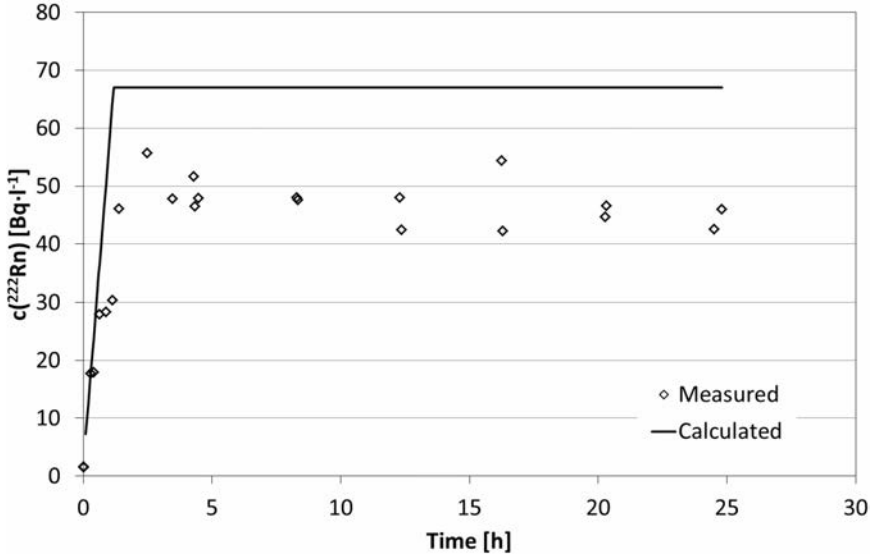


FIG. 4. Radon-222 concentrations measured in the water after filtration, compared with calculated values.

of ^{222}Rn through the quiescent water surface into the ambient atmosphere, the emanation is calculated from the following equation [9]:

$$K_e = \frac{c_{\text{Rn, measured}} - c_{0\text{Rn}}}{c_{\text{Rn, calculated}} - c_{0\text{Rn}}} \quad (3)$$

where

- K_e is the emanation coefficient of the filtration sand;
- $c_{\text{Rn, measured}}$ is the concentration of ^{222}Rn in water, measured after filtration (Bq/L);
- $c_{\text{Rn, calculated}}$ is the concentration of ^{222}Rn in water after filtration, calculated (Bq/L);

and $c_{0\text{Rn}}$ is the concentration of ^{222}Rn in water before filtration (Bq/L).

The emanation coefficient K_e of the filtration sand under operational conditions was found to be 0.70. The coefficient will be probably be dependent on the physical and chemical properties of the different filtration sands used in various treatment plants and will be specific to raw water of a particular quality.

2.3. Radon-222 concentration in the air of the plant

During the operation of the plant, slow filtration is alternated with washing of all six filters every three days. The filters are washed with a high flow rate to create turbulent conditions. During this process, the ^{222}Rn is vigorously released into the air. While monitoring the ^{222}Rn concentration in the air of the plant, a rapid increase in the ^{222}Rn concentration was detected when a filter was being washed. On completion of the filter washing process, the ^{222}Rn concentration values returned rapidly to their normal values. The highest peaks were detected in the filtration hall itself. An example of the time dependence of the ^{222}Rn concentration in the air of the filtration hall is shown in the Fig. 5. The maximum measured concentration was 2163 Bq/m^3 , compared with an average concentration of 141 Bq/m^3 .

The relationship between the maximum ^{222}Rn concentration in the air of the filtration hall and the ^{226}Ra concentration in the sand was also investigated. A linear relationship with a regression coefficient R^2 of 0.7845 was obtained, as shown in Fig. 6. However, this relationship was obtained over a short period of only three days and cannot be considered to be representative of the long term relationship taking in account all available data over the period 1996–2011. The relationship can be influenced by various factors such as seasonal changes, ventilation, and fluctuations in the rate of ^{222}Rn release from the sand particles

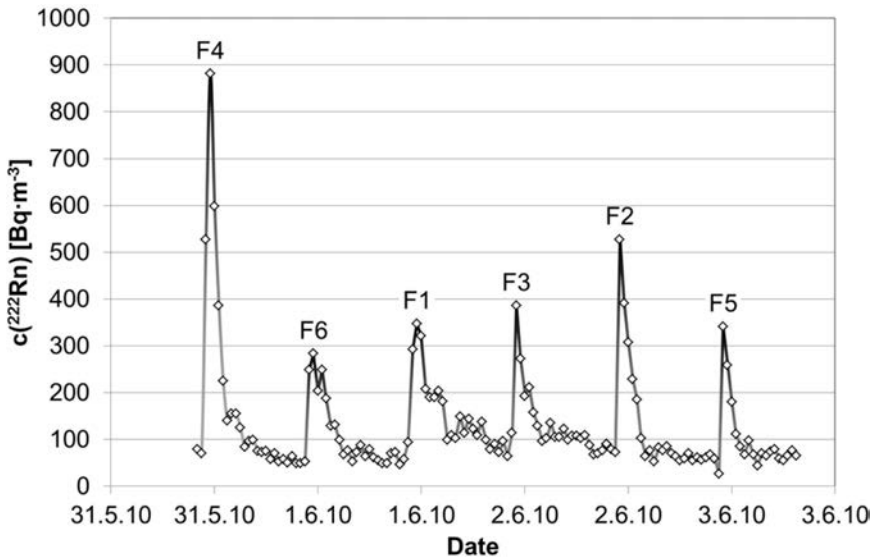


FIG. 5. Radon-222 concentration in air in the filtration hall during filter washing cycles.

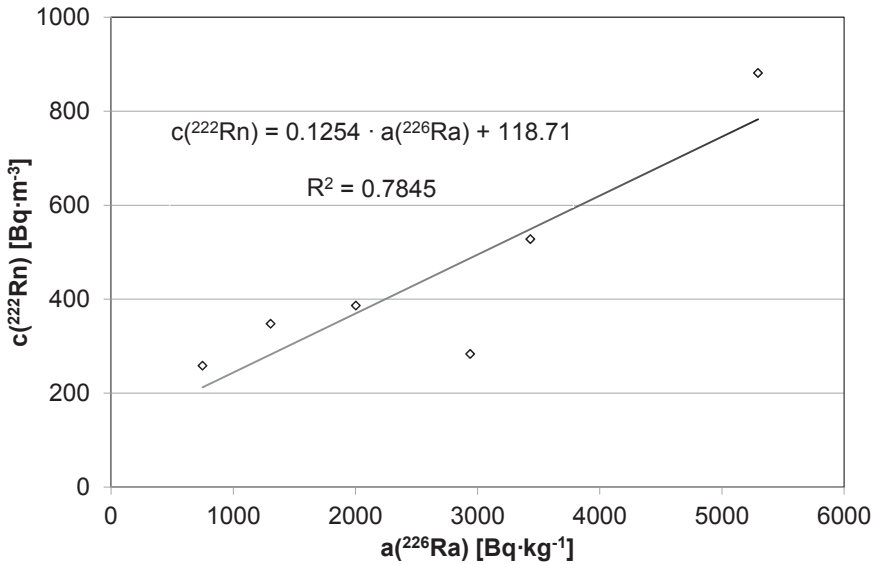


FIG. 6. Relationship between the maximum ^{222}Rn concentration in the air of the filtration hall and the ^{226}Ra activity concentrations in the filtration sand over a three day period.

caused by variations in the structure and thickness of the ferric and manganese oxide layer.

In other parts of the plant, including the staff room, the elevations in ^{222}Rn concentration were not as pronounced but were nevertheless significant. In the staff room, the maximum radon concentration was 645 Bq/m^3 , with an average value of 84 Bq/m^3 .

3. DOSE RATE AS A MEASURE OF ^{226}Ra ACTIVITY CONCENTRATION IN THE FILTRATION SAND

The ^{226}Ra concentration in the (dried) filtration sand, averaged over all filters, was 1.83 Bq/g . The range of concentrations was $0.336\text{--}5.623 \text{ Bq/g}$. Gamma dose rates at 15 locations on the surfaces of each of the drained filters were measured concurrently with the activity concentration measurements. The relationship between dose rate and ^{226}Ra concentration was linear, as shown in Fig. 7, and could be represented by the following equation:

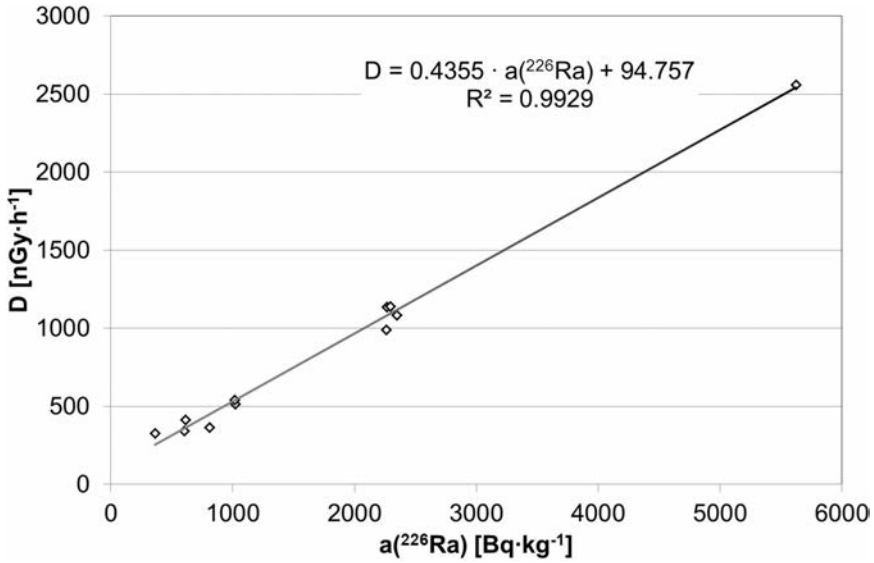


FIG. 7. Relation between gamma dose rate and ^{226}Ra activity in the (dried) filter sand.

$$D = 0.4355 \cdot a_{\text{Ra}} + 94.757 \quad (4)$$

where D is the gamma dose rate (nGy/h) and a_{Ra} is the ^{226}Ra activity concentration of the (dried) filtration sand (Bq/kg). The linear fit was very good, with a regression coefficient R^2 of 0.99. This linear relationship was used for calculating the ^{226}Ra content in the filter sand where a direct gamma spectrometric measurement was not available. A similar relationship between dose rate and ^{226}Ra activity concentration was observed for soils in areas of uranium mining. The study, described in Ref. [10], derived the following equation for dose rate measured 1 m above a surface with a homogenous ^{226}Ra distribution:

$$D = 0.459 \cdot a_{\text{Ra}} \quad (5)$$

which is very similar to Eq. (4).

4. CONCLUSION

Because elevated concentrations of ^{222}Rn can negatively affect human health, Czech legislation¹ requires the monitoring of ^{222}Rn in facilities where there is a possibility of elevated concentrations. Groundwater treatment plants are such facilities. In the plant investigated in the study, the ^{222}Rn concentrations in the air and in the treated water remained below the applicable reference values.

A secondary source of ^{222}Rn was identified in this work. During the water treatment process, ^{226}Ra is extracted from the raw water and retained in the filtration sand, thus ensuring that the treated water is suitable for drinking purposes. However, decay of the ^{226}Ra that accumulates in the filtration sand generates ^{222}Rn , which is then released back into the treated water or liberated into the air of the plant.

The ^{222}Rn concentrations in the air of the plant fluctuate with time. A rapid increase was recorded when a filter was being washed, followed by a subsequent rapid decay. A short term (3 day) relationship was observed between ^{222}Rn concentration in the air and the ^{226}Ra concentration in the filters. Over a long period, this relationship would change because of variations in other parameters related to the operation of the plant. Nevertheless, to optimize the radiation protection of the plant workers, filters with elevated ^{226}Ra concentrations are to be washed during periods when other plant workers are not present.

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¹ Regulation No. 307/2002 Coll. of radiation protection, as amended by regulation No. 499/2005 Coll.

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RAPPORTEUR SUMMARY OF TOPICAL SESSIONS 2 AND 3

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1. INTRODUCTION

This rapporteur summary covers the technical programme in Sessions 2 and 3 of the symposium, which included five papers on the topic of regulatory aspects and eleven papers on environmental aspects of NORM. A brief summary of each of the papers in both the sessions are given in this report.

2. REGULATORY ASPECTS OF NORM: MEETING THE NEW BSS REQUIREMENTS

2.1. Radon challenges in uranium mines and other workplaces

Mining and processing of uranium has been regulated many years as part of the nuclear fuel cycle. One of the important radiological hazards in mines and enclosed spaces is radon. The recent work of the International Commission on Radiological Protection (ICRP) on the risks associated with radon and move toward a fully dosimetric approach for regulating workplace exposure to radon has generated much discussion. This paper provided an industry perspective on associated issues which include, among other things, deficiencies in the ICRP's proposed dose coefficients arising from incomplete consideration of the carcinogenic effect of smoking and practical issues associated with the lack of dosimetrically relevant data to support a dosimetric approach. There are considerable uncertainties associated with the implementation of a fully dosimetric approach and the need for an operationally acceptable protocol for such measurements. Until such data are available, the current practice for monitoring, reporting and regulating miners' exposure to radon progeny is to continue. Radioactivity in NORM production is an important issue for public and regulators. While levels are relatively low, there is a concentration of radionuclides at some steps in processing, and care is needed. Proven radiation protection practices are available to protect workers and the public, and it is very important to communicate with the local public.

2.2. Radiological protection against radon exposure

ICRP Committee 4 is working on documents for existing exposure situations, including radon, NORM, cosmic radiation and contaminated sites, among others. The presentation covered the work of the task group on the updated guidance on radiological protection against radon exposure. Progression from ICRP 60 to ICRP 103 has led to a network of exposure situations — planned, existing and emergency situations; optimization below a dose restriction — a similar approach for all situations; dose limits and dose constraints for planned exposure situations; and a reference level for existing and emergency exposure situations. Existing exposure situations need to be characterized before trying to control difficult to control sources, but perhaps they can control pathways. The situation is usually characterized by a large distribution of individual exposures, radiation protection culture is often missing and consideration is on the long term perspective. NORM is considered mainly an existing exposure situation from an ICRP perspective. However, in some cases it can be planned (when the source is removed). The same approach is recommended whether exposure is deliberate or adventitious. Occupational exposure is considered as the responsibility of the management. The principle of justification — do more good than harm — is applicable. For NORM residues, special attention to recycling or use as a by-product needs to be considered. Optimization is below a reference level of dose and this level is not to be exceeded and below which the ALARA (as low as reasonably achievable) principle is applied. For radon in existing exposure situations, 10 mSv/a (middle of ICRP's band of 1–20 mSv/a) with a long term target of 1 mSv/a was recommended in the task group report. Graded approach and involvement of concerned parties were underlined.

To control the main part of radon exposure, the ICRP recommends an integrated approach focused as far as possible on the management of the building or location in which radon exposure occurs, whatever the purpose of the building and the types of its occupants. This approach is based on the optimization principle and a graded approach according to the degree of responsibilities at stake, notably in workplaces, and the level of ambition of the national authorities. The report emphasized the importance of preventive actions. The report also provided recommendations on how to control radon exposure in workplaces when workers' exposure can reasonably be regarded as being the responsibility of the operating management. In such a case, workers' exposures are considered as occupational and controlled using the corresponding requirements on the basis of the optimization principle and the application, as appropriate, of the dose limit.

2.3. Radiation safety regulatory policy and rule for NORM industries in China

Regulatory issues related to NORM industries in China were presented in the paper. The Ministry of Environmental Protection (MEP) is the responsible authority for NORM. Natural radiation exposures vary widely in China. Average annual background exposure from natural sources to public is about 3.1 mSv. South-east and south-west China have the highest external gamma levels. Laws on prevention of radioactive contamination and management of radioactive waste exist in the country. The Chinese BSS 2002 clearly defines requirements for NORM. The first list of requirements for exploration and exploiting of mineral resources was made available in 2013. The MEP published regulatory guide for non-uranium mines with NORM greater than 1 Bq/g. Regulatory framework for NORM includes special assessment for radiological impacts — assess and then check.

Coal mining is the largest industrial sector involving NORM in China. Several data tables with doses from NORM and indoor radon levels were presented. Around 800 NORM industries exist. Industries with elevated radioactivity greater than 1 Bq/g ^{238}U , ^{226}Ra or ^{232}Th include:

- Rare earths, 5709 nGy/h;
- Niobium–tantalum, 3263 nGy/h;
- Zircon, 1592 nGy/h.

The paper concluded with the observation that 10 million people work in mining, 100 million living in high radon homes and NORM is an urgent problem in China.

2.4. Preparing the building industry for the new Euratom BSS

Nucleair Technologisch Centrum (NuTeC) works with industry in Belgium. According to the EU BSS, building materials with NORM incorporation are regulated in the country as well as EU Member States. Regulation specifies the use of 1 mSv/a as the reference level for external gamma, uses the activity concentration index (ACI) — considering ^{226}Ra , ^{232}Th and ^{40}K activity in the material — and treats radon separately. To comply with these regulations, practical screening tools are needed. A new industrially useful protocol for measurement of ACI was developed to assess the applicability of newly developed materials for the European building market. The applicability of an in situ measurement methodology was investigated with the specific objective of aiding the industry in its search for cost efficient measurement techniques. An important aspect of the

investigation was the validation of the so-called B-NORM method that had been developed. A database from a large scale study from the port of Antwerp and the NuTeC project was compiled and analysed. The study compared on-site screening with $\text{LaBr}_3(\text{Ce})$ to high purity germanium, and good agreement was noted. In a reply to question from France, the European Union is developing two standards on the use of NORM and dose calculations and much discussion resulted from the European Atomic Energy Community (Euratom) based ACI index.

2.5. Progress in the regulation of radiation environmental safety in the utilization of mineral resources in China

The scope and requirements of regulation on the radiation and environmental safety in the utilization of mineral resources in China were stated. For the prevention of radioactive pollution, mines need study to receive approval that is adapted to the social environment and is consistent with international practices. The urgent need to regulate radiation in minerals industry was highlighted. The authors suggested international experience was the best precedent, such as ICRP 104 recommendations, NORM with less than 1 Bq/g exempted, IAEA regulatory guides and Safety Reports (e.g. Safety Report No. 49), the Euratom graded approach and United States Environmental Protection Agency NORM studies. However, it generally follows IAEA guidance. In 2013, the MEP issued regulatory directions for five industry sectors in China:

- Mining;
- Beneficiation and processing of rare earths;
- Niobium–tantalum;
- Zircon and zirconia;
- Vanadium and anthracite.

3. ENVIRONMENTAL ASPECTS OF NORM

3.1. NORM levels in mine pit lakes in south-western Spain

This paper investigated and analysed the natural radionuclide concentration at a former mining site Iberian pyrite belt, a historical mining area in Spain. The study examined radiological and environmental effects on four pit lakes. Water in the mine pits showed low pH (~2–3) and dissolution of metals and radionuclides was observed. The Odiel River transports the dissolved heavy metals and radionuclides all the way to the estuary. On mixing with seawater at the estuary, the river water is neutralized and the contaminants are precipitated. The Odiel

estuary is therefore serving as a repository for the contaminants liberated by the former mining activities. Some of the observations are:

- High arsenic levels;
- High levels of uranium and ^{230}Th in waters in three of the four pit lakes;
- Sediments show normal gamma values;
- Correlation between ^{234}U and iron, and conductivity;
- Impact on the fluvial system;
- Activity ratios affected by leaching;
- Chemistry changed on entering estuary.

The estuary was also affected by wastes from past fertilizer production.

3.2. Transfer of radioactivity in the treatment of wastewater by phosphate

The aim of this work was to study the transfer of ^{238}U and ^{232}Th during the treatment of wastewater by infiltration percolation through a phosphate bed. To assess radiation dose due to the treated water which is used in agriculture and for the irrigation of gardens in urban areas, these radionuclides were measured in wastewater, phosphate samples and treated water by using solid state nuclear track detectors. There has been increased demand to reuse water caused by mining in areas with low water potential — a new process to treat water using a bed of phosphate was proposed. Uranium and thorium have increased, but they rarely exceed World Health Organization (WHO) drinking water guideline levels and hence are tolerable. Results are not dependent on phosphate or water not contaminated by phosphate.

3.3. ^{222}Rn , ^{220}Rn concentrations and miner doses in non-uranium mines in China

Results of a survey carried out in 44 non-uranium mines in 12 provinces of China were presented. The workforce numbers around 21 million miners in non-uranium mines. The Ministry of Health wants to consider underground mining as occupational radiation exposure including metal mines (copper, ferrous and rare earths), non-metal (chemical materials and building materials) mines, energy sector mines (coal, oil and gas) and water or gas (mineral water and helium) facilities. Monitoring is carried out using track etch detectors hung in work areas of 25 metal mines, 18 non-metal and 1 spa (radon and thoron). Some of the mines showed high levels of ^{222}Rn (up to 7000 Bq/m^3) but lower levels of ^{220}Rn (around 220 Bq/m^3). High levels in rare earth facilities were also noted, but they need more study. The average dose to coal miners is 0.76 mSv/a .

3.4. Residual environmental contamination at a former ferroniobium production site in Belgium

From an aerial gamma survey, a site with residues from a former ferroniobium production facility was identified in Belgium. In close collaboration with the current site operator, an inventory of the mineral residue piled on the ground was made and the material was characterized in terms of its radionuclide content (<1–12 and <1–60 Bq/g for radionuclides of the uranium and thorium decay series, respectively). Some slags showed ^{226}Ra (4–5 Bq/g), ^{232}Th (60–70 Bq/g) and ^{40}K (5 Bq/g). Nearly 6000 t of ferroniobium residues were sent to the nuclear waste site. A pathway analysis for workers and public exposure was carried out and a maximum dose of 0.18 mSv/a from drum filling was noted. Leaching test showed leaching from barium, which implied a need for treatment. Dose estimates of the aquatic pathway indicated a maximum dose from drinking water of 2 L/d of 51 $\mu\text{Sv/a}$ — not a radiological health problem. The dose from using the site was estimated to be below 1 mSv/a, which means no immediate need to remediate. However, a long term strategy to clean up the entire site may be required.

3.5. Application of ICRP radiation protection principles in existing exposure situations with large volumes of NORM: The Wismut Case Study

The paper presented the background on the Wismut legacy — mines, waste rock and tailings totalling nearly 800 million t of lead to remediation of the site in Germany. The German Government earmarked €6.6 billion for remediation. Remediation goals driven by mining law to enable future land use and targeted reference level of 1 mSv/a for use of sites. Local rivers and groundwater are contaminated. Optimization of remediation took place based on best practice/experience, cost–benefit analysis, risk benefit (collective dose) and multiattribute analysis with the involvement of stakeholders. Waste rock cover pile in Schlema had a goal of 50 Bq/m³ of radon — but it could not be met, owing to convection affected cover performance and radon venting was not effective. Control of radon near surface mine galleries also posed challenges — turning off ventilation caused very high radon levels indoors. Protection of the environment may be an issue in future, and there may be a need to develop a culture of reference level acceptance. Whenever possible, transfer ‘wastes’ to productive use would be the better option.

3.6. The distribution characteristics of ^{222}Rn and ^{220}Rn concentrations in soil gas in Zhongshan City, Guangdong Province, China

A survey of ^{222}Rn and ^{220}Rn concentrations in soil gas was conducted at 67 locations across an area of 1800 km² in Zhongshan City, Guangdong Province. It was found that ^{222}Rn concentrations increased with increasing depth, whereas ^{220}Rn concentrations were almost unchanged at different depths. The sites with high ^{222}Rn values were located mainly in granite outcrops, while those with low values were located in a sedimentary region. The distribution patterns of ^{220}Rn coincide with the patterns of activity concentration of ^{232}Th in soil.

3.7. Dosimetric evaluation of thoron exposure in the three typical rural indoor environments of China

In rural areas of China, many homes are typically built from soils such as brick houses, mud houses and special buildings (caves), usually characterized by uncoated surfaces from which thoron gas is easily exhaled. In order to evaluate the dose from inhalation of thoron in these indoor environments, field measurements were carried out using portable devices for measuring the concentration and size distribution of thoron progeny. Indoor levels of radon and thoron are quite variable, and therefore focused measurement of progeny levels. Particle size distribution for ^{212}Pb was measured using double screen device. Particle size and equilibrium equivalent concentration for three kinds of rural dwelling were reported. In Yangjiang, the smallest particle size is in caves (50 nm) at 10 mSv/a (no indication of excess of cancer in this area in the last twenty years) and indicated that dose evaluation using dosimetric approach would be much higher.

3.8. Brazilian NORM industries: Lessons and challenges

NORM industries in various locations of Brazil include, among others, fertilizer, copper, niobium and coal. Uranium and thorium concentrations versus different types of NORM were presented. The main radiological issues relate to acid mine drainage in gold and coal mines, internal and external dose to workers and public and inadequate disposal of residues. Small companies often do not know there is an issue, and the need for better education was highlighted. There are concerns of ^{210}Pb in pyrometallurgical activities. CNEN Standard NE.4.01 applies only to NORM in mining, not oil and gas. It was mentioned that it would be easier to work with activity levels rather than dose based on land use. There were calls to stimulate the use of residues and, for instance, adopt 1 Bq/g for agriculture and work on phosphogypsum in civil construction.

3.9. Assessment of uranium in drinking water in Kitwe, Chambeshi and Chingola in the Copperbelt region of Zambia

This paper discussed uranium in drinking water in three towns located in the uraniumiferous Copperbelt region of Zambia. Mining copper is a major economic activity, along with agriculture and tourism. Wastewater from copper mining is discharged to environment. Copperbelt region has granites (uranium and pyrite), and Chambeshi is the largest open pit mine in the world. Underground and surface water analyses showed that most were below the WHO screening level concentration (gross alpha 0.5 Bq/L), and estimated doses were well below the screening dose of 0.1 mSv/a. An estimate of the dose from the highest concentration observed at Chambeshi was 20 μ Sv/a.

3.10. Contamination at the legacy site of a rare earths plant: A case study

A former rare earths plant known as YL used to be a very large rare earth smelting and processing plant and had serious pollution with high levels of ^{226}Ra and ^{232}Th . The MEP initiated decommissioning in 2009, completed in 2010 for unrestricted future use. Eighty buildings and structures and waste piles were cleaned up. A contamination map showed exposure rates up to 800 μ Sv/h indoors, a total area of about 30 600 m² was contaminated, and radioactivity levels were higher close to the finished product area. Disposals were in a municipal landfill, an engineered near surface facility and a small amount in a deep engineered facility. Source term was complicated. Other features included:

- Assumed vertical contamination was consistent with surface expression.
- Radon was used to help to identify subsurface contamination.
- Radioactive waste was classified by activity but was onerous for analysis.
- Classifying expedited by dose rate.
- Improved regulatory mechanisms.
- Proper education.

3.11. ^{222}Rn at a groundwater treatment plant

Radon was measured in a ground water treatment plant (minimum of 30 Bq/m³, highest 2000 Bq/m³). The dose rate was also measured in the plant. A hall with open gravity fed sand filters, iron, manganese added for radionuclide removal are the features of the treatment. Radium-226 concentration was quite stable at 0.186 and 0.072 Bq/L after treatment. The assumption was radon in groundwater at about 6 Bq/L released to air via aeration and estimated radon was greater than measured values, at an average of about 140 Bq/m³. Linear relation

between dose rate and radon observed. A short term (three day) relationship was observed between ^{222}Rn concentration in the air and the accumulation of ^{226}Ra in the filters.

4. CONCLUSIONS

There has been some progress in the harmonization of standards and regulatory approaches for exposure to NORM. However, the new BSS requirements pose additional challenges in applying the graded approach to regulation that is commensurate with the magnitude and likelihood of exposures. This is especially true in the case of radon and progeny exposures considering the newer approach of the ICRP and the new reference level concepts in the BSS. Natural radionuclides incorporated in building materials also need careful consideration.

Non-uranium mines in China involve a large workforce in millions and control of exposures to NORM is a challenge. Environmental contamination due to NORM at many sites was reported at the symposium. In most cases, exposure to workers and public is below 1 mSv/a.

**SITE SPECIFIC MEASUREMENTS, DOSE ASSESSMENT
AND APPLICATION OF THE ALARA PRINCIPLE**

(Topical Session 4)

Chairpersons

P.V. SHAW
United Kingdom

LING CHEN
China

V.M.F. JACOMINO
Brazil

QIFAN WU
China

Invited Paper

EXPOSURE ASSESSMENT OF WORKERS HANDLING INDUSTRIAL NORM IN JAPAN

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Abstract

In the study, a dose assessment of workers handling typical industrial NORM was carried out to obtain information for the future regulation system in Japan. The annual effective dose received by workers was estimated using measurements of dose rate and activity concentrations in raw materials, products and aerosols, as well as concentrations of ^{222}Rn and ^{220}Rn decay products in workplaces at plants processing zircon, monazite and titanium ore in Japan. From the results of the dose assessment, a relationship between the concentration of NORM and the average annual dose received by the worker was discussed.

1. INTRODUCTION

Japan is deficient in natural resources, and most mines, other than those for limestone, were closed more than 20 years ago. Thus, the exposure of workers who deal with various types of industrial NORM becomes a significant issue. Most of the materials are currently not subject to strict regulation in terms of the legal system. Only NORM containing high levels of uranium or thorium are subject to the law for the Regulation of Nuclear Source Material, Nuclear Fuel Material and Reactors. The main purpose of this law is to prevent nuclear disasters and to ensure security of the material. A tentative guideline was developed by the regulatory body for industrial materials containing relatively low concentrations of radionuclides of natural origin. The guideline does not define the activity concentration criteria for control but designates the types of materials which are to be subject to control. Information on dose is not yet

sufficient to establish a regulatory system. In this study, a dose assessment for workers handling industrial NORM was carried out to obtain information for the future regulatory system.

2. INDUSTRIAL USE OF NORM AND REGULATION IN JAPAN

Table 1 shows information on the use of NORM as an industrial raw material. These data were obtained from a database developed by the National Institute of Radiological Sciences [1, 2] and the report Exemption of Naturally Occurring Radioactive Materials by the Administrative Group of the Radiation Council of the Japanese Government [3]. The legal framework for NORM regulation in Japan is not yet established. Regarding nuclear fuel material, the exemption level for notification of use of nuclear source material is as follows:

- (a) Radioactive concentration: 74 Bq/g (liquid or gas) or 370 Bq/g (solid);
- (b) Sum of the amounts of thorium and three times of the amount of uranium: 900 g.

TABLE 1. INDUSTRIAL RAW MATERIALS WHICH COULD BE SUBJECT TO REGULATORY CONTROL AS NORM

	Amount imported (t/a)	Typical activity concentration (Bq/g)		End product	Approximate av. worker dose (mSv/a)
		U-238	Th-232		
Monazite	Several tens (in the past)	17–60	100–500	Consumer goods	0.3
Zircon	78 000 (in 2005)	1.8–3.9	0.37–1.9	Refractory	0.14
Bastnäsite	2 000 (in 2003)	1.1	5.8	Abrasive	0.4
Titanium ore	510 000 (in 2005)	0.05–0.56	0.04–0.57	Titanium	0.000 7–0.27
Phosphate rock	770 000 (in 2005)	0.1–1.5	0.006–0.046	Fertilizer	0.28

The concept of exemption level is different from that defined by the International Commission on Radiological Protection because exemption criteria are stipulated in the law for the Regulation of Nuclear Source Material, Nuclear Fuel Material and Reactors, the main purpose of which is to prevent disasters, and their levels are not related to dose criteria from the point of view of radiation protection of workers or the public. Regarding NORM regulation, a guideline was developed in 2009. In the guideline, manufacturers and importers are required to conduct self-management of the materials. The requirements of the guideline include the measurement of dose rate at 1 m from raw materials or products and the evaluation of the annual doses received by workers and local residents or users of the products. If the dose exceeds 1 mSv/a, measures to control exposure are required.

3. DOSE ASSESSMENT OF WORKERS HANDLING INDUSTRIAL NORM

3.1. Method

An investigation of the exposure of workers in NORM industries was carried out in three types of facility: those manufacturing refractories from zircon, those using monazite and those using titanium ore [4, 5]. Workers were categorized into groups with similar patterns of work in various workplaces. The annual effective dose for each group of workers was estimated using exposure scenarios and parameters from Ref. [6], the results of dose rate measurements at various locations in the workplace, and activity concentration measurements of radionuclides of natural origin including ^{222}Rn , ^{220}Rn and their decay products.

3.2. Results

The results of the dose assessment are shown in Table 2.

4. RESULTS AND DISCUSSION

The doses estimated in this study were found to be less than 1 mSv/a except for a case where monazite was used without the use of a mask. The concentrations of ^{232}Th and ^{238}U in one raw material (monazite) were about 340 and 38 Bq/g, respectively. It is stated in IAEA Safety Standards Series No. RS-G-1.7, Application of the Concepts of Exclusion, Exemption and Clearance [7], that doses to individuals as a consequence of an activity concentration of 1 Bq/g for

TABLE 2. SUMMARY OF DOSE ASSESSMENT RESULTS FOR WORKERS

	Type of plant	Annual effective dose (mSv/a)		
		External exposure only	External and internal exposure	
			Using mask	Without mask
Zircon	Manufacture of unshaped refractory	0.000 045–0.000 48	0.000 079–0.000 59	0.000 079–0.000 8
	Manufacture of shaped refractory	0.002 2–0.093	0.002 9–0.18	0.002 9–0.19
	Manufacture of electrocast refractory	0.026–0.28	0.045–0.77	0.046–0.77
Monazite	Manufacture of consumer products	0.001 3–0.43	0.004 7–0.8	0.013–1.6
Titanium ore	Storage of residue	0.000 6	0.000 72	0.000 72

radionuclides of natural origin would be unlikely to exceed about 1 mSv in a year, excluding the contribution from the emanation of radon. The effective doses estimated in this study include the contributions from radionuclides in the ^{238}U and ^{232}Th series and ^{40}K and from the inhalation of ^{222}Rn , ^{220}Rn and their decay products. Even where the concentrations of ^{238}U and ^{232}Th exceed 1 Bq/g, the annual effective dose would not exceed 1 mSv/a in most cases.

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Invited Paper

RADIONUCLIDE RELEASE FROM THE COMBUSTION OF COAL: A CASE STUDY

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Abstract

Under certain geological conditions, uranium and other metallic elements are absorbed into coal or even mineralized to form coal–uranium–polymetallic deposits. The coal–uranium–polymetallic ores are generally burnt in blast furnaces to produce metal concentrates from which metals are subsequently extracted (e.g. by smelting). This investigation was carried out in a plant located south-west of Kunming. The coal is enriched in uranium and rare earth elements. After its combustion at a temperature of more than 1000°C, the rare earth content of the fly ash removed from the bag filters was 2.32% compared with 0.053% in the original coal. Radioactivity in the coal becomes concentrated in the ash: some in the bottom ash and some in the fly ash. The rest is released to the atmosphere. In 2010, the activity released to the atmosphere from the plant was 15.5 MBq for ²³⁸U, 11.7 MBq for ²²⁶Ra, 41.4 MBq for ²¹⁰Pb and 50.7 MBq for ²¹⁰Po. The release of radionuclides in the off-gas was much greater than the amount contained in the fly ash. As a result, the level of radioactivity in the environment was enhanced.

1. INTRODUCTION

Coal contains radionuclides of natural origin at low concentrations. The typical concentrations in coal are 0.010–0.025 Bq/g for both ^{238}U series and ^{232}Th series radionuclides [1]. During combustion, the radionuclides become concentrated in the ash, but large amounts are discharged into the environment, resulting in enhanced exposure of the public [1–3]. Figures 1 and 2 show the concentrations of radionuclides in most of the coal in China [4]. The concentrations widely vary. In most coal, the concentrations are less than 0.1 Bq/g but about 6% exceeds 0.5 Bq/g for ^{238}U and ^{226}Ra and 0.2 Bq/g for ^{232}Th . In some areas, coal, uranium and other elements occur together. For example, coal–uranium mines exist in Xinjiang and Yunnan.

China is the world’s largest coal producer and the largest coal consumer. The total consumption of coal was about 3220 Mt in 2010. Most of it was used

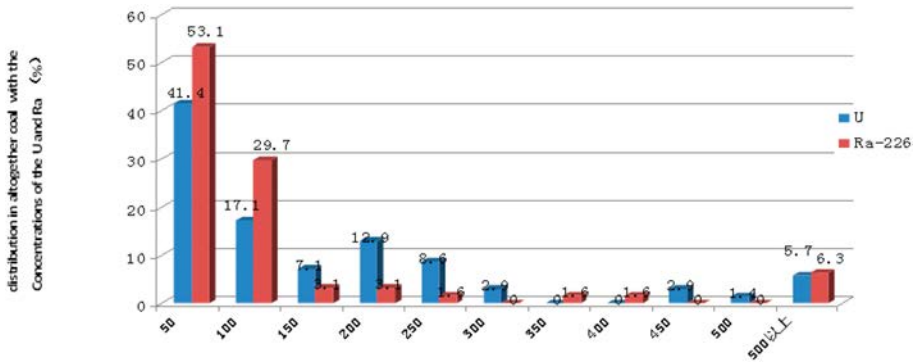


FIG. 1. Uranium-238 and ^{226}Ra concentrations in coal produced in China (Bq/kg).

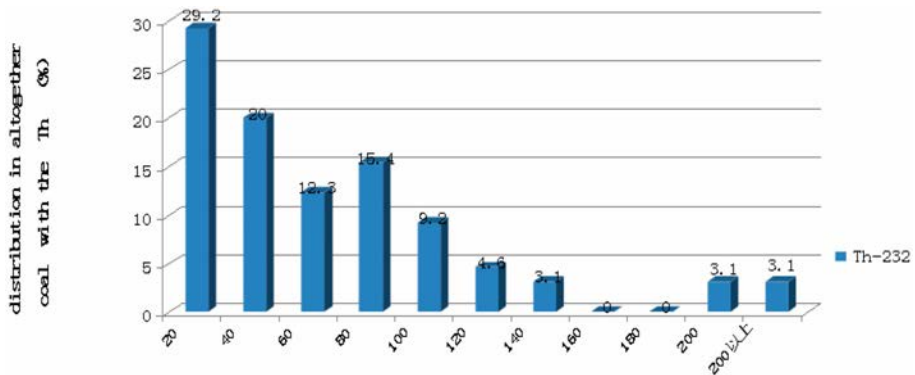


FIG. 2. Thorium-232 concentrations in coal produced in China (Bq/kg).

to generate electricity. Some coal contains elevated concentrations of uranium and other metallic elements and is used as a feedstock for metallic element extraction [5]. Coal fired power plants, with their vast coal consumption, are a major source of activity released to the environment and thus contribute significantly to enhanced levels of activity in the environment. The radiological impact on workers, members of the public, and the environment from coal fired power plants has been assessed [6–11]. Fly ash removed from the bag filters contains accumulations of metallic elements. The amount of activity released to the atmosphere is based on the amounts of particles and gases that pass through the filters and electrostatic precipitators. This study focuses on the radioactivity released from coal combustion, based on a mass balance in the process of extraction of rare metal elements.

2. RARE EARTHS PRODUCTION PROCESS

The plant that was the subject of this investigation is located in Lincang, south-west of Kunming. Coal in this area is enriched in uranium, but thorium and potassium are at normal background levels. There are some tens of coal mining sites in this area which, in 2010, produced about 100 000 t of coal as a source of rare earths concentrates and a further 100 000 t as bunker coal for domestic and industrial use. Table 1 shows the uranium concentrations in coal from 11 coal mining sites in the Lincang Basin. Generally, the coal with elevated uranium concentrations also has elevated rare earth element concentrations — this is illustrated by the fact that coal mines CJ, DZ and MW in Table 1 produce coal for rare earths extraction and their uranium concentrations exceed 1 Bq/g.

The plants for the production of rare earths concentrate from coal are located near the mine sites. A diagram of the process is shown in Fig. 3. Coal from underground is transported by conveyer belt to the plant. After crushing, the coal is burnt in furnaces. During combustion, much of the carbon and other combustible components leave the furnace as off-gas, while non-combustible matter such as clay minerals and some unburnt coal remain in the ash. The coarser, heavier ash falls to the bottom of the furnace as bottom ash. The finer, lighter ash (fly ash) escapes with the off-gas. The off-gas and fly ash are driven by a fan through snaked pipes for cooling and then directed to the dust collection house where about 99% of the fly ash is removed by bag filters. The gaseous components and the remnants of the fly ash are carried by the gas stream to the stack and discharged to the atmosphere after desulphurization. The rare earth content of the coal (0.053%) accumulates in the fly ash to a concentration of 2.32%. The fly ash thus becomes a product in the form of a rare earth concentrate. Radionuclides also become concentrated in the fly ash.

TABLE 1. ACTIVITY CONCENTRATIONS IN COAL FROM THE LINCANG BASIN

Mine	Activity concentration (Bq/g)	
	U-238	Th-232
ZT	0.305 3	0.073 3
BL	0.208 0	0.135 9
MT	0.502 2	0.074 9
DBS	0.092 3	0.121 3
LDK	0.491 0	0.375 2
MTK	0.525 8	0.079 5
SZ	0.313 8	0.005 8
AK	0.518 4	0.015 5
CJ	4.960 8	0.156 5
DZ	2.052 4	0.084 6
MW	1.346 5	0.004 2

Source: See Ref. [5].

In the coal, uranium exists as two phases [6]:

- (a) The first phase comprises uranium that has been absorbed as uraninite (UO_2) and has become highly dispersed in the coal. During combustion at more than 1000°C , a strong physical and chemical reaction occurs. Coal reacts with air to produce carbon dioxide. The UO_2 reacts with oxygen to produce highly volatile uranium trioxide. Some of the uranium progeny such as ^{210}Pb and ^{210}Po are volatile elements, which react with air at high temperature to form oxides.

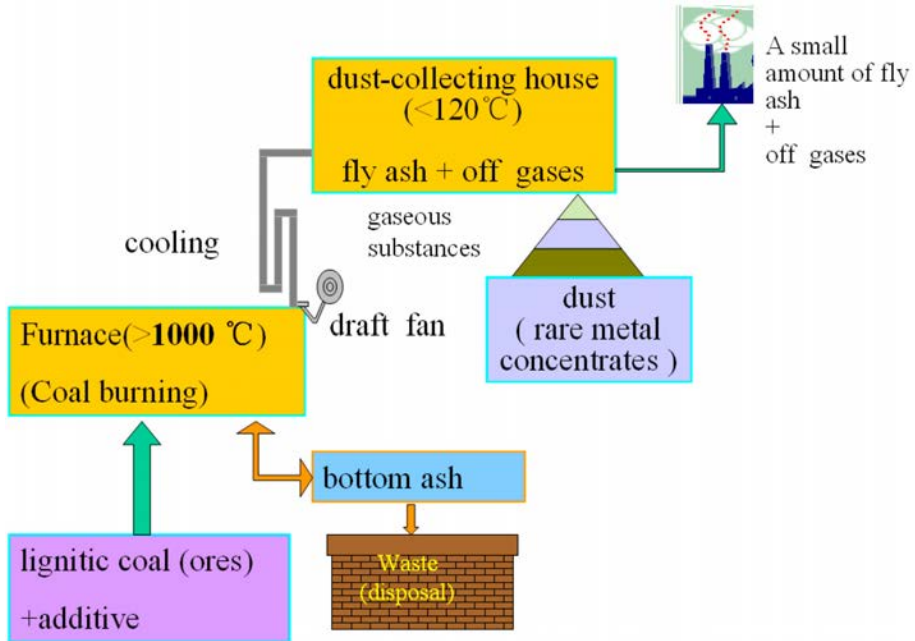


FIG. 3. Rare earth concentrate production.

- (b) The second phase comprises uranium distributed either in the crystal lattice of coffinite ($U(SiO_4)_{1-x}(OH)_{4x}$), which forms part of the clay content of the coal, or in aluminosilicate minerals. Thorium and potassium are also distributed in the aluminosilicate minerals. Uranium, thorium and their progeny, and potassium, remain in the ash but most of the ^{210}Pb and ^{210}Po in the clay or bottom ash escapes with the other gaseous substances in the furnace.

3. RELEASE OF RADIONUCLIDES

3.1. Radionuclide activity concentrations

Table 2 shows the activity concentrations of radionuclides and the enrichment factors (EFs) in the samples from the plant. The EFs are the activity concentrations in the ash relative to those in the coal. The activity concentrations of ^{232}Th and ^{40}K are at background levels and are insignificant in

terms of radiological impact. The activity concentrations of ^{238}U and its progeny (^{226}Ra , ^{210}Pb and ^{210}Po) in the coal feedstock are 1.14–1.20 Bq/g, with the radionuclides of the ^{238}U decay chain being approximately in equilibrium. The EFs for uranium and radium are about 1.3 in the bottom ash and about 4 in the fly ash. The volatile elements lead and polonium are depleted in the bottom ash, with EFs of 0.5 and 0.2, respectively, but are highly enriched in the fly ash, with EFs of about 37 and 21, respectively.

TABLE 2. ACTIVITY CONCENTRATIONS IN COAL AND ASH SAMPLES

	U-238	Ra-226	Pb-210	Po-210	Th-232	K-40
Activity concentration (Bq/g)						
Coal feedstock	1.20	1.14	1.17	1.14	0.035 3	0.307
Bottom ash	1.58	1.61	0.57	0.20	0.058 3	0.546
Fly ash	4.81	4.73	42.90	24.30	0.128	0.668
Enrichment factor						
Bottom ash	1.3	1.4	0.5	0.2	1.65	1.76
Fly ash	4.0	4.2	36.7	21.3	3.63	2.18

3.2. Release of radionuclides into the atmosphere

The furnaces in the plant had eight burners which, in 2010, consumed 50 000 t of coal and generated 24 t of fly ash product in the bag filters. The recovery of the fly ash was 99%. Potassium was deposited with the non-combustible materials which remained in the ash after combustion. Assuming the mass of potassium in the coal burnt was equal to the total inventory of potassium in the bottom ash and fly ash, an estimated 28 100 t of bottom ash was generated in 2010. Therefore, the radionuclide activity concentrations in the coal, bottom ash and fly ash can be calculated from the data in Table 2. The amount of activity of a given radionuclide in coal, bottom ash or fly ash is the product of the activity concentration and the mass of the coal, bottom ash or fly ash, respectively. The total activity of a radionuclide discharged to the atmosphere can be calculated from the radionuclide balance, as follows:

RADIONUCLIDE RELEASE FROM THE COMBUSTION OF COAL

$$\begin{aligned}
 A_d(j) &= A_c(j) - A_{ba}(j) - A_{bf}(j) \\
 &= AC_c(j) \cdot m_c - AC_{ba}(j) \cdot m_{ba} - AC_{bf}(j) \cdot m_{bf}
 \end{aligned}
 \tag{1}$$

where

$A_d(j)$, $A_c(j)$, $A_{ba}(j)$, $A_{bf}(j)$ are the total activities of radionuclide j in the discharged gas, coal, bottom ash and bag filter fly ash, respectively (Bq);

$AC_c(j)$, $AC_{ba}(j)$, $AC_{bf}(j)$ are the activity concentrations of radionuclide j in the coal, bottom ash and bag filter fly ash, respectively (Bq/g);

and m_c , m_{ba} , m_{bf} are the amounts of coal, bottom ash and bag filter fly ash, (50×10^9 , 28.1×10^9 and 24×10^6 g, respectively, in 2010).

Taking ^{238}U as an example, the total activity discharged during 2010 can be calculated from Eq. (1) by substituting the relevant activity concentrations from Table 2, giving:

$$\begin{aligned}
 A_d(^{238}\text{U}) &= (50 \times 10^9 \times 1.2) - (28.1 \times 10^9 \times 1.58) - (24 \times 10^6 \times 4.81) \\
 &= 15.5 \times 10^9 \text{ Bq}
 \end{aligned}
 \tag{2}$$

The total activities for 2010 of all the relevant radionuclides in the process material throughputs and in materials discharged to the atmosphere are given in Table 3. The total amounts of ^{238}U , ^{226}Ra , ^{210}Pb and ^{210}Po activity in the coal burnt in the plant during 2010 were each about 58×10^9 Bq. Of these, the amounts released to the atmosphere are 15.5×10^9 Bq for ^{238}U , 11.7×10^9 Bq for ^{226}Ra , 41.5×10^9 Bq for ^{210}Pb and 50.8×10^9 Bq for ^{210}Po . The 1% of the fly ash that escapes into the atmosphere accounts for only a very small fraction of the total activity released — the amounts range from 3.1×10^4 Bq for ^{232}Th to 1.04×10^7 for ^{210}Pb .

TABLE 3. RADIONUCLIDE ACTIVITIES IN THE PROCESS MATERIALS AND DISCHARGED TO THE ATMOSPHERE IN 2010

	U-238	Ra-226	Pb-210	Po-210	Th-232
Annual activity in process materials (10^9 Bq)					
Coal feedstock	60.0	57.0	58.5	57.0	1.77
Bottom ash	44.4	45.2	16.0	5.62	1.64
Fly ash	0.115	0.114	1.03	0.583	0.003 07
Annual activity discharged to the atmosphere (10^9 Bq)					
Fly ash only	0.001 17	0.001 15	0.010 4	0.005 89	0.000 031
Total	15.5	11.7	41.5	50.8	0.125
Proportion of total coal activity discharged (%)					
	25.8	20.5	70.9	89.1	7.1

4. CONCLUSIONS AND DISCUSSION

The activity concentrations of radionuclides in the coal, bottom ash and fly ash are higher than the exemption level. The combined activity of ^{238}U , ^{226}Ra , ^{210}Pb and ^{210}Po released to the atmosphere exceeds 10^{11} Bq annually. As a result, the activity concentrations of radionuclides determined from aerosol monitoring in the plant were ten times higher than the background levels near the plant. Radiation levels were enhanced because of the deposition of waste and the discharge of off-gas.

The result of this investigation is significantly different from results reported in the literature concerning radionuclides released from coal fired plants for electric power generation. The activity concentrations of radionuclides in coal used in coal fired power plants are mostly below 0.1 Bq/g, while the enrichment in the bottom ash is usually more than three times, and even higher in fly ash, especially for ^{210}Pb and ^{210}Po , which can be enriched by up to ten times. Some radionuclide release to the atmosphere is due to the incomplete removal of dust from the filters. Radionuclides are carried by the escape of fly ash from filters, which is less than 5% of the total fly ash. Radionuclides absorbed in the carbonaceous portion of the coal tend to be released as part of the off-gas and thus contribute significantly to the total amount of radionuclides released to the atmosphere. The activity released in the off-gas is much more than that remaining

in the fly ash. The results of this investigation show that the proportions of radionuclides in the coal that are released to the atmosphere are 26% for ^{238}U , 21% for ^{226}Ra , 71% for ^{210}Pb and 89% for ^{210}Po . The amounts released in the escaping fly ash are very small.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the Department of Nuclear Safety Management of the Ministry of Environmental Protection for financial support. They would like to thank Pan Ziqiang for his technical discussions. They also thank Ma Jizeng, P.P. Haridasan and Fan Zhiwen for their help and encouragement.

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OCCUPATIONAL EXPOSURE IN AMMONIUM PHOSPHATE FERTILIZER PLANTS

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Abstract

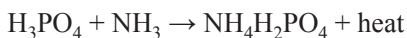
Occupational exposures and activity concentrations have been assessed in two industrial plants producing mono-ammonium phosphate and di-ammonium phosphate fertilizers, located in south-western Spain. The annual effective doses received by the workers are below 1 mSv/a, with the contribution from external exposure being similar to that from internal exposure. The dose contribution from inhalation of dust has been estimated to be about 0.12 mSv/a, while the ^{222}Rn concentrations inside the plants are of no concern. Consequently, no additional radiation protection measures need to be taken to protect the workers in these facilities.

1. INTRODUCTION

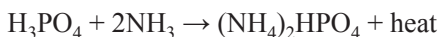
According to the Spanish regulation affecting NORM industries, assessments of occupational exposure should be performed in all plants producing phosphoric acid and derived phosphate fertilizers in order to determine the need for radiation protection measures to control the doses received by the workers. Most of the dosimetric studies performed in the phosphate industry in Spain have been focused on the determination of occupational exposures associated with the handling of phosphate rock and the generation of phosphoric acid, mostly because, at this stage of the production process, all the radionuclides in the uranium decay series, and originally present in the raw material, are involved. However, no similar studies on the production of ammonium phosphate fertilizers — mono-ammonium phosphate (MAP) and di-ammonium phosphate (DAP) — have been performed until now in Spain, and few data can be found in the open literature [1–3] concerning occupational exposures in this type of facility.

The phosphoric acid used as a raw material in ammonium phosphate fertilizer production is enriched in ^{238}U but is deficient in the progeny radionuclides ^{226}Ra , ^{210}Pb and ^{210}Po [4]. A dosimetric evaluation was carried out in one MAP plant and one DAP plant which form part of a fertilizer manufacturing complex in Huelva, Spain. External radiation and dust inhalation were considered to be the main exposure pathways to workers, but ^{222}Rn determinations were also performed in both plants.

In the MAP plant, the final product is generated by reacting phosphoric acid with ammonium until an $\text{NH}_3:\text{PO}_4$ molar ratio of 1 is reached:



In the DAP plant, the final product is generated by reacting phosphoric acid with ammonium until an $\text{NH}_3:\text{PO}_4$ molar ratio of 2 is reached:



2. MATERIALS AND METHODS

2.1. Gamma dose rate determination

External gamma dose rate determinations were performed using a calibrated Berthold LB1236 gas dosimeter coupled to an electronic unit LB1230. This system works under a proportional regime and has a working range of 0.01–10 000 $\mu\text{Sv/h}$. It is sensitive to photons with energies in the range of 30 keV–1.2 MeV. The energy dependence of the dosimeter response is about $\pm 20\%$ within the working energy range. The data readings obtained with this equipment were corrected for contributions arising from cosmic radiation and electronic noise.

2.2. Aerosol analysis

Aerosol sampling was performed 2 m above the floor level at a common representative place within each of the two plants using Andersen PM10 high volume samplers (flow rate 68 m^3/h) fitted with 25.4 cm \times 20.3 cm rectangular quartz microfibre filters. The sampling frequency was once in every 2 weeks, and the normal period of collection was 48 h.

Using the methodology described in Ref. [5], the mass activity concentrations of uranium isotopes, thorium isotopes and ^{210}Po in each filter were determined by alpha spectrometry. The mass activity concentrations of

^{210}Pb were also determined by gamma spectrometry, along with the concentrations of other gamma emitting radionuclides such as ^{226}Ra , ^{234}Th , ^{234}U , ^{228}Ra , ^{224}Ra , ^{228}Th and ^{40}K . The calibration efficiency of the gamma spectrometric system for atmospheric filters is described in Ref. [6].

The dust concentrations in the air were determined by the company using individual dust samplers worn by various representative workers.

2.3. Radon-222 determination

The volumetric activity concentrations of ^{222}Rn were determined by performing integrated measurements with CR-39 nuclear track detectors. The exposure time of each detector was 3 months. In each plant, detectors were placed at two sites characterized by their high worker occupancy, in order to obtain representative values and to determine the uniformity of the ^{222}Rn inside the facilities.

3. RESULTS AND DISCUSSION

3.1. Exposure to gamma radiation

A map of the gamma dose rate increments above background was created for each of the two plants surveyed. It was observed that the values were low and always in the range of 0–0.08 $\mu\text{Sv/h}$. These low values are to be expected when one considers that the raw materials and final products are enriched only in radionuclides which are mainly alpha emitters, and are depleted particularly in ^{226}Ra and its progeny, ^{232}Th and its progeny, and ^{40}K , which in NORM facilities represent the main contributors to external gamma radiation. The activity concentrations in the 54% phosphoric acid used for the production of MAP and DAP were as follows:

- 3 Bq/g for ^{234}U and ^{238}U ;
- 2 Bq/g for ^{230}Th ;
- <0.02 Bq/g for ^{226}Ra , ^{226}Ra progeny, ^{232}Th , ^{232}Th progeny and ^{40}K .

The doses received by workers in the MAP and DAP plants were determined from the gamma dose rate measurements using detailed occupancy information obtained from the plant operator. The results for various categories of worker are summarized in Table 1 and may be considered generally representative of plants of this type.

TABLE 1. DOSE FROM EXTERNAL EXPOSURE, BY WORKER CATEGORY

	Annual effective dose (mSv)
Control operator in MAP and DAP plants	0.134
Chief technician in MAP and DAP plants	0.087
Operator in MAP plant	0.090
Operator in DAP plant	0.076
Granulator operator in DAP plant	0.055

Note: DAP — di-ammonium phosphate; MAP — mono-ammonium phosphate.

3.2. Exposure to airborne dust

The results of the radiometric analyses performed on the aerosol filters are given in Table 2. The concentrations of ^{238}U series radionuclides were found to be elevated above the levels determined in a representative background area, while the concentrations of ^{232}Th series radionuclides and ^{40}K were generally similar to background levels. The ^{238}U series radionuclides with long half-life (^{234}U , ^{230}Th and ^{226}Ra) were found to be in approximate equilibrium with ^{238}U . It was important to note in this regard that, even though the activity concentrations of ^{226}Ra and its progeny in the phosphoric acid feedstock were more than two orders of magnitude lower than those of ^{238}U and ^{234}U , the potential for generating airborne MAP and DAP dust particles was very limited since MAP is produced as a powder with a moisture content of about 5% and DAP is produced as granules. The contributions of MAP and DAP particles to the airborne dust inside the plants were therefore insignificant, with most of the dust originating from the handling, milling and transport of phosphate rock in the neighbouring phosphoric acid plants.

For purposes of dose assessment, the activity concentration of each radionuclide in the ^{238}U decay series was assumed to be 0.67 Bq/g, the average of the measured ^{238}U concentrations. This value was multiplied by the measured dust concentration to determine the activity concentration in the air breathed by the worker. The committed effective dose due to dust inhalation was then determined according to the methodology described in Ref. [5]. The dust concentrations in air and the calculated doses for selected worker categories are shown in Table 3. For all workers, the doses are less than 0.11 mSv.

OCCUPATIONAL EXPOSURE IN AMMONIUM PHOSPHATE FERTILIZER PLANTS

TABLE 2. DUST ACTIVITY CONCENTRATIONS

Sample number	Activity concentration (Bq/g)			
	U-238		Th-232	
	Mean	Standard deviation	Mean	Standard deviation
1	0.090	0.023	0.032	0.004
2	0.830	0.090	0.036	0.004
3	0.580	0.060	0.020	0.003
4	0.180	0.030	0.010	0.003
5	0.880	0.100	0.036	0.007
6	1.160	0.110	0.026	0.003
7	0.430	0.050	0.079	0.007
8	0.990	0.100	0.230	0.030
9	0.970	0.090	0.024	0.003
10	0.760	0.060	0.053	0.006

TABLE 3. DUST CONCENTRATION IN AIR BREATHED BY THE WORKER AND THE RESULTING INHALATION DOSE, BY WORKER CATEGORY

	Av. dust concentration (mg/m ³)	Inhalation dose (mSv/a)
Operator in MAP and DAP plants	2.175	0.107
Control operator in MAP and DAP plants	0.955	0.047
Granulator operator of in DAP plant	1.300	0.054
Chief technician in MAP and DAP plants	2.080	0.103

Note: DAP — di-ammonium phosphate; MAP — mono-ammonium phosphate.

3.3. Exposure to ^{222}Rn

The mean ^{222}Rn concentrations at the two sampling locations in the MAP plant were 18 and 16 Bq/m³ while the corresponding values in the DAP plant were 15 and 18 Bq/m³. In all cases, the standard deviation was 3 Bq/m³. These levels are typical of normal indoor concentrations and far below the 400 Bq/m³ reference level for workplaces [7, 8]. This result is not surprising, since the ^{226}Ra content of the feedstocks and products is less than that of normal soil and the plants are well ventilated. Exposure to ^{222}Rn is thus of no radiological concern.

4. CONCLUSIONS

Exposure to gamma radiation and airborne dust are the main contributors to the dose received by workers in the MAP and DAP plants, with no significant contribution from ^{222}Rn . The dose is in all cases less than 0.3 mSv/a. No specific radiation protection measures need to be taken in order protect the health of the workers.

ACKNOWLEDGEMENTS

The authors are very grateful to the staff of the Fertiberia company in Huelva for all the help and support given in the performance of this work.

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REMEDICATION OF A NORM CONTAMINATED SITE IN AUSTRIA

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Abstract

The paper describes the remediation of a NORM contaminated site in the Austrian province of Lower Austria. In 2007, it was planned to erect a school in the vicinity of the site. As result of reports in the literature about historical activities involving ^{226}Ra at the site and after a small contaminated area was detected and remediated, the Austrian Agency for Health and Food Safety was engaged to ensure that no further NORM contamination would harm the people in this area. The site history indicated that between 1910 and 1917 a chemical plant produced radium containing products such as oils and salts for bathing. The paper provides a description of the strategy to control the area, the measurement methods used and the results and consequences of those measurements. Details are given of the detection of subsurface contamination, on-site measurements on a large scale, the waste acceptance criteria established for various disposal options and the methodology to derive them, as well as the regulatory framework, amounts of NORM waste disposed of by various routes and unexpected difficulties encountered during the remedial work.

1. INTRODUCTION

In 2007, the Austrian Agency for Health and Food Safety (AGES, Österreichische Agentur für Gesundheit und Ernährungssicherheit) was instructed to conduct minor remediation of a contaminated site in the Austrian province of Lower Austria and subsequently to examine the site with respect to any radiological hazards, including the measurement of dose rate, soil activity concentration and radon. An investigation of the history of the site showed that between 1910 and 1917 a chemical plant was producing beauty products containing radium. Between 1917 and 1925, the chemical plant produced further products until closure of the plant. It is unclear as to whether radium continued to be used in the process or not. Between 1925 and 2007, records show that the site was used as a textile factory and later as a municipal workshop. Challenges included the existence of a time constraint because of the planned erection of a school in 2009 and the need to optimize the costs of comprehensive remediation of the area.

2. INITIAL SITUATION, FIRST RESULTS AND REMEDIAL ACTION TAKEN DURING 2008

In 2007, the first investigations of the site took place as a result of information relating to the early use of the site [1]. After a short survey of the site, a small area contaminated with ^{226}Ra (area 2 m^2 , depth 20 cm) was detected and remediated by Nuclear Engineering Seibersdorf in 2007. Consequently, AGES was contracted as an independent body to control the remediated area between an old villa and the workshop by performing a radiological examination of the site, including measurements of dose rate, soil activity concentration and radon. In March 2008, an AGES team of four investigated the buildings and the agricultural field over a four day period. First the offices and group room were investigated by surveying with a dose rate meter (device type FH 40 G) with an output in nanosieverts per hour (nSv/h) and a contamination monitor (device type FHT 111 M) with an output in counts per second (cps). The measurements identified slightly elevated radiation levels (160–320 nSv/h and 40–50 cps). The workshops (carpentry, metal workshop and a forge) were investigated in the same way. From these investigations, six local and superficial hot spots (190–800 nSv/h and 200–800 cps) were detected, sampled and marked. No suspicious measurement results were detected in the old villa. After that, short term radon measurement devices (PicoRad LS vials) were installed in the offices, group room and workshops for about 48–72 h. Finally, the agricultural field was investigated in a grid of $10\text{ m} \times 10\text{ m}$ at 1 m height using dose rate monitors and contamination monitors. On the strength of the results of approximately 185 dose rate and count rate measurements, 35 soil samples were identified and removed for analysis. The dose rates were 120–250 nSv/h and the count rates were 15–23 cps. A short summary of the first evaluation results of the site with an overview of the site and the location of the buildings is given in Fig. 1.

The preliminary results of the sampled hotspots showed, as expected, high ^{226}Ra activity concentrations of up to 40 Bq/g. In contrast, the results of the soil samples from the agricultural field were quite normal with ^{226}Ra activity concentrations of about 0.03 Bq/g. Concerns arose after the average radon concentration results became available: 100–2000 Bq/m³ in the storage facilities; 200–5000 Bq/m³ in the workshops; and 12 000–20 500 Bq/m³ in the offices and group rooms. As a first consequence, a rough dose assessment based on the short term radon measurements was done, which led to a decision to move the municipal workers into container accommodation. Next, detailed radon measurements with an AlphaGuard radon monitoring instrument were performed, which led to a detailed dose assessment based on occupancy and radon concentration. The radon inhalation doses assessed for two office worker were in the same range as those

REMEDIATION OF A NORM CONTAMINATED SITE IN AUSTRIA

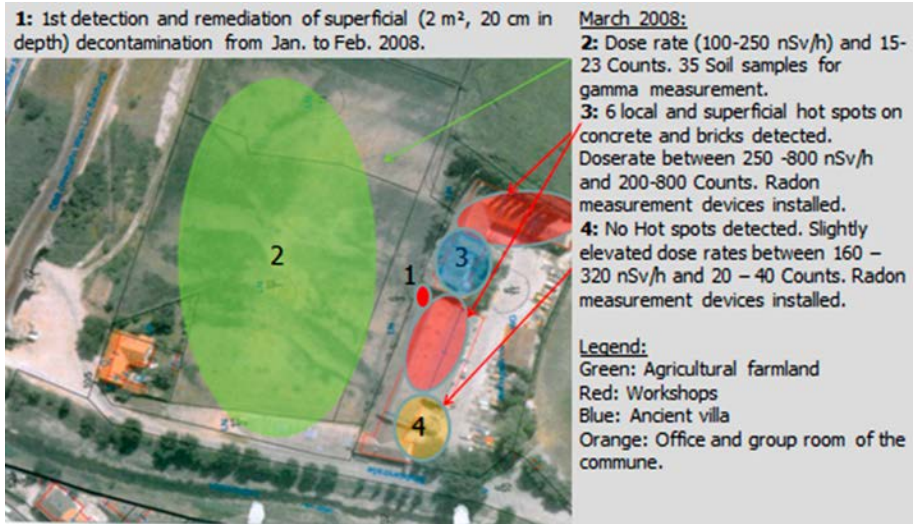


FIG. 1. Aerial photograph of the site and first evaluation results.

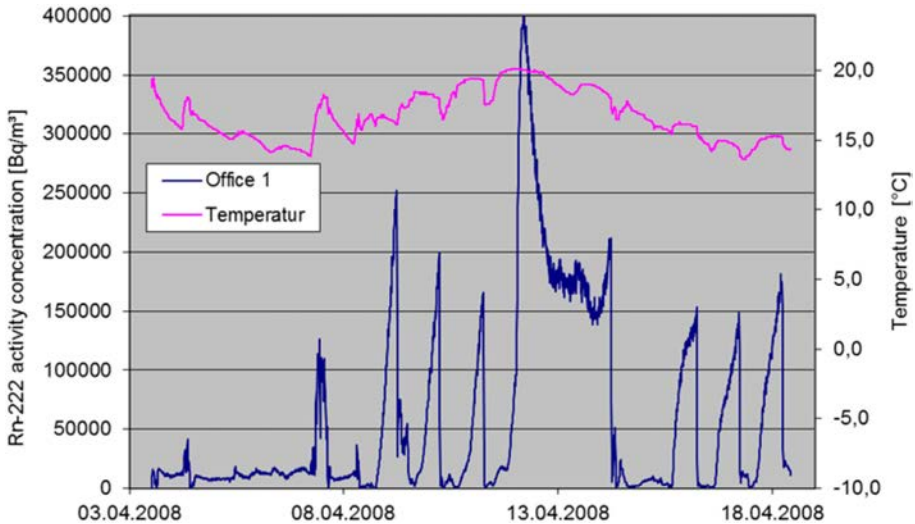


FIG. 2. Radon activity concentration in one of the offices.

for a ‘Category A worker’ (i.e. 6–20 mSv/a). A typical portion of a continuous radon monitoring result is given in Fig. 2.

On the basis of these initial results, new objectives for the year 2008 were considered. A plan for the demolition of the non-contaminated parts of the building and for the remediation of the six hotspots was established. The demolition plan consisted of fencing of the area, on-site measurements to identify the presence of radioactive material, instruction of the demolition workers on radioactivity, radium and health and safety measures, and surveillance of the workers with aerosol measurements and personal dosimetry. The observance of these measures was performed by an independent radiation protection officer. A view of the demolition of the non-contaminated parts of the building is shown in Fig. 3. For the remediation of the hotspots, a characterization of the depth and the activity concentration was performed using core drilling. The vertical distribution of the ^{226}Ra activity concentration is shown in Fig. 4. The remediation of a hotspot with simple measures such as hand digging, percussion drilling and vacuum cleaning is shown in Fig. 5. All these activities were done between the summer and autumn of 2008. After an analysis of the results of actions taken up to that point and after the remediation of the hotspots, it was clear that the main source of the high radon activity concentration had still not been found. In winter 2008, all activities on site were stopped and new objectives for the year 2009 were defined.



FIG. 3. Demolition of the non-contaminated parts of the building.

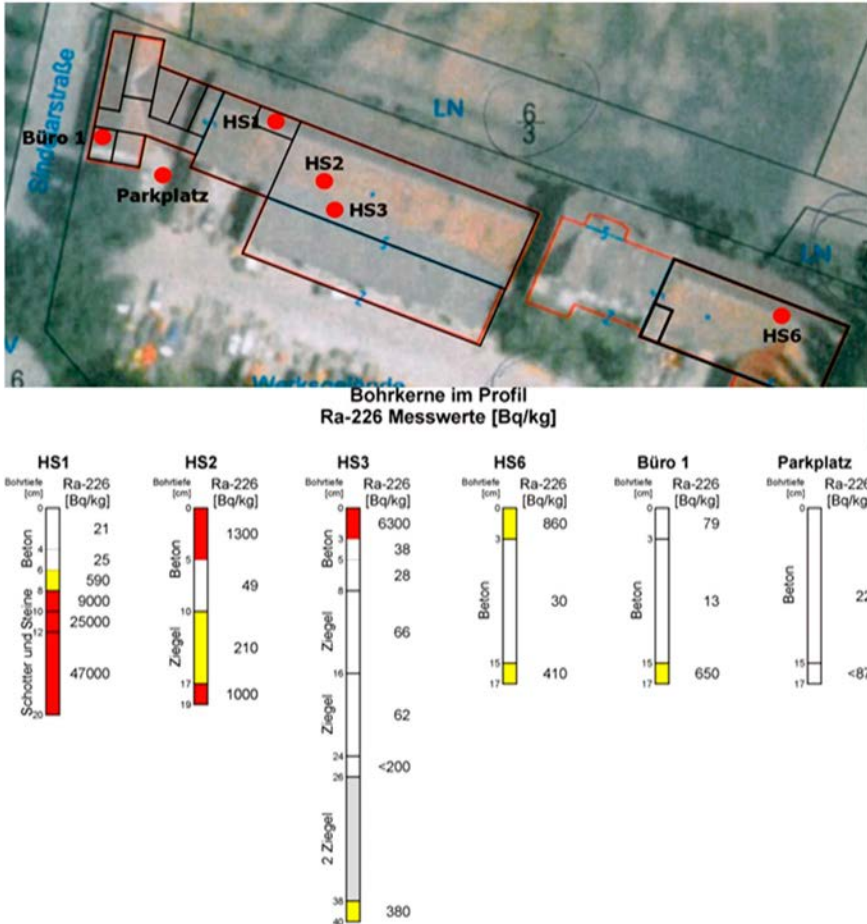


FIG. 4. Vertical distribution of the ^{226}Ra contamination at the six hotspots.

3. OBJECTIVES, RESULTS AND REMEDIAL ACTION TAKEN DURING 2009

3.1. Objectives

The objectives for 2009 were:

- (a) Detection of the subsurface contamination, particularly around the office buildings;
- (b) Characterization of the expected contaminated materials;
- (c) Estimation of the expected amount of radioactive waste;



FIG. 5. Remediation of a hotspot.

- (d) On-site measurements during the remedial work using a high purity germanium (HPGe) detector and in situ measurements of the barrels filled with contaminated soil.

3.2. Measurement results

During the spring of 2009, core drilling to a depth of up to 4 m was performed on the whole site. The drilling profiles in terms of ^{226}Ra activity concentrations around the office building are shown in Fig. 6. It was remarkable to find that almost no contamination under the office buildings was identified by the radon measurements and that the main contamination was situated outdoors in front of the offices. From the results of the core drilling and after the first excavation it became clear that the expected amount of radioactive waste would not be in line with prior estimations and the objectives for 2009 therefore had to be adapted to the new situation.

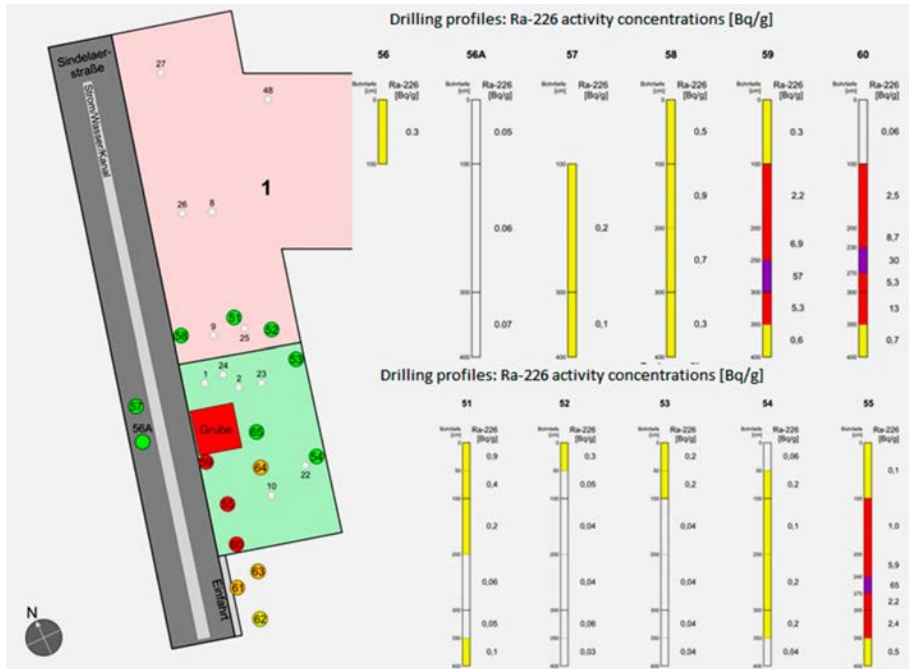


FIG. 6. Drilling profiles around the office buildings.

3.3. Remedial action

The excavation work was performed by two or three workers wearing protective suits. One independent expert from AGES surveyed the work on site and performed random control measurements with a contamination monitor to check roughly that only contaminated soil was put into the barrels. On any one day, some 10–20 barrels were filled and the barrels from the day before were measured for about 15 min on a rotating platform with an ISOCS calibrated HPGe detector. The filled barrels were temporarily stored and measured in a building still remaining on site. Views of the excavation work are shown in Fig. 7, while Fig. 8 shows the excavated pit during final inspection. The car park eventually constructed on the site and the adjacent school are shown in Fig. 9.



FIG. 7. The excavation work: (a) early stages and (b) highly contaminated zone.



FIG. 8. The excavated pit during final inspection.

3.4. Waste acceptance criteria

After several days, the data from the barrel measurements and a chemical description of the subsurface contamination were sufficient for making a start on the process of defining waste acceptance criteria for various disposal options such as restricted release to a controlled disposal site and retention of contaminated



FIG. 9. Car park and adjacent secondary school in 2010.

soil on site. As a first step, the consequence of possible leaching and transfer into groundwater was determined by means of a simulation based on the RESRAD code [2]. For this purpose, scaled aerial photography of the remediated site and the future disposal site were necessary. Several future exposure scenarios, such as farm land, a school, dwellings and a freshwater well, were analysed using conservative assumptions on occupancy periods and rates of inhalation and ingestion. All results showed that leaching into the groundwater was the most important exposure pathway. Depending on consumer behaviour, the ingestion pathway could also be a significant pathway, but to a much lesser extent. The inhalation of dust was not included in the assessment because it was only relevant during the remediation work. The inhalation of radon was also excluded because the future uses of both the remediated site and the waste disposal site were restricted. The final waste acceptance criteria established on the basis of the simulations resulted in the following waste management routes:

- (a) Two hundred barrels, each containing 150–200 kg of soil contaminated with ^{226}Ra and its progeny at activity concentrations of 8–500 Bq/g, were sent to a controlled storage facility for radioactive waste.
- (b) About 2000 t of soil contaminated with ^{226}Ra and its progeny at activity concentrations of about 7 Bq/g or less (and contaminated also with non-radioactive chemically hazardous material) was sent for disposal at a controlled waste repository.
- (c) Soil contaminated with ^{226}Ra and its progeny at activity concentrations of 2 Bq/g or less remained on site, covered by 2–4 m of uncontaminated soil, and its presence was recorded in the cadastral register.

3.5. Regulatory framework

This initiative was based on the Radiation Protection Law §26 – Orphan Radioactive Sources and Radioactively Contaminated Sites.¹ The remediation of the small hotspots and the main areas of contamination was regulated in terms of the Radiation Protection Law §36 (b) and (c). The displacement of the municipal workers into container accommodation was done on the basis of the Radiation Protection Law §36 l – Scope for the Execution of Interventions; Ordinance for Intervention.² The surveillance of the workers was based on the Radiation Protection Law §24. Disposal of the moderately contaminated material at the controlled waste disposal site was based on the Radiation Protection Law §36 (h) and NORM Ordinance.³

4. CONCLUSIONS AND LESSON LEARNED

In the initial site survey, the use of contamination monitors proved to be more effective than the use of dose rate meters because the readings of most dose rate meters were compromised by the count statistics. Once contamination had been found, a dose rate meter reading was necessary in order to satisfy and check against legal requirements. When ²²⁶Ra and its progeny are present in the contamination, radon measurement is always a good choice. The definition of waste acceptance criteria for various disposal options is to be considered in order to avoid very large amounts of waste having to be sent to a temporary radioactive waste repository. The use of an established software package for the simulation of exposure scenarios was helpful in identifying the most likely exposure pathways and in determining final waste acceptance criteria. After completion of the remedial work, environmental monitoring of the sediments of a nearby river and of well water on site was performed.

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² Interventionsverordnung – IntV, BGBl. II Nr. 145/2007.

³ Natürliche Strahlenquellen-Verordnung – NatStrV, BGBl. II Nr. 2/2008.

MOBILE UNIT FOR SITE CHARACTERIZATION IN ENVIRONMENTAL REMEDIATION PROJECTS

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Abstract

The determination of the extent of contamination and contaminant distribution, among other things, in areas undergoing remediation forms part of an environmental remediation plan. Traditionally, this activity involves the collection of different environmental samples and laboratory analysis of the relevant radionuclides (and eventually other contaminants such as heavy metals). When the results are available they are interpreted and then a decision is made. This process is normally very expensive and time consuming. In recent years, many techniques have been made available for in situ measurement that can provide reliable information on the contamination profile of the contaminated area. Such measurements tend to be less expensive and more rapid and, with the aid of GPS/GIS systems, decisions can be made immediately on site. Mobile units may also be useful to States that have laboratory analysis facilities but are faced with large, unforeseen characterization challenges such as those following an accident or radiological emergency. To cater for this situation, data acquisition and control module (DACM) technology was developed. Instruments based on this technology can be modified by the user at any time without special knowledge. DACM technology offers a set of components which can be configured, parameterized and controlled to suit the requirements on site. Typical components include: radon–thoron modules (soil gas, water, air, exhalation and contiguous flux measurement at different depths); signal inputs for sensors for carbon dioxide, methane and sulphur dioxide; control outputs for pumps, magnetic valves for exhalation measurements and also complex functional blocks such as spectrometers; a GPS receiver; and PID regulators. A complex sampling schedule can be created within minutes by a graphical software interface. With dimensions of 235 mm × 140 mm × 255 mm and a mass of less than 6 kg, the full system is very handy.

1. INTRODUCTION

As part of an environmental remediation plan to be applied to areas affected by past activities and accidents, the characterization of the site is a mandatory step. This activity will determine the extent of the contamination and the contaminant distribution, among other things [1], and involves the collection of various environmental samples and the laboratory analysis of the relevant radionuclides (and eventually other contaminants such as heavy metals). When

the results are available they are interpreted and then a decision is made. This process is normally very expensive and time consuming. In recent years, many techniques have been made available for in situ measurement that can provide reliable information on the contamination profile of contaminated land. Such measurements tend to be less expensive and more rapid and, with the aid of GPS/GIS systems, decisions can be made immediately on site. Many States facing the challenge of implementing environmental remediation projects do not have an adequate analytical infrastructure for site characterization. They will need to first establish such an infrastructure before carrying out site characterization work. Therefore, remedial work will be delayed or not implemented at all. Mobile units may also be useful to States that do have laboratory analysis facilities but are faced with large, unforeseen characterization challenges such as those following an accident or radiological emergency.

2. DATA ACQUISITION AND CONTROL MODULE SYSTEM TECHNOLOGY

Data Acquisition and Control Module (DACM) technology offers a universal platform for instrumentation which can be adapted to any application without changes in the hardware or special knowledge in computer programming. Compared with conventional data acquisition systems, DACM is able to control complex sampling procedures in addition to data logging and management. Scalability and portability make the DACM architecture a universal and future oriented system. The system performance can be expanded or specialized in any order while the basic structure of system control and data flow is not touched. The user is able to subsequently modify all instrument functions with respect to the user's own requirements.

Each DACM based instrument contains a system core and a set of so called 'components' (see Figs 1–3). The system core includes the basic instrument properties such as device control, data storage, interfacing, component management and sampling control. The components implemented in the DACM can be considered as a library of functional units which can be manipulated to adapt them to the user's requirements:

- (a) Each component can be specifically configured (e.g. transfer function and operation mode).
- (b) The state of a component can be changed during the sampling cycle.
- (c) Components can generate, display and store measurement data.
- (d) Components can generate alerts or react to alert states.

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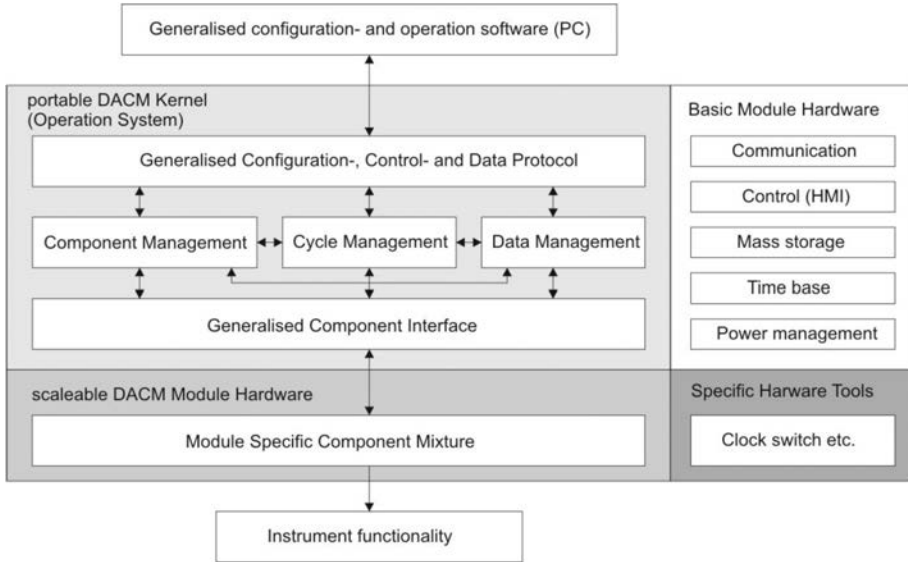


FIG. 1. DACM topology.

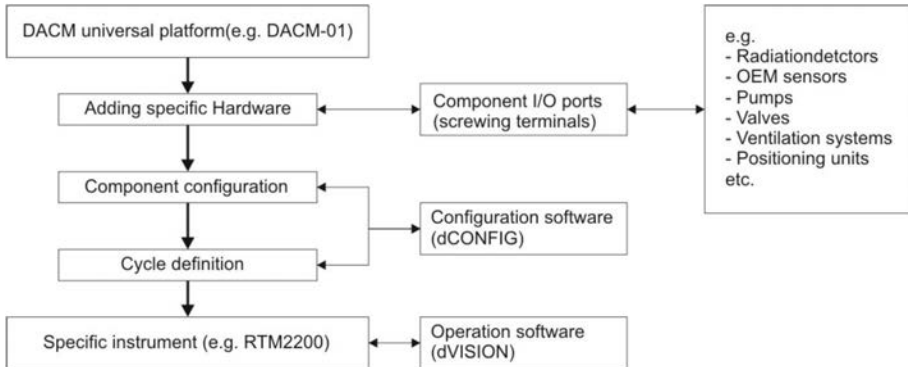


FIG. 2. DACM instrument synthesis.

2.1. Sampling control

To set and change the component status during the data acquisition process, at first a so called ‘sampling cycle’ has to be defined. The cycle can also be interpreted as the basic storage interval because each measurement series consists of a number of repetitions of this cycle. All acquired data are stored at the end of the cycle even if the physical sampling has been taken at any time within this cycle. A cycle can be split into a maximum of 32 sequential subdivisions. The

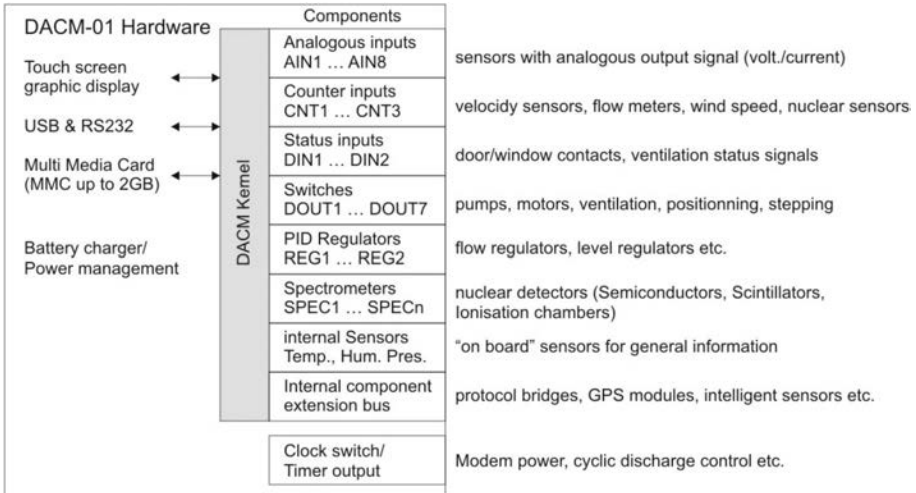


FIG. 3. DACM hardware basic implementation.

status of each component can be defined for each subdivision by the user. This is realized by a control bit or a control word. The time period of a subdivision can be set from 1 s up to the cycle interval period in steps of 1 s. The configuration software running on a PC allows the cycle definitions to be easily edited. All components are listed in a table and their status information is shown as a time chart. Only a simple mouse click into the time chart is necessary to change the status of any component within a subdivision. Several cycles may be stored on the DACM's non-volatile memory and called for execution.

2.2. Component configuration

Each implemented component can be separately configured by the user. The number and type of the configuration parameters are defined by the type of the component. A module can contain a maximum of 64 components. These components could be of the same or different types.

2.3. Data flow

All generated data are stored in binary format as it is generated by the component of origin. This ensures a 100% retraceability of the acquired data resulting in a high level of quality assurance. A component can generate one or more measurement values from the basic data. To display and transmit recent readings, all results are calculated by the module using these basic data. If the

complete data are transferred to the PC, the packed basic data are transmitted together with all necessary module and configuration information. That means that the calculation procedures are implemented in the DACM as well as in the PC software. If the data are stored in the replaceable card memory (SD card), a copy of the whole module and component configuration will be saved in parallel on the card. Thus, the results can be calculated correctly, independently of the origin of the data.

2.4. Alert management

Several components are able to generate alerts (alert sources). The type of alert is defined by the component type and required alert conditions (alert level). Both are elements of the component configuration. The alert check is carried out once per second for all components which are enabled and activated during the recent cycle subdivision. Other components are able to process alerts (i.e. to act as an alert destination). If a component acts as an alert source, a complete list of all available alert destinations is provided to select the component which will react in case of an arising alert. How the component responds to the alert situation is also defined by the component type and configuration of the alert destination.

2.5. PC interfacing

Operation and configuration software are delivered as two separate programs. Thus, once configured, the instrument looks like an instrument designed only for that purpose. This simplifies the handling and operation of the unit if it is used by unskilled staff. The configuration software allows the operator:

- (a) To transfer the configuration information and the cycle definitions from and to the module;
- (b) To edit the common module information;
- (c) To edit the configuration of the implemented components;
- (d) To edit the cycle definitions;
- (e) To save the configuration and cycle information on the hard disk.

After loading the actual module and component configuration from the DACM or hard disk, a list of all components available in the DACM will appear. Several tables show the settings of the configuration parameters clearly arranged for all components of the same type. A simple mouse click into the table opens a dialogue window to edit the parameter settings of the selected component. The changed parameters can be transferred to the DACM or saved on the hard

disk after finishing the configuration process. The cycle definition is also very easily editable by a time chart (cycle subdivisions of the x axis) which lists all components vertically. Subdivisions can be created or deleted and component states can be changed by a few mouse clicks, clicking onto the desired chart position.

2.6. Operation software (dVISION)

The operation software allows the operator:

- (a) To set the DACM real time clock;
- (b) To set the clock switch;
- (c) To start and stop the cycles uploaded to the module;
- (d) To display and transmit the recent measurement results;
- (e) To transmit the complete measurement data;
- (f) To save measurement data as a set or as single tracks;
- (g) To show the measurement data in configurable diagrams;
- (h) To export the measurement data to Excel/GIS compatible test files.

The data loaded from the module will be saved as a binary data file with a predefined filename and folder structure. This file contains all measurement data of the components, the configuration of the components and the module information. For further data processing, this file is split into several data tracks, one for each measurement value. These tracks can be saved and exported separately. Any combination of tracks can be loaded into the chart view at a later time. This allows the simultaneous display of data generated by several instruments or acquired during several time periods. The graphic options are as follows:

- (a) Selection of any time period for the chart view;
- (b) Combination of various data tracks, each with a manually or automatically scaled y axis;
- (c) Selection of the line width, line colour and line style of any data track;
- (d) A grid, cross line cursor and sliding result box for a selected track;
- (e) A scale definition for each track.

3. EXPERIMENTAL RESULTS

An experiment was carried out using mobile instruments to assess the effectiveness of remedial actions to reduce surface radiological hazards at a site

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contaminated with NORM [2]. The contaminant was in the form of charcoal with a high radium content as a result of its use as an absorber for iodine production. The remediated area was in the vicinity of Baku, Azerbaijan. One of the instruments used was the NucScout (see Fig. 4), a handy and robust 50 mm × 50 mm (optionally 75 mm × 75 mm) NaI(Tl) radionuclide identifier and quantifier. The instrument weighs less than 2 kg including GPS and ZigBee wireless connection, can be calibrated for use at a height of 1 m above the soil and gives a direct readout of activity concentration for up to 6 radionuclides chosen from a library of more than 50 radionuclides. With a time resolution of 10 s, a surveying speed of 1 m/s and a spatial resolution of 10 m, it was possible to detect a surface soil activity concentration of less than 0.2 Bq/g.



FIG. 4. NucScout with wireless 2.4 GHz ZigBee data transfer.

The contaminated material which had been deposited on the site was partially covered by a protective soil layer and the surface soil activity was therefore only slightly elevated. The activity concentrations measured at the site (see Fig. 5) showed that the 0.2 Bq/g value was only slightly higher in areas that had not yet been remediated. An A²M 4000 area monitor (see Fig. 6) enabled several parameters to be measured in a short time. The results in Fig. 7 show that the ²¹⁴Bi count rate and radon soil gas concentration were in good agreement. The data showed also that the thoron concentration in the covering material was slightly elevated because of a small, natural elevation of the thorium concentration in this soil material. However, the dose rate measured above the soil remained below 0.2 µSv/h.

4. CONCLUSION

The mobile devices used for the characterization of this remediated site demonstrated that it was possible to survey an area of more than 30 000 ha in less than two days. The new DACM technology allows a device with particular customer specifications to be created that can provide immediate results on site and that can reduce the need for time consuming and costly laboratory procedures.

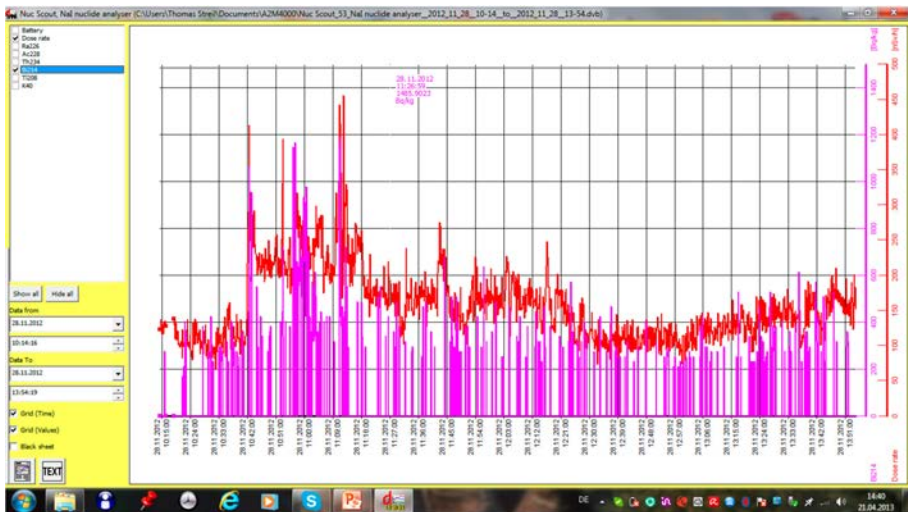


FIG. 5. NucScout screening measurements of ²¹⁴Bi in the surface soil.

MOBILE UNIT FOR SITE CHARACTERIZATION



FIG. 6. The A²M 4000 monitor for measuring radon and thoron activity concentration in soil gas together with the gamma dose rate.

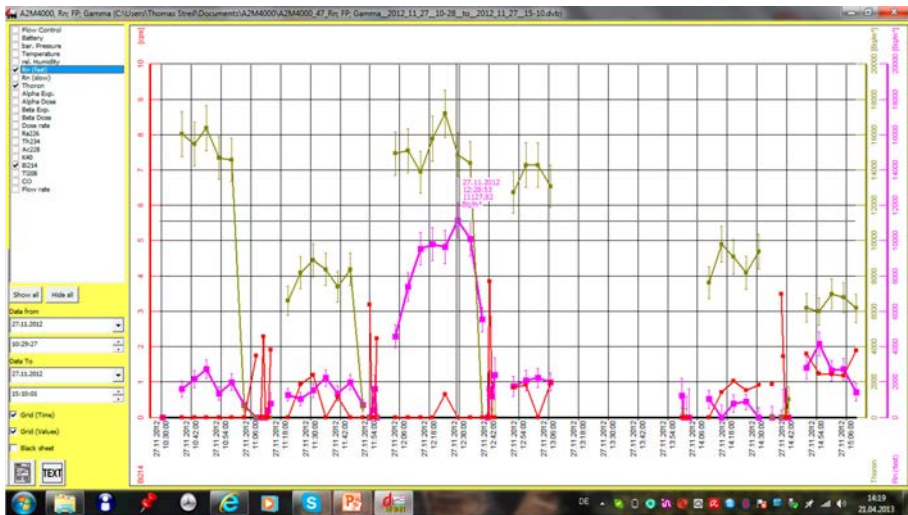


FIG. 7. Results of simultaneous measurements of radon and thoron activity concentrations in soil gas and ²¹⁴Bi count rate.

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^{222}Rn , ^{220}Rn AND PROGENY MEASURED IN A LIMESTONE CAVE AND THE ASSOCIATED RADIATION DOSE

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Abstract

Concentrations of radon, thoron and the attached and unattached fractions of the short lived alpha emitting decay progeny were measured at different locations in a limestone cave by means of CR-39 and LR-115 type II solid state nuclear track detectors in order to assess the dose due to inhalation. The committed equivalent doses per hour of exposure due to the attached and unattached fractions of ^{218}Po and ^{214}Po were evaluated in different tissues of the respiratory tract. The influence of the activity of the attached and unattached fractions of ^{218}Po and ^{214}Po and the mass of the tissue on the committed equivalent dose per hour of exposure was investigated. The annual committed effective doses due to the attached and unattached fractions of ^{218}Po and ^{214}Po were determined. A maximum value of 1.7 mSv was found for workers spending 1 h/d during the summer months inside the cave.

1. INTRODUCTION

Radon-222 and its short lived progeny (^{218}Po , ^{214}Pb , ^{214}Bi and ^{214}Po) and ^{220}Rn and its short lived progeny (^{216}Po , ^{212}Pb , ^{212}Bi and ^{212}Po) are alpha, beta and gamma emitting radionuclides. The inhalation of these radionuclides, which occurs in the open air and, at higher concentrations, in indoor air is the main source of exposure of individuals in most countries [1–6]. Since ^{222}Rn and ^{220}Rn are inert gases, their behaviour is not affected by chemical processes. In addition, their concentration levels depend strongly on geological and geophysical conditions, as well as on atmospheric parameters such as barometric pressure and rainfall. Having been formed by the decay of ^{238}U and ^{232}Th occurring naturally in the Earth's crust, they are free to move through soil pores and rock fractures until they escape into the atmosphere. Once in the atmosphere, the ^{222}Rn and ^{220}Rn atoms decay, producing isotopes of polonium, lead and bismuth (and also thallium for ^{220}Rn). These elements are heavy metals which are chemically very active and which may exist briefly as ions or free atoms before forming molecules in the condensed phase or attached to airborne dust particles, typically

in the submicron range, forming radioactive aerosols. A variable proportion of airborne ^{222}Rn and ^{220}Rn progeny remains unattached and is referred as the airborne unattached fraction. This fraction may be inhaled and deposited in the respiratory tract, where alpha emission occurs. The particle distribution of the aerosol attached fraction in the inhaled air also influences the dose to the airways, because particles of different sizes deposit preferentially in different areas of the respiratory tract.

In controlling the exposure of individuals inside caves, it is necessary to measure the concentrations of ^{222}Rn , ^{220}Rn and their progeny in the air. Instantaneous ^{222}Rn concentrations in the air of a limestone cave have been measured using alpha scintillation cells [7]. The progeny of ^{222}Rn and ^{220}Rn have been measured in the air by beta counting using an end window Geiger–Muller counter [8]. This technique has some limitations and drawbacks. Tl-208 cannot be determined and the relative error of ^{218}Po and ^{212}Bi concentration measurements is much higher than that for ^{214}Pb , ^{214}Bi and ^{212}Pb , requiring long duration counting to achieve the best accuracy. Alpha counting applied to the measurement of ^{222}Rn and ^{220}Rn progeny concentrations has some disadvantages such as the absorption of alpha particles and the degradation of alpha particle energy in the membrane filter of the counter. The use of gamma spectrometry for measuring ^{222}Rn and ^{220}Rn progeny concentrations in air suffers from some disadvantages such as low efficiency, high background and high cost. In the work described in this paper, a technique based on using CR-39 and LR-115 type II SSNTDs was used for measuring the concentration of ^{222}Rn , ^{220}Rn and their progeny at different locations inside a limestone cave. The annual committed effective doses due to the inhalation of ^{222}Rn short lived progeny were also assessed.

2. METHODS

2.1. Description of the cave

The cave that was the subject of this investigation is located in Turonian continental limestone in the Aoufous area of the province of Errachidia in the high Atlas region of Morocco (see Fig. 1). The cave is 48 m long, 2–12 m wide and 8 m high. It is visited by speleologists as well as collectors of stalactites and stalagmites.



FIG. 1. Map of Morocco showing the location of the cave studied in this investigation.

2.2. Determination of alpha activity concentrations

The alpha activity concentrations of ^{222}Rn , ^{220}Rn and their progeny were measured using the following types of solid state nuclear track detectors (SSNTDs):

- (a) CR-39 discs, 500 μm thick, manufactured by Pershore Mouldings Ltd, United Kingdom;
- (b) LR-115 type II discs, 2 cm in diameter, comprising 12 μm of cellulose nitrate on a 100 μm thick polyester base, manufactured by Kodak Pathé, France, and marketed by Dosirad, France.

The detectors were hung at different locations inside the cave for 6 h (09:00–15:00) in June 2011 (see Fig. 2). During the exposure time, alpha particles emitted by ^{222}Rn , ^{220}Rn and their progeny bombarded the SSNTD films. After the irradiation, the exposed CR-39 material was etched for 2 h at 60°C in a sodium hydroxide (NaOH) solution of 2.5 normality and the LR-115 II material was etched for 7 h at 70°C in an NaOH solution of 6.25 normality [9]. After chemical treatment, the track densities were determined using a microscope at 40x magnification. Background levels for the SSNTDs were established by placing five unexposed films inside small, well closed plastic pockets for 6 h (09:00–15:00) at each of the cave locations and determining the resulting track densities. For the experimental etching conditions, the residual thickness of the LR-115 type II SSNTD was 5 μm , which corresponds to the lower and upper

energy limits (1.6 and 4.7 MeV, respectively) for the registration of tracks of alpha particles in such LR-115 II films [10]. All alpha particles emitted by ^{222}Rn , ^{220}Rn and their progeny with a residual energy between 1.6 and 4.7 MeV and reaching the LR-115 II detector surface at an angle smaller than its critical angle of etching θ'_c are registered as bright track holes. The CR-39 detector is sensitive to all alpha particles reaching its surface at an angle smaller than its critical angle of etching θ_c . The critical angles of etching θ'_c and θ_c were calculated using the method described in Ref. [11].

The global track density rates (tracks $\cdot\text{cm}^{-2}\cdot\text{s}^{-1}$) due to alpha particles emitted by ^{222}Rn and its progeny (three alpha emitters) and ^{220}Rn and its progeny (four alpha emitters) registered on the CR-39 detectors (ρ_G^{LR}) and LR-115 II detectors (ρ_G^{LR}), after subtracting the corresponding background levels, were obtained as follows [12]:

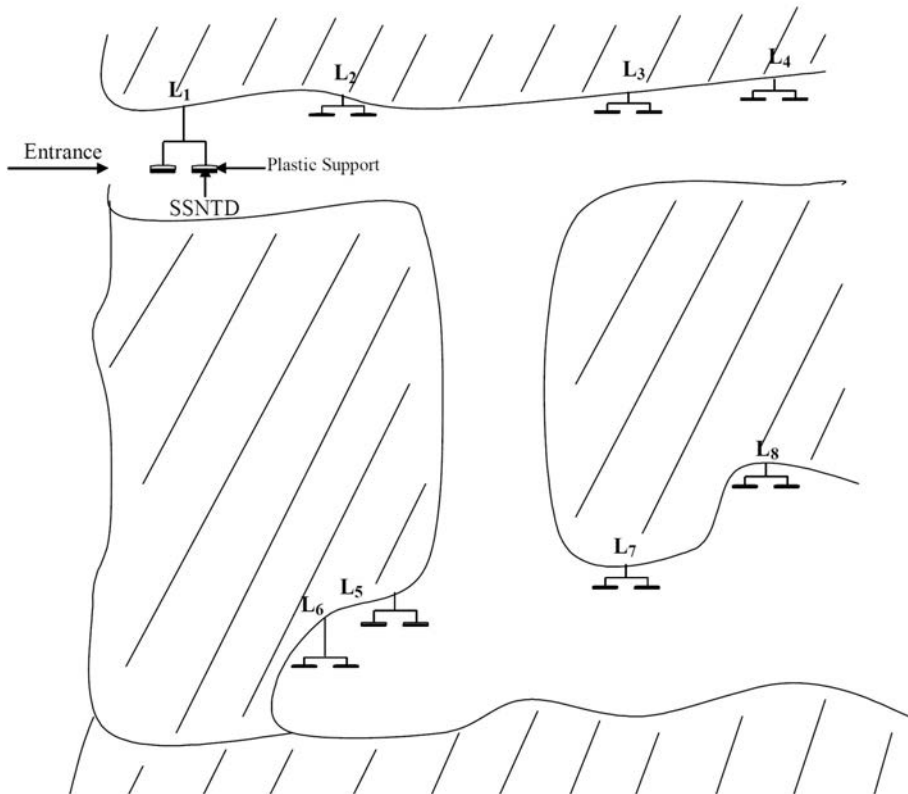


FIG. 2. Deployment of the CR-39 and LR-115 type II SSNTDs in the cave.

$$\rho_G^{CR} = \frac{\pi q^2}{2S_d} \left[\sum_1^3 A_c(j) K_j \varepsilon_j^{CR} R_j + \sum_1^4 A_c(j) K'_j \varepsilon'_j{}^{CR} R'_j \right] \quad (1)$$

and

$$\rho_G^{LR} = \frac{\pi q^2}{2S'_d} \left[\sum_1^3 A_c(j) K_j \varepsilon_j^{LR} R_j + \sum_1^4 A_c(j) K'_j \varepsilon'_j{}^{LR} R'_j \right] \quad (2)$$

where

- q is the diameter of the detector films (2 cm);
- S_d and S'_d are the surface areas of the CR-39 and LR-115 II films, respectively (cm²);
- $A_c(j)$ is the alpha activity concentration of the j^{th} alpha emitter (Bq/cm³);
- R_j and R'_j are the ranges in air of an alpha particle of index j and initial energy E_j emitted by ²²²Rn and its progeny and by ²²⁰Rn and its progeny, respectively, in the air of the cave (cm);
- K_j and K'_j are the branching ratios corresponding to the decay of ²²²Rn and its progeny and of ²²⁰Rn and its progeny, respectively;
- ε_j^{CR} and $\varepsilon'_j{}^{CR}$ are the detection efficiencies of the CR-39 film for alpha particles from ²²²Rn and its progeny and from ²²⁰Rn and its progeny, respectively;

and ε_j^{LR} and $\varepsilon'_j{}^{LR}$ are the detection efficiencies of the LR-115 II film for alpha particles from ²²²Rn and its progeny and from ²²⁰Rn and its progeny, respectively.

The relationship between the alpha activity concentration of the $(j-1)^{\text{th}}$ radionuclide $A_c(j-1)$ and that of its j^{th} unattached progeny $A_{c,u}$ and attached progeny $A_{c,a}$ is given by the following [13]:

$$A_c(j) = A_{c,u}(j) + A_{c,a}(j)$$

$$A_{c,u}(j) = \frac{\lambda_j A_{c,u}(j-1) + r_{j-1} \lambda_j A_{c,a}(j-1)}{V + \lambda_j + q^u + X} \quad (3)$$

$$A_{c,a}(j) = \frac{(1-r_{j-1}) \lambda_j A_{c,a}(j-1) + X A_{c,u}(j)}{V + \lambda_j + q^a}$$

where

- λ_j is the decay constant of radionuclide j (s^{-1});
- r_j is the recoil factor of the aerosol-attached ^{222}Rn or ^{220}Rn progeny j ;
- V is the air exchange rate at the location inside the cave (h^{-1});
- X is the mean attachment rate (h^{-1});
- q^u is the plate-out rate of the unattached ^{222}Rn or ^{220}Rn progeny;

and q^a is the plate-out rate of the attached ^{222}Rn or ^{220}Rn progeny.

The values of j for ^{222}Rn and its progeny are:

- Radon-222: $j = 0$;
- Polonium-218: $j = 1$;
- Lead-214: $j = 2$;
- Bismuth-214: $j = 3$;
- Polonium-214: $j = 4$.

The corresponding values for ^{220}Rn and its progeny are:

- Radon-220: $j = 0$;
- Polonium-216: $j = 1$;
- Lead-212: $j = 2$;
- Bismuth-212: $j = 3$;
- Polonium-212: $j = 4$.

Since ^{222}Rn and ^{220}Rn are gaseous, all of the activity is unattached, that is:

- $A_{c,u}(0) = A_c(0)$;
- $A_{c,a} = 0$.

The recoil factors for the attached ^{222}Rn and ^{220}Rn progeny are as follows [14]:

- $r_1 = r_4 = 0.8$;
- $r_2 = r_3 = 0$.

The air exchange rate V was measured using a carbon dioxide tracer method and was found to be in the range of $0.10\text{--}0.64\text{ h}^{-1}$. The mean attachment

rate is in the range of 5–500 h⁻¹ for low ventilation rates (less than 0.5 h⁻¹) [13], The plate-out rate of the unattached ²²²Rn and ²²⁰Rn progeny q^u is in the range of 5–110 h⁻¹ [15] and the corresponding figure for the attached progeny q^a is 0.05–1.1 h⁻¹ [13].

2.3. Dose assessment

In terms of the International Commission on Radiological Protection (ICRP) model [15], the human respiratory tract is divided into two major regions: the thoracic region TH and the extrathoracic region ET. The thoracic region is divided into four subregions (alveolar interstitium AI, bronchioles bb, bronchi BB and lymphatics LN_{TH}), while the extrathoracic region is divided into three subregions (anterior nasal ET₁, posterior nasal passage, larynx, pharynx and mouth ET₂ and lymphatics LN_{ET}). There are ten compartments in the thoracic region, numbered 1 to 10: AI₁, AI₂, AI₃, bb₁, bb₂, bb_{seq}, BB₁, BB₂, BB_{seq} and LN_{TH}, respectively. The extrathoracic region contains four compartments numbered 11 to 14: ET₂, ET_{seq}, LN_{ET} and ET₁, respectively.

Inhaled ²²²Rn and ²²⁰Rn progeny are assumed to be attached to particles with an activity median aerodynamic diameter (AMAD) of 0.2 μm with a geometric standard deviation σ_g of 2.5 in natural caves (i.e. without diesel engines) [13]. The rate of change of the alpha activity of the attached fraction of the j^{th} decay progeny in compartment i of the respiratory tract at any time is given by the following [15]:

$$\frac{dA_{c,a}^i(j')}{dt} = F_d(i)BA_{c,a}(j') + \sum_n (m_{n,i} + S_s)A_{c,a}^n(j') - \left[\sum_n (m_{i,n} + S_s) + \lambda_{j'} \right] A_{c,a}^i(j') \quad (4)$$

where

- $F_d(i)$ is the fractional deposition in compartment i of the respiratory tract of individuals;
- B is the average breathing rate for individuals in the cave;
- $A_{c,a}(j')$ is the alpha activity of the attached fraction of the j^{th} decay progeny at the location inside the cave (Bq/m³);
- $m_{n,i}$ is the clearance rate from region n to region i due to particle transport;

S_s is the clearance rate due to particle absorption into blood; the rate of absorption of a material into blood is the same in all regions of the respiratory tract except in the anterior nasal passages (ET_1), where no absorption occurs [15]; the value of $(m_{n,i} + S_s)$ is zero for $i = 1, 2, 3, 5, 6, 8, 9, 12$ and 14 [15];
 $m_{i,n}$ is the clearance rate from region i to region n due to particle transport;
 and $\lambda_{j'}$ is the decay constant of the j^{th} decay progeny.

Alpha activities corresponding to the attached fraction of the j^{th} decay progeny in each of the 1–14 compartments of the respiratory tract as functions of time are obtained by solving Eq. (4). The alpha equivalent dose rate (in Sv/s) in tissue T of the respiratory tract of an individual due to the inhalation of the attached fraction of the j^{th} decay progeny is given by:

$$\dot{H}_T^a(j')(t) = A_{c,a}^T(j')(t)D_{SP}^T(j')w_R \quad (5)$$

where $A_{c,a}^T(j')(t)$ is the alpha activity of the attached fraction of the j^{th} decay progeny in tissue T of the respiratory tract (Bq), and $D_{SP}^T(j')$ is the specific alpha dose deposited by alpha particles emitted by 1 Bq of the j^{th} decay progeny inside tissue T (Gy).

The value of the specific alpha dose $D_{SP}^T(j')$ is given by:

$$D_{SP}^T(j') = k \frac{K_{j'}R_{j'}S_{j'}}{m_T} \quad (6)$$

where

m_T is the mass of target tissue T [15];
 $K_{j'}$ is the branching ratio;
 $R_{j'}$ is the range of the alpha particle emitted by the j^{th} decay progeny;
 $S_{j'}$ is the stopping power of tissue T for the emitted alpha particle;

and k is a conversion factor of 1.6×10^{-10} .

$R_{j'}$ and $S_{j'}$ were calculated using the TRIM program [16] (using the elemental chemical composition of tissues given in Ref. [17]). The equivalent

dose to tissue T of the respiratory tract for the attached fraction of the j^{th} decay progeny is given by:

$$H_T^a(j') = \int_0^{t'_e} H_T^a(j')(t) dt \tag{7}$$

where t'_e is the exposure time of tissue T .

The rate of change of the alpha activity of the unattached fraction of the j^{th} decay progeny (AMAD = 1 nm) [13] in compartment i of the respiratory tract at any time is given by [15]:

$$\begin{aligned} \frac{dA_{c,u}^i(j')}{dt} = & F_d(i)BA_{c,u}(j') + \sum_n (m_{n,i} + S_s)A_{c,u}^n(j') \\ & - \left(\sum_n (m_{i,n} + S_s) + \lambda_{j'} \right) A_{c,u}^i(j') \end{aligned} \tag{8}$$

where $A_{c,u}(j')$ is the alpha activity of the unattached fraction of the j^{th} decay progeny at the location inside the cave (Bq/m³) and the other terms are as in Eq. (4). Alpha activities corresponding to the unattached fraction of the j^{th} decay progeny in each of the 1–14 compartments of the respiratory tract as functions of time are obtained by solving Eq. (8). The alpha equivalent dose rate (in Sv/s) in tissue T of the respiratory tract of an individual due to inhalation of the unattached fraction of the j^{th} decay progeny is given by:

$$\dot{H}_T^u(j')(t) = A_{c,u}^T(j')(t)D_{SP}^T(j')w_R \tag{9}$$

where $A_{c,u}^T(j')(t)$ is the alpha activity of the unattached fraction of the j^{th} decay progeny in tissue T of the respiratory tract (Bq). The equivalent dose in tissue T of the respiratory tract for the unattached fraction of the j^{th} decay progeny is given by:

$$H_T^u(j') = \int_0^{t'_e} H_T^u(j')(t) dt \tag{10}$$

Regional doses, weighted with factors assigned for the partition of radiation detriment (e.g. A_{BB} and A_{bb}), are summed to give a value of committed equivalent dose for the thoracic region $H_{TH}(j')$ and the extrathoracic region $H_{ET}(j')$:

$$H_{TH}(j') = A_{BB} \{H_{BB}^a(j') + H_{BB}^u(j')\} + A_{bb} \{H_{bb}^a(j') + H_{bb}^u(j')\} + A_{AI} \{H_{AI}^a(j') + H_{AI}^u(j')\} + A_{LN_{TH}} \{H_{LN_{TH}}^a(j') + H_{LN_{TH}}^u(j')\} \quad (11)$$

and

$$H_{ET}(j') = A_{ET_1} \{H_{ET_1}^a(j') + H_{ET_1}^u(j')\} + A_{ET_2} \{H_{ET_2}^a(j') + H_{ET_2}^u(j')\} + A_{LN_{ET}} \{H_{LN_{ET}}^a(j') + H_{LN_{ET}}^u(j')\} \quad (12)$$

where $H_{BB}^a(j')$, $H_{BB}^u(j')$, $H_{bb}^a(j')$, $H_{bb}^u(j')$, $H_{AI}^a(j')$, $H_{AI}^u(j')$, $H_{LN_{TH}}^a(j')$ and $H_{LN_{TH}}^u(j')$ are the equivalent doses to the BB, bb, AI and LN_{TH} tissues of the thoracic region, respectively, and $H_{ET_1}^a(j')$, $H_{ET_1}^u(j')$, $H_{ET_2}^a(j')$, $H_{ET_2}^u(j')$, $H_{LN_{ET}}^a(j')$ and $H_{LN_{ET}}^u(j')$ are the equivalent doses to the ET_1 , ET_2 and LN_{ET} tissues of the extrathoracic region, respectively. The weighting factors for the partition of radiation detriment for the BB, bb, AI and LN_{TH} tissues of the thoracic region are, respectively [15]:

- $A_{BB} = 0.333$;
- $A_{bb} = 0.333$;
- $A_{AI} = 0.333$;
- $A_{LN_{TH}} = 0.001$.

The weighting factors for the partition of radiation detriment for the ET_1 , ET_2 and LN_{ET} tissues of the extrathoracic region are, respectively [15]:

- $A_{ET_1} = 0.001$;
- $A_{ET_2} = 1$;
- $A_{LN_{ET}} = 0.001$.

The total committed effective dose (mSv/h) due to the unattached and attached fractions of the ^{222}Rn decay progeny ^{218}Po and ^{214}Po is given by:

$$E = E^u + E^a \quad (13)$$

where

$$\begin{aligned}
 E^u &= 0.12[H_{\text{TH}}^u(^{218}\text{Po}) + H_{\text{TH}}^u(^{214}\text{Po})] + 0.025[H_{\text{ET}}^u(^{218}\text{Po}) + H_{\text{ET}}^u(^{214}\text{Po})] \\
 E^a &= 0.12[H_{\text{TH}}^a(^{218}\text{Po}) + H_{\text{TH}}^a(^{214}\text{Po})] + 0.025[H_{\text{ET}}^a(^{218}\text{Po}) + H_{\text{ET}}^a(^{214}\text{Po})]
 \end{aligned}
 \tag{14}$$

3. RESULTS AND DISCUSSION

3.1. Alpha activity concentrations of ^{222}Rn , ^{220}Rn and their progeny

The activity concentrations of ^{222}Rn and ^{220}Rn at the various locations in the cave are shown in Table 1. From the statistical error on the track counting, the error associated with the track density rate measurements could be determined, from which a relative uncertainty of 8% was established for the ^{222}Rn and ^{220}Rn concentrations. The ^{222}Rn concentrations were higher than the ^{220}Rn concentrations. Owing to the short half-life of ^{220}Rn (55 s), its diffusion length in air is shorter than that of ^{222}Rn (half-life 3.82 d). The concentrations increased with the distance from the entrance of the cave as a result of the decreasing rate of air exchange. Thus, the highest exposures are at those locations furthest from the entrance (L7 and L8 in Table 1). In ICRP Publication 65 [18], the ICRP recommends a radon action level of 500–1500 Bq/m³ for workplaces and this range has been adopted by the European Commission [19]. The UK Health and Safety Executive has adopted a radon action level of 400 Bq/m³ for workplaces, based on advice from the former National Radiological Protection Board [20]. In the United States of America, a reference level of 150 Bq/m³ is adopted for workplaces [21]. In Ireland, the advisory reference level for workplaces is 200 Bq/m³ [22]. For workplaces in Morocco, the ICRP Publication 65 recommendations are applied [18]. The activity concentrations of ^{222}Rn and ^{220}Rn progeny were determined from Eqs (1)–(3) and the results are shown in Table 2. The relative uncertainty of the radon progeny determination was 8%. The concentrations of the attached fraction are clearly higher than those of the unattached fraction.

TABLE 1. ^{222}Rn AND ^{220}Rn CONCENTRATIONS

Location code	Air exchange rate (h^{-1})	Activity concentration (Bq/m^3)			
		Rn-222		Rn-220	
		Mean	SD	Mean	SD
L1	0.64	96	7	11	1
L2	0.35	147	11	14	1
L3	0.31	176	14	15	1
L4	0.28	200	16	22	2
L5	0.22	286	22	28	2
L6	0.19	376	30	31	2
L7	0.14	564	45	44	3
L8	0.10	798	63	70	5

Note: SD — standard deviation.

3.2. Dose

The committed equivalent doses per hour of exposure of tissues in the respiratory tract to the attached and unattached fractions of short lived ^{222}Rn progeny were determined for an adult with a breathing rate corresponding to light exercise. The time variations in ^{218}Po and ^{214}Po activities in various tissues for different age groups are given in Ref. [23]. The results for ^{218}Po and ^{214}Po are shown in Tables 3 and 4, respectively. The statistical relative uncertainty of the committed equivalent dose determination was about 10%.

TABLE 2. ²¹⁸Po AND ²¹⁴Po CONCENTRATIONS

Location code	Air exchange rate (h ⁻¹)	Activity concentration (Bq/m ³)							
		Po-218				Po-214			
		Attached fraction		Unattached fraction		Attached fraction		Unattached fraction	
		Mean	SD	Mean	SD	Mean	SD	Mean	SD
L1	0.64	67	5	9.7	0.7	32	2	0.01	0.000 8
L2	0.35	106	7	15	1	61	4	0.02	0.001
L3	0.31	127	9	18	1	76	5	0.02	0.001
L4	0.28	144	10	20	1	88	6	0.03	0.002
L5	0.22	207	16	29	2	134	10	0.04	0.003
L6	0.19	273	20	38	2	181	13	0.05	0.003
L7	0.14	411	30	57	4	285	21	0.08	0.006
L8	0.10	584	43	81	6	419	31	0.12	0.008

Note: SD — standard deviation.

The committed equivalent dose to a given tissue is influenced by the activity integral (the integral of the activity–time curve given by Eq. (7) or (10) as appropriate), the mass of the target tissue (m_T) and the weighting factor for the partition of the radiation detriment (Eqs (11) and (12)). The committed equivalent doses are negligible in the LN_{TH} (thoracic region) and LN_{ET} (extrathoracic region) tissues. This is due to the fact that these tissues show lower activity integrals

TABLE 3. EQUIVALENT DOSE TO TISSUES IN THE RESPIRATORY TRACT: ^{218}Po

Location code	Committed equivalent dose per hour of exposure												
	Thoracic region						Extrathoracic region						
	AI ($\text{Sv}/10^9$)		bb ($\text{Sv}/10^6$)		BB ($\text{Sv}/10^6$)		ET ₁ ($\text{Sv}/10^6$)		ET ₂ ($\text{Sv}/10^6$)				
Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
L1	14	1	1.4	0.1	0.50	0.05	0.92	0.09	34	3			
	13		1.5		0.53			0.8	37				
L2	21	2	2.2	0.2	0.78	0.07	1.4	0.1	56	5			
			2.4		0.82	0.08			57				
L3	26	2	2.6	0.2	0.94	0.08	1.7	0.1	67	6			
	25		2.9		1.0	0.1			69				
L4	29	2	3.0	0.2	1.1	0.1	2.0	0.1	76	7			
			3.3	0.3					78				
L5	42	4	4.3	0.4	1.5	0.1	2.8	0.2	110	10			
	41		4.8		1.6								
L6	56	5	5.6	0.5	2.0	0.2	3.7	0.3	140	10			
	54		6.3	0.6	2.1				150				

TABLE 3. EQUIVALENT DOSE TO TISSUES IN THE RESPIRATORY TRACT: ²¹⁸Po (cont.)

Location code	Committed equivalent dose per hour of exposure											
	Thoracic region						Extrathoracic region					
	AI (Sv/10 ⁹)		bb (Sv/10 ⁶)		BB (Sv/10 ⁶)		ET ₁ (Sv/10 ⁶)		ET ₂ (Sv/10 ⁶)		Mean	SD
L7	84	8	8.5	0.8	3.0	0.3	5.6	0.5	220	20		
	82		9.5	0.9	3.2							
L8	120	10	12	1	4.3	0.4	8.0	0.7	310	30		
			13		4.5							
Unattached fraction												
L1	0.04	0.003	0.66	0.06	0.89	0.08	0.9	0.08	31	3		
	0.06	0.006	0.84	0.08	0.92	0.09		0.09	32			
L2	0.06	0.005	1.0	0.1	1.4	0.1	1.4	0.1	47	4		
	0.09	0.009	1.3						49			
L3	0.07	0.007	1.2	0.1	1.6	0.1	1.7	0.1	59	5		
	0.11	0.01	1.5		1.7							
L4	0.08	0.007	1.4	0.1	1.8	0.1	1.9	0.1	64	6		
	0.12	0.01	1.7		1.9				67			
L5	0.11	0.01	2.0	0.1	2.6	0.2	2.7	0.2	92	8		
	0.18	0.01	2.5	0.2	2.7				96	9		

TABLE 3. EQUIVALENT DOSE TO TISSUES IN THE RESPIRATORY TRACT: ^{218}Po (cont.)

Location code	Committed equivalent dose per hour of exposure												
	Thoracic region						Extrathoracic region						
	AI ($\text{Sv}/10^9$)		bb ($\text{Sv}/10^6$)		BB ($\text{Sv}/10^6$)		ET ₁ ($\text{Sv}/10^6$)		ET ₂ ($\text{Sv}/10^6$)				
Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
L6	0.15	0.01	2.6	0.2	3.5	0.3	3.5	0.3	3.5	0.3	120	10	
	0.23	0.02	3.3	0.3	3.6		3.6		3.6		130		
L7	0.23	0.02	3.9	0.3	5.2	0.5	5.3	0.5	5.3	0.5	180	10	
	0.35	0.03	4.9	0.4	5.4		5.4		5.4		190		
L8	0.32	0.02	5.5	0.5	7.4	0.7	7.5	0.7	7.5	0.7	260	12	
	0.49	0.04	7.0	0.7	7.7		7.7		7.7		270	20	

Note: SD — standard deviation. Where two values are given, they relate to a female and male worker, respectively.

TABLE 4. EQUIVALENT DOSE TO TISSUES IN THE RESPIRATORY TRACT: ²¹⁴Po

Location code	Committed effective dose per hour of exposure												
	Thoracic region						Extrathoracic region						
	AI (Sv/10 ¹⁷)	Mean	SD	bb (Sv/10 ¹³)	Mean	SD	BB (Sv/10 ¹⁴)	Mean	SD	ET ₁ (Sv/10 ¹⁴)	Mean	SD	ET ₂ (Sv/10 ¹²)
Attached fraction	L1	620	60	6.3	0.6	22	2	41	4	21	2		
		610		7.0		24							
L2	1 200	100	12	1	44	4	81	7	40	4			
	1 100		14		46				41				
L3	1 400	100	15	1	54	5	100	10	49	4			
			17		57				51	5			
L4	1 700	100	18	1	63	6	120	10	58	5			
			20		67				59				
L5	2 600	200	27	2	96	9	180	10	87	8			
			30		100	10			90				
L6	3 500	300	36	3	130	10	240	20	120	10			
			40	4	140								

TABLE 4. EQUIVALENT DOSE TO TISSUES IN THE RESPIRATORY TRACT: ^{214}Po (cont.)

Location code	Committed effective dose per hour of exposure												
	Thoracic region						Extrathoracic region						
	AI (Sv/10 ¹⁷)		bb (Sv/10 ¹³)		BB (Sv/10 ¹⁴)		ET ₁ (Sv/10 ¹⁴)		ET ₂ (Sv/10 ¹²)				
Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD
L7	5 600	500	57	5	200	20	380	30	180	10			
	5 500		64	6	210				190				
L8	8 300	800	84	8	300	20	550	50	270	20			
	8 100	700	94	9	320	30			280				
Unattached fraction													
L1	0.004	0.000 4	0.007	0.000 5	0.09	0.008	0.09	0.008	0.04	0.003			
	0.006	0.000 6	0.008	0.000 7	0.10	0.01	0.10	0.01	0.10	0.004			
L2	0.080	0.007	0.013	0.001	0.18	0.01	0.18	0.01	0.08	0.007			
	0.012	0.001	0.017						0.08				
L3	0.080	0.007	0.013	0.001	0.18	0.01	0.18	0.01	0.08	0.007			
	0.012	0.001	0.017						0.08				
L4	0.010	0.001	0.020	0.002	0.027	0.02	0.27	0.02	0.12	0.01			
	0.017		0.025		0.28				0.12				
L5	0.015	0.002	0.026	0.002	0.036	0.03	0.36	0.03	0.16	0.01			
	0.023		0.033	0.003	0.37		0.37		0.16				

TABLE 4. EQUIVALENT DOSE TO TISSUES IN THE RESPIRATORY TRACT: ²¹⁴Po (cont.)

Location code	Committed effective dose per hour of exposure											
	Thoracic region						Extrathoracic region					
	AI (Sv/10 ¹⁷)		bb (Sv/10 ¹³)		BB (Sv/10 ¹⁴)		ET ₁ (Sv/10 ¹⁴)		ET ₂ (Sv/10 ¹²)			
Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	Mean	SD	
L6	0.020	0.002	0.033	0.003	0.045	0.04	0.45	0.04	0.20	0.02		
	0.029		0.042	0.004	0.46		0.46					
L7	0.030	0.003	0.052	0.004	0.071	0.06	0.72	0.06	0.31	0.03		
	0.046	0.004	0.067	0.006	0.07	0.07	0.73	0.07	0.32			
L8	0.046	0.004	0.078	0.006	1.1	0.1	0.11	0.1	0.47	0.04		
	0.070	0.007	0.10	0.01					0.49			

Note: SD — standard deviation. Where two values are given, they relate to a female and male worker, respectively.

and weighting factors for the partition of radiation detriment. The following observations can be made from Tables 3 and 4:

- (a) The committed equivalent doses from the attached and unattached fractions of ^{218}Po were higher in the ET region than in the TH region. This was because, after inhalation, ^{218}Po and ^{214}Po are deposited in the extrathoracic region ET and thoracic region TH of the lung. According to the ICRP compartmental model [15], some ^{218}Po atoms are transferred from the TH region to the ET region, since this radionuclide has a half-life of 3.05 min.
- (b) The committed equivalent doses from the attached and unattached fractions of ^{214}Po are smaller than those from ^{218}Po in both the ET and TH regions. This is due to the fact that the half-life of ^{214}Po (164 μs) is very short compared with the exposure time of the tissues. This means that the ^{214}Po comes essentially from the decay of ^{218}Po present in the ET and TH regions.
- (c) The committed equivalent doses are clearly higher in the bb and BB tissues than in the AI tissue of the thoracic region, even though the latter tissue shows a higher activity integral [23]. This is due to the greater mass of the AI tissue.
- (d) The committed equivalent doses are higher in the ET_2 tissue than in the ET_1 tissue of the extrathoracic region, even though the latter tissue has a smaller mass and a higher activity integral. This is because the ET_2 tissue shows a higher weighting factor for the partition of radiation detriment.
- (e) The committed equivalent doses in the bb tissue are higher for an adult male than for an adult female. This is due to the predominance of the activity integral [23].

The annual committed effective dose per hour of exposure to ^{218}Po and ^{214}Po was calculated from Eq. (13) for individuals undertaking light work within the cave. The results are shown in Table 5. The statistical relative uncertainty of the committed effective dose determination was about 10%. The dose rates were highest at the L8 location because of the higher activity concentrations there (see Table 2).

Equation (13) was also used to calculate the dose to members of the public by assuming a breathing rate corresponding to an adult at rest. The results are shown in Table 6 together with doses calculated from the ^{222}Rn concentration using the dose coefficients of 9 nSv/h per Bq/m^3 published by United Nations Scientific Committee on the Effects of Atomic Radiation [24] and 10.5 nSv/h per Bq/m^3 published by the ICRP [25]. There was good agreement between the three methods of calculation.

TABLE 5. ANNUAL EFFECTIVE DOSE RATES RECEIVED FOR INDIVIDUALS WORKING IN THE CAVE DURING THE SUMMER MONTHS

Location code	Dose rate ($\mu\text{Sv/h}$)					
	Attached fraction		Unattached fraction		Total	
	Mean	SD	Mean	SD	Mean	SD
L1	1.1	0.1	1.0	0.1	2.1	0.2
	1.2				2.2	
L2	1.8	0.1	1.5	0.1	3.3	0.3
	1.9				3.4	
L3	2.2	0.2	1.8	0.1	4.0	0.4
			1.9		4.1	
L4	2.5	0.2	2.0	0.2	4.5	0.4
			2.1		4.7	
L5	3.5	0.3	2.9	0.2	6.5	0.6
	3.7		3.0		6.7	
L6	4.6	0.4	3.9	0.3	8.5	0.8
	4.8		4.0		8.8	
L7	7.0	0.7	5.8	0.5	13	1
	7.3		6.0		6.6	
L8	9.9	0.9	7.2	0.7	17	1
	10		1		8.5	

Note: SD — standard deviation. Where two values are given, they relate to a female and male worker, respectively.

TABLE 6. ANNUAL EFFECTIVE DOSE RATES FOR MEMBERS OF THE PUBLIC VISITING THE CAVE

Location code	Aerosol fraction	Dose rate ($\mu\text{Sv/h}$)				
		Female	Male	Av.	Calculated from Rn-222 concentration	
					9 nSv/h per Bq/m^3 [24]	10.5 nSv/h per Bq/m^3 [25]
L1	Attached	0.3 ± 0.03	0.3 ± 0.03			
	Unattached	0.1 ± 0.01	0.4 ± 0.04			
	Total	0.3 ± 0.03	0.7 ± 0.07	0.5 ± 0.05	0.51	0.60
L2	Attached	0.4 ± 0.04	0.5 ± 0.05			
	Unattached	0.1 ± 0.01	0.6 ± 0.06			
	Total	0.5 ± 0.05	1.1 ± 0.1	0.8 ± 0.08	0.79	0.93
L3	Attached	0.5 ± 0.05	0.6 ± 0.06			
	Unattached	0.1 ± 0.01	0.7 ± 0.07			
	Total	0.6 ± 0.06	1.3 ± 0.1	0.9 ± 0.09	0.95	1.11
L4	Attached	0.5 ± 0.05	0.7 ± 0.07			
	Unattached	0.2 ± 0.02	0.8 ± 0.08			
	Total	0.7 ± 0.07	1.5 ± 0.1	1.1 ± 0.1	1.08	1.26
L5	Attached	0.8 ± 0.08	1.0 ± 0.1			
	Unattached	0.2 ± 0.02	1.1 ± 0.1			
	Total	1.0 ± 0.1	2.1 ± 0.2	1.5 ± 0.1	1.54	1.80
L6	Attached	1.0 ± 0.1	1.3 ± 0.1			
	Unattached	0.3 ± 0.03	1.5 ± 0.1			
	Total	1.3 ± 0.1	2.7 ± 0.2	2.0 ± 0.2	2.03	2.36

TABLE 6. ANNUAL EFFECTIVE DOSE RATES FOR MEMBERS OF THE PUBLIC VISITING THE CAVE (cont.)

Location code	Aerosol fraction	Dose rate ($\mu\text{Sv/h}$)				
		Female	Male	Av.	Calculated from Rn-222 concentration	
					9 nSv/h per Bq/m^3 [24]	10.5 nSv/h per Bq/m^3 [25]
L7	Attached	1.5 ± 0.1	1.9 ± 0.1			
	Unattached	0.5 ± 0.04	2.2 ± 0.2			
	Total	2.0 ± 0.2	4.1 ± 0.4	3.1 ± 0.3	3.04	3.55
L8	Attached	2.2 ± 0.2	2.7 ± 0.2			
	Unattached	0.6 ± 0.06	3.2 ± 0.3			
	Total	2.8 ± 0.2	5.9 ± 0.5	4.3 ± 0.4	4.31	5.02

4. CONCLUSION

The alpha activity concentrations of ^{222}Rn , ^{220}Rn and their attached and unattached decay progeny were determined at various locations inside a limestone cave using CR-39 and LR-115 type II SSNTDs. This measurement method has the advantage of being inexpensive, accurate, and sensitive, without the need for standard calibration sources. The committed equivalent doses to tissues of the respiratory tract were evaluated for the decay progeny ^{218}Po and ^{214}Po . It was shown that the dose rate increased with increasing airborne activity concentration and with decreasing tissue mass. The maximum dose rate was found to be $19 \mu\text{Sv/h}$. For a worker spending 90 h/a inside the cave collecting stalactites and stalagmites, this corresponds to an annual committed effective dose of 1.7 mSv. This dose level is higher than the 1.15 mSv/a average worldwide effective dose from radon progeny inhalation given in Ref. [26].

ACKNOWLEDGEMENT

This work was performed under a URAC-15 research contract with the National Centre for Scientific and Technical Research (CNRST), Rabat, Morocco.

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Invited Paper

HAZARD ASSESSMENT AT THE SITE OF A FORMER COAL FIRED POWER PLANT

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Abstract

The precautionary principle (PP) is laid down explicitly in the EU Treaty as one of the starting points of environmental policy. The European Commission indicated that this principle should be applied whenever there is a reasonable suspicion of the existence of a health or environmental risk. Furthermore, the Commission indicated that measures based on the PP should not be aimed at completely eliminating any risk; it was assumed that such an effort would be unrealistic. Decades of industrialization and extensive exploitation of natural resources have left certain areas heavily polluted. It is essential that environmental burdens left behind by past state controlled industries be addressed. What were once (theoretically) government problems have now been transferred to new owners, in most cases without a clear assignment of the environmental responsibility. Contaminated industrial legacy sites often pose a hazard to individuals living nearby, either directly through negative health impacts or indirectly through pollutants in the food chain. Additionally, increasing industrialization and population density have led to situations where humans and the environment are exposed to a variety of hazards. Often little is known about the mid term and long term health and ecological consequences of these hazards, especially when they occur together. The assessment of chronic low level mixed exposures presents considerable challenges for methodology and data interpretation. Determining the effects of mixed hazards is complex, as the contaminants may interact at different levels. One of the measures based on the PP is hazard characterization of the site, enabling an appropriate remediation programme to be established. The site of interest presented in the paper was heavily polluted with coal sludge and fly ash originating from a former power plant. The coal had a relatively high content of radionuclides of natural origin. The combustion residue was deposited next to the sea, separated by a simple but robust stone wall. The plant had been closed for eight years before a radiological characterization of the sludge was initiated. Research was focused on gathering knowledge about the local Mediterranean biodiversity and microbiota systems which could be used during the remediation of this polluted site. The results are also expected to be helpful in investigating the by-product use of the coal sludge and ash because most if not all of the potential risks are

essentially due to mixed contaminant exposure situations with a mix of radionuclides, heavy metals, metalloids and/or organic pollutants.

1. INTRODUCTION

The precautionary principle (PP) introduces a set of measures to be taken while utilizing a certain technology in order not to cause harm [1]. For radiological hazards, as for other types of hazard, the most efficient measure is a hazard assessment of the site of interest, enabling an appropriate remediation programme to be established if necessary. The site described in this paper was heavily polluted with coal sludge and fly ash from a former power plant. In the literature, the term 'risk' is often confused with term 'hazard'. Radiation protection professionals, epidemiologists and professionals in the field of occupational and environmental health define risk as the probability or chance that a hazard posed by a stressor will cause injury or other harm to a worker or member of the public. Thus, 'hazard' is the potential to cause harm, while 'risk' is associated with the likelihood of harm occurring in defined circumstances. Risk is usually qualified by some statement of the severity of harm. Thus, risk can be seen as the likelihood of occurrence multiplied by the seriousness once an incident has occurred (see, e.g., the definition of risk in Ref. [2]). The relationship between hazard and risk must be treated very cautiously when using these terms for evaluating situations involving radiological pollutants, chemical pollutants or a mixture of both. An additional PP measure is 'health impact assessment' to the public, which should ideally be one of the key principles during the process of decision making in physical planning and the construction of industrial infrastructure and other facilities. It is also necessary to integrate 'environmental health' in the process of strategic environmental and health assessment because of the fact that harmful non-radioactive contaminants can cause much more intense environmental pollution than radioactive contaminants. Situations involving a mixture of non-radiological and radiological contaminants pose an immense research challenge. The objective is to understand the mechanisms and processes by which mixtures of contaminants interact to induce adverse effects on biota and the environment in order to determine whether radiation protection criteria are sufficient when considering a mixed contaminant situation. An occupational health impact assessment is also to be included in the process because very often new types of workplace situation will be created during the remediation process and in the development of new technology aimed at by-product use of residues. The so called European Waste Framework Directive (WFD) [3] gives an explanation of when waste ceases to be waste and becomes a secondary raw material (so called end-of-waste criteria), and describes how to

distinguish between waste and by-products. It includes two new recycling and recovery targets to be achieved by 2020: 50% preparing for use and recycling of certain waste materials from households and other similar origins; and 70% preparing for use, recycling and other recovery of construction and demolition waste. As the site of interest was a former coal combustion plant, the possibilities for by-product use and recycling were of great interest. The site consisted of construction and demolition waste and combustion residue deposit (fly ash and slag) deposited directly next to the sea. The deposit was separated from the sea by a simple but robust stone wall. The coal that had been burnt in the plant had a relatively high content of radionuclides of natural origin. A radiometric analysis of the combustion residue was needed, as well as measurements of the parameters of the relevant exposure pathways to remediation workers and members of the public. The measurement strategies needed to be appropriate for the relatively diverse radionuclide content and physical characteristics of the material (e.g. scale, bulk material and dust). In addition, a rapid, on-site exchange of measurement experience among the radiation protection experts was desirable. Interactions between radiological and non-radiological contaminants also needed to be investigated.

The radiological characterization of the sludge residue was initiated in order to facilitate remediation decisions and to ensure the minimum possible hazard to the environment and the public. At this time, the power plant had been closed for eight years, no human activities had been carried out near the residue deposit and the site had been left undisturbed. An independent microecological terrestrial system had established itself on the residue surface. The WFD requires that waste be managed without endangering human health and harming the environment (in particular, without risk to water, air, soil, plants or animals), without causing a nuisance through noise or odour, and without adversely affecting the countryside or places of special interest. Consequently, an extensive radiological investigation of the specific local ecosystem that had established itself on the coal slag and ash residue was launched. The research challenge was to acquire as much relevant radiological data as possible about the behaviour of biota populating the site, growing and feeding itself exclusively on the nutrients incorporated into the residue. The interaction between the terrestrial flourishing biota with the seawater life immediately next to the sea wall was also investigated. Knowledge was needed on the transfer of pollutants between different environmental compartments and on the impact of cumulative stressors, including chemical mixtures. This knowledge was to be gathered by developing and using improved assessment tools and novel models to reduce the uncertainty in current risk assessment and screening methodologies, for example by improving the scientific basis for setting safety factors or benchmarks.

2. SURVEY APPROACH

Most if not all exposure situations involving radionuclides of natural origin at legacy sites are essentially mixed contaminant exposure situations. However, it was decided that the health risk assessment would be performed for radiological hazards only, on the basis that radiological measurement methods and some of the applicable methodologies could be used to deal with non-radiological contaminants existing at the site. Radiation exposure conditions entail a variety of radionuclides which are treated in impact and risk assessments in an additive way using appropriate weighing factors for the various radiation types. The site to be surveyed was divided into subareas, as shown in Fig. 1, using as a criterion an annual effective dose to a member of the public of 1 mSv above the



FIG. 1. Aerial view of the site.

local background level [4]. The 1 mSv criterion was interpreted in terms of the corresponding dose rate at a height of 1 m. A view of the area delineated in Fig. 1 as Area D, as it appeared at the start of the radiological survey, is shown in Fig. 2. This area contained deposits of fly ash and slag and was therefore regarded as the most important of the various subareas. Subarea C shown in Fig. 1 contained no combustion residues and was regarded as uncontaminated. The sea is visible as area Dm.

3. ROUGH RADIOACTIVITY MAP OF THE SITE

A simple, rapid and reliable field dose rate survey method was developed, incorporating a global positioning system (GPS) which provided location and time information in all weather conditions. The survey was performed by walking randomly over the site, collecting time, GPS location and dose rate data. More than 15 000 sets of data were recorded. The data were incorporated into a single time dependent file and a tracking path was formed, as shown in Fig. 3. The ambient dose rate values and GPS Gauss Krüger coordinates obtained by the GPS formed the basis for the rough radioactivity map shown in Fig. 4. At the

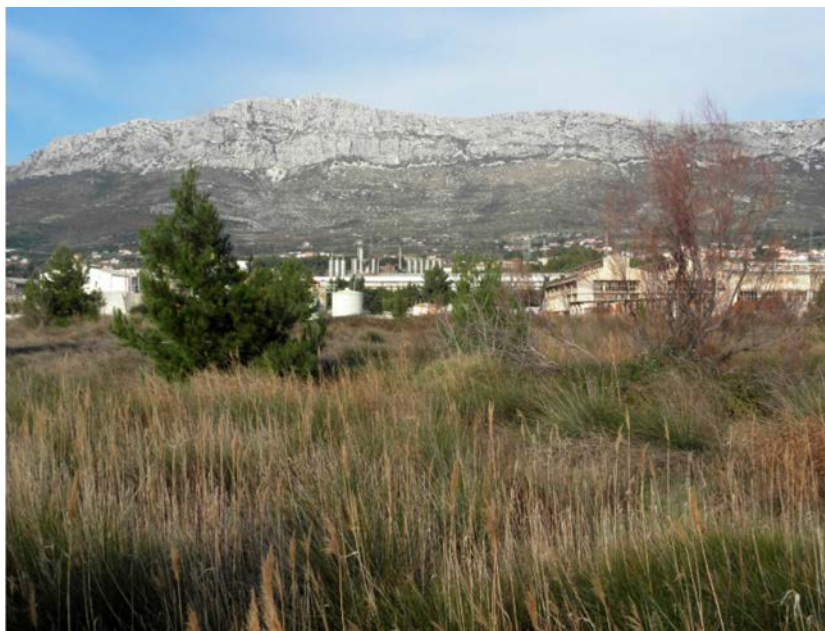


FIG. 2. Area D containing combustion residue deposit.

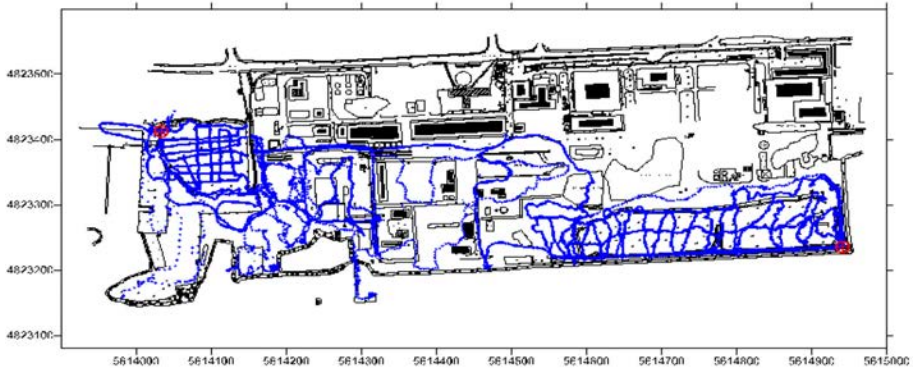


FIG. 3. Ambient dose rate tracking path formed by randomly walking over the site.

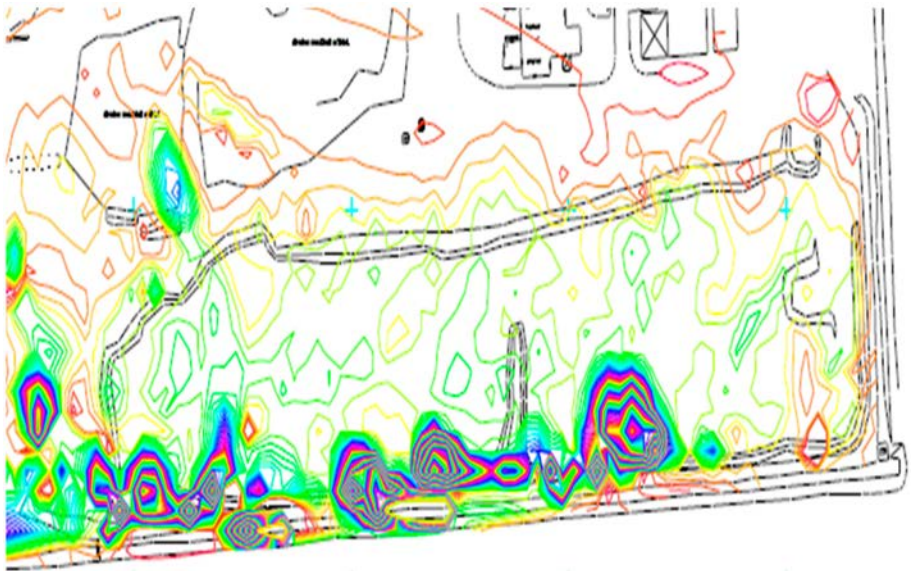


FIG. 4. Rough radioactivity map of ambient dose rates above the surface of the residue deposit.

spots where the rough radioactivity map suggested higher activity concentrations, as shown in Fig. 4, soil samples were collected and the activity concentrations measured in situ. The soil sampling was performed in accordance with recognized area-volume sampling methods [5].

4. RELATIONSHIP BETWEEN DOSE AND ACTIVITY CONCENTRATION

To assist in deriving a relationship between dose and activity concentration, the radium equivalent concept was used:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (1)$$

where A_{Ra} , A_{Th} and A_{K} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively. It allowed a single 'hazard index' H_{ex} to be derived from different radionuclide compositions in the coal slag and ash samples and, importantly, allowed a relationship between dose and activity concentration to be established in accordance with the PP. The measured ambient dose was subsequently converted to effective dose in order to determine the associated health impacts. A reliable relationship was established between the dose rate measured at 1 m above the soil surface and the measured activity concentrations in the samples collected from a 1 m² area at the same point. It was assumed that the predominant gamma emitting radionuclide in the residue was ^{226}Ra . Because of the health impact associated with this radionuclide, all dose rate results and activity concentrations were calculated using the dose conversion factor of 0.345 nSv/h per Bq/kg for ^{226}Ra . The activity concentration in the soil, assuming an infinite soil depth, is given by:

$$A_{Ra} \text{ (Bq/kg)} = \frac{\text{Effective ambient dose rate (nSv/h)}}{0.345 \text{ (nSv/h per Bq/kg)}} \quad (2)$$

The effective ambient dose from in situ gamma spectrometry measurements could also be estimated using the ^{238}U dose conversion factor of 0.45 nSv/h per Bq/kg. Data reported in the literature [6, 7] support that assumption, because all radionuclides of natural origin contribute, to some extent, to the total effective dose. Dose calculations were performed assuming, firstly, that the effective dose was entirely attributable to ^{238}U and, secondly, that it was entirely attributable to ^{226}Ra . Since the dose conversion factor for ^{226}Ra is smaller than that for ^{238}U , the effective dose calculated using ^{226}Ra will always be overestimated to a certain degree. This gives confidence in the approach taken, namely that the measured dose rates (using the fast GPS based method in the field) can be taken as a benchmark value for a decision making process, even if only a small number of in situ measurements are made and a limited number of samples are subjected to a more accurate laboratory analysis.

5. FINAL RADIOACTIVITY MAP OF THE SITE

The final radioactivity map of the residue site was generated by superimposing the ambient dose rate values on a real geodesic map of the site. This allowed additional modelling and/or calculations of the ambient dose rates to be reliably assigned to the residue surface or volume of interest. The final radioactivity map (see Fig. 5) shows the ^{226}Ra activity concentrations determined on the assumption that the effective dose is entirely attributable to this radionuclide. Dose rates corresponding to the activity concentrations measured in situ are shown in Table 1. The ‘in situ gamma’ (IG) sampling locations were selected from the initial, rough radioactivity map (see Fig. 4). All possible contributions to the total effective dose from radionuclides of natural origin were calculated.

Some selected, final conversion values between the measured ambient dose rates and activity concentration are presented in Table 2. Assuming a maximum dose rate of 800 nSv/h and taking the more conservative conversion factor based on ^{226}Ra , the estimated maximum value of ^{226}Ra concentration was 2318 Bq/kg. It is obvious that pockets of higher activity concentration exist in the coal sludge and ash at the investigated site (see Fig. 5). Table 2 shows the ^{238}U and ^{226}Ra contributions to the total effective dose, estimated assuming that those two radionuclides were the only ones of interest and that the presence of each in turn was 100% in the soil. The reason for such an assumption lies in the fact that ^{238}U is the parent of the decay series and ^{226}Ra is the progeny with the greatest health impact.

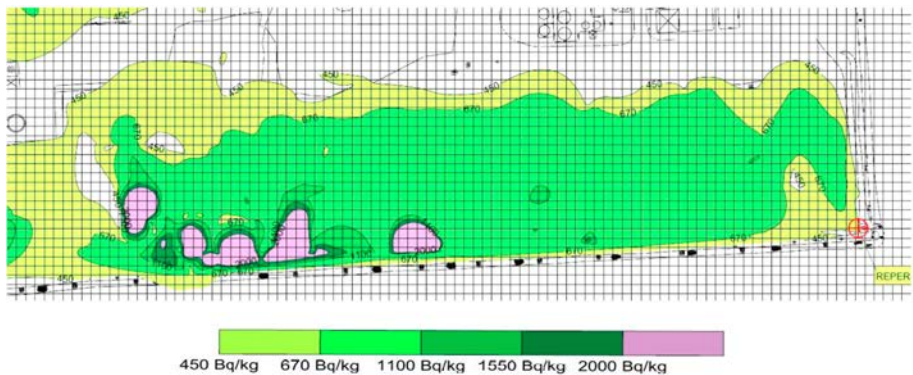


FIG. 5. Final radioactivity map generated using the ^{226}Ra dose conversion factor.

HAZARD ASSESSMENT OF A FORMER COAL FIRED POWER PLANT

TABLE 1. DOSE RATES CORRESPONDING TO MEASURED ACTIVITY CONCENTRATIONS

In situ gamma location code	Altitude (m)	Area	Dose rate (nSv/h)				
			Total	Contributions from U, Th, K			
				U series	Th series	K-40	Combined
IG18	3.48	D	79	27	16	5	47
IG19	3.54	D	557	366	46	5	417
IG20	4.01	D	608	409	61	6	475
IG21	4.33	D	535	379	40	4	422
IG24	3.95	D	287	265	24	3	292
IG25	4.25	D	279	231	33	6	270
IG28	5.76	Background	68	30	23	5	59

TABLE 2. ESTIMATED MAXIMUM ACTIVITY CONCENTRATIONS IN SLUDGE AND ASH DEPOSITS FOR VARIOUS DOSE RATES

Ambient dose rate at 1 m above the ground (nSv/h)	Activity concentration (Bq/kg)	
	Assuming dose is due to U-238	Assuming dose is due to Ra-226
200	445	580
300	666	870
400	889	1159
500	1111	1449
639	1420	1850
800	1778	2318

The calculations made for the radioactivity map were purposely done for the maximum activity concentrations of ^{238}U and ^{226}Ra rather than the averages. This means that the activity concentrations assigned to the high activity pockets on the radioactivity map (see Fig. 5) are overestimated. The surface areas of the pockets will also be overestimated, leading to an overestimation of the volumes of sludge and ash with possible enhanced activity concentration which have to be remediated or processed for by-product use. This constitutes a conservative approach from a radiation protection point of view. In reality, the calculated volume of slag and ash will not have the same enhanced activity concentrations distributed homogeneously over the depth of pockets. On the other hand, it is possible that, despite the use of a fine geodesic mesh size (up to 2 m resolution in the field at the surface), the ambient dose rate measurement equipment was unable to detect some of the small pockets of enhanced activity concentrations located at depth. In future, the collimator technique for enhancing the detection resolution is to be introduced and used for ambient dose rate measurements.

ACKNOWLEDGEMENTS

The in situ gamma spectrometry and overall sampling and analysis were done by our colleagues J. Sencar, G. Marovic and B. Skoko. A contribution to a model of fast ambient dose rate measurements was made by Z. Vucic. All the results presented are reported to a responsible governmental organization in terms of a contract for carrying out the radiological characterization of this site [8].

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WORKER EXPOSURE AT A COPPER MINING AND BENEFICIATION FACILITY

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Abstract

Palabora Mining Company is a fully integrated copper producer, dealing with mining, beneficiation, smelting, refining and casting processes. It has held a nuclear authorization since 1993 and has a well developed and mature radiation protection programme in place. The object of the paper is to report on the outcome of the most recent worker dose assessment, including a comparison with a previous study. It also provides some views on the determination of future monitoring protocols and reflects on the programme within the broader framework of an integrated risk management programme.

1. INTRODUCTION

Palabora Mining Company is a fully integrated copper producer, dealing with mining, beneficiation, smelting, refining and casting processes. The company embarked on its radiation protection programme approximately twenty years ago and has entrenched the management of NORM in its integrated risk management framework. The first worker exposure assessment was conducted in the early to mid-1990s and the two most recent assessments were carried out in 2008 and 2012 [1]. An integrated facility of this nature poses challenging process variations when compared with nuclear installations. First, the size of the operation poses a challenge to conventional monitoring strategies (i.e. 1 m × 1 m measurement matrices), while secondly it is also necessary to recognize the physical characteristics of the source, namely low activity levels but with uniform distribution within a particular process. There is thus a different balance between the level of risk and the utilization of resources to ensure an effective strategy.

1.1. Geology and radionuclide activity concentrations

The dominating rock type in the Phalaborwa area, more than 3000 million years old, is granite–gneiss of the Archaic Complex. Intrusive in this are younger rock types of the Phalaborwa Igneous Complex. Inclusions of serpentine, talc

and amphibole schist are found in the granite–gneiss and igneous rock. The main mineral content of the ore is as follows:

- (a) Apatite, $\text{Ca}_2(\text{PO}_4)_3\text{F}$;
- (b) Magnetite, Fe_3O_3 ;
- (c) Phlogopite, $\text{KMg}_3(\text{AlSi}_3\text{O}_{10})(\text{FOH})_2$;
- (d) Copper sulphide, CuS ;
- (e) Baddeleyite, ZrO_2 .

Typical average radionuclide activity concentrations in process materials are shown in Table 1.

1.2. Process description

In past years, the ore was removed from an open pit. Currently, the remaining ore body lies below the open pit and is mined using the block caving method. The ore is beneficiated by crushing, milling and flotation. Then it is melted and refined and the copper product is cast into rods. A diagram of the mining and mineral processing operation is shown in Fig. 1.

TABLE 1. TYPICAL ACTIVITY CONCENTRATIONS IN PROCESS MATERIALS

	Activity concentration (Bq/g)		
	Copper concentrate	Magnetite	Copper extraction tailings
U-238	1.43	0.14	3.52
Ra-226	1.14	0.14	1.81
Pb-210	0.56	0.08	8.84
Th-232	0.56	0.11	2.09
Ra-228	1.04	0.16	1.60
Th-228, Ra-224	1.04	0.16	1.60

WORKER EXPOSURE AT A COPPER MINING AND BENEFICIATION FACILITY

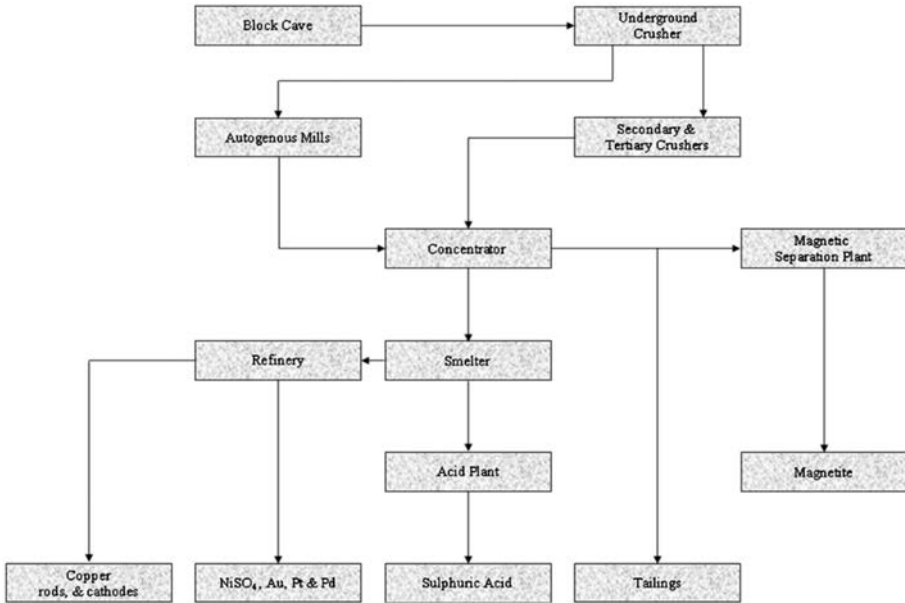


FIG. 1. Mining and mineral processing operation.

2. SURVEY METHODOLOGY

For the purposes of dose assessment, data collected through the routine monitoring programme are complemented by a single area gamma dose rate survey of the mine and works. The dose rate measurements emphasize the homogeneous distribution of the radionuclide content within particular processes, with a standard deviation for a 5 m × 5 m matrix of less than 0.07 $\mu\text{Sv/h}$. A summary of the survey methodology is given in Table 2. This methodology has several advantages:

- It better reflects the true status of the area as it does not reflect a 'once off' assessment;
- More data points represent a better statistical average;
- It provides a greater opportunity to identify abnormal conditions.

TABLE 2. SURVEY METHODOLOGY

	Type of measurement	Equipment used	Survey strategy	
			Routine programme	'Once off'
Gamma dose rate	Area	Electra GM+	X	X
Inhalation of airborne dust	Personal	Personal air samplers	X	
Inhalation of radon and thoron progeny	Area	Track etch cup	X	

3. DETERMINATION OF FUTURE MONITORING STRATEGY

Guidance is usually provided on the monitoring frequency for a particular type of classified area, for example continuous use of personal dosimeters in a controlled area with a high gamma component. The statistical value to use, however, is very often ambiguous. A possible decision matrix is shown in Fig. 2.

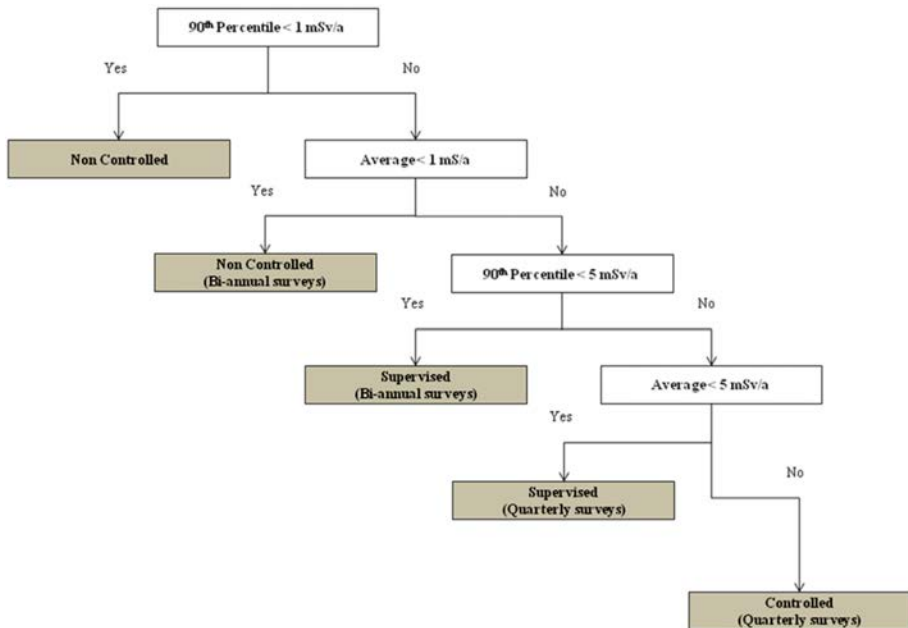


FIG. 2. Possible decision matrix.

The decision matrix in Fig. 2 has the following advantages:

- (a) It is not too conservative.
- (b) It takes cognizance of the potential of an area.
- (c) It provides for a structured escalation (increased monitoring) in terms of protection.
- (d) Protection and economics are both considered.
- (e) It is aligned with the ALARA (as low as reasonably achievable) principle.

4. ASSESSMENT RESULTS

The results of the worker dose assessment are summarized in Table 3.

It is not fully understood and quantified why the doses in most areas had decreased from their 2008 values. Three possible options are proposed:

- (a) The 2012 data may be more representative. The data set used in 2012 does not rely on a single measurement protocol, but utilizes values obtained over a 4–5 year period and at different time intervals. It is thus more representative of the routine operating conditions. Nevertheless, it should be recognized that within this framework the 2012 assessment could potentially have shown an increase in comparison with 2008.
- (b) The radionuclide content of the ore body has changed. This possibility has not yet been investigated.
- (c) The radiation protection programme has been incorporated into the integrated management system for safety, health, environment and quality. This has created various advantages, especially in view of the fact that the company's 'zero harm' focus is included in its core business values. This focus thus drives a 'safe production' frame of mind and subsequently the desired behaviour. Radiation protection processes and procedures are also, as a consequence, subjected to regular internal and external audits other than the routine regulatory compliance inspections. This reinforces the desired culture of compliance.

TABLE 3. RESULTS OF THE DOSE ASSESSMENT

	Dose (mSv/a)			Area classification	Recommended monitoring frequency
	2008 av.	2012			
		Av.	90th percentile		
Mining					
Underground	1.89	0.84	1.33	Non-controlled	Bi-annual
VO operations	0.54	0.36	0.47	Non-controlled	Annual
Surface mining	0.92	0.67	1.00	Non-controlled	Annual
Concentrator					
Concentrator	1.76	0.90	2.62	Non-controlled	Bi-annual
Secondary crushers	0.99	0.86	1.18	Non-controlled	Bi-annual
Autogenous mills	1.07	1.02	1.57	Supervised	Bi-annual
Smelter					
Reverb	0.78	0.56	1.02	Non-controlled	Bi-annual
Converter	0.78	0.45	0.99	Non-controlled	Annual
Anode casting	0.78	0.53	1.17	Non-controlled	Bi-annual

TABLE 3. RESULTS OF THE DOSE ASSESSMENT (cont.)

	Dose (mSv/a)			Area classification	Recommended monitoring frequency
	2008 av.	2012			
		Av.	90th percentile		
Refinery					
Tankhouse	0.40	0.39	0.57	Non-controlled	Annual
Tankhouse workshop	0.40	0.25	0.26	Non-controlled	Annual
Corrosion workshop	0.40	0.22	0.24	Non-controlled	Annual
Acid plant	0.69	0.74	1.01	Non-controlled	Bi-annual
Lead burning area	0.40	0.28	0.33	Non-controlled	Annual
Rod casting	0.40	0.34	0.55	Non-controlled	Annual
Rod casting workshop	0.40	0.26	0.28	Non-controlled	Annual
Nickel plant	—	0.25	0.31	Non-controlled	Annual
Other areas					
Decontamination area	0.70	2.01	2.70	Supervised	Bi-annual
Scrapyard	0.37	1.82	2.28	Supervised	Bi-annual
Dump 4	—	—	2.90	Supervised	Bi-annual

5. MANAGEMENT SYSTEM

The practical management of radiation protection in a large mining and mineral processing operation requires a different approach than, for example, that in a nuclear power plant. Radiation exposure is but one relatively small risk when compared with other types of risk that are subject to legal requirements (e.g. conventional safety and health management, and environmental protection), each with its own subcomponents. Considering only environmental protection, its subcomponents for a facility such as Palabora Mining Company includes water management, air quality management, waste management, biodiversity and invasive plant control. Each subcomponent is subject to its own legislative requirements with each legislative requirement deserving a response proportional to the risk. A further complication is that the radiation component is inseparably infused in basically all of these components. It is thus imperative that these requirements are dealt with through an integrated process — the running of two separate systems, with potentially different fundamental approaches, for one specific aspect (i.e. water management) is almost impossible to manage effectively.

For Palabora Mining Company this meant the incorporation of the requirements of the ISO 9000 series and ISO 14000 series international standards [2, 3], as well as the requirements of the nuclear regulatory body, into a single integrated management system. The quality management requirements of the nuclear regulatory body for NORM facilities (regulatory document RD-005) [4] are aligned with the requirements of ISO 9001:2000 and with IAEA standards.¹ These codes are claimed to be compatible, although with a different emphasis, with ISO 9001:2000 focusing on meeting customer satisfaction, while the IAEA standards emphasize nuclear safety. RD-005 continues to emphasize safety, with one of the objectives being for the holder of an authorization to demonstrate the ability to consistently ensure safety to the satisfaction of the regulatory body, workers and the public. Alignment with ISO 9001:2000 is evident, for instance, through the need to seek continual improvement. The combination of the two different philosophies, however, creates challenges. To illustrate this point, some examples relating to quality management requirements are given in Table 4.

¹ INTERNATIONAL ATOMIC ENERGY AGENCY, Quality Assurance for Safety in Nuclear Power Plants and Other Nuclear Installations, IAEA Safety Series No. 50-C/SG-Q, IAEA, Vienna (1996).

TABLE 4. EXAMPLES OF QUALITY MANAGEMENT REQUIREMENTS

Nuclear regulatory body RD-005 requirement [4]		Comment
6.2(k).	Have a process to measure the extent to which its personnel are aware of the relevance and importance of their activities and how they contribute to the achievement of the quality objective	The term ‘quality objective’ becomes ambiguous where a company has ISO 9001:2000 certification, thus lessening effectiveness
7.1.	The organization shall plan and develop the processes needed for product realization. In planning the product realization, the organization shall determine the ... quality objectives and requirements for the product	Not clear how this applies when the emphasis is on radiation protection Ambiguity again present when the company has ISO 9001:2000 certification

The management of radiation safety requires a structured management system. It has been demonstrated in South Africa that specialized management systems, such as those for HIV/AIDS management, can be integrated into general management systems such as ISO 9001:2000. Integration provides unique advantages, for example: (a) it ensures a consistent response to similar levels of risk from different disciplines; (b) it optimizes available resources; and (c) provides for more frequent audits. However, it is very necessary for the radiation management system not to be too unique and detailed. Its purpose is not to try to address the management of radiation safety, but address the system to follow. Where the focus is on the system, the detail of radiation safety management is then captured typically under ‘operational control’.

Palabora Mining Company is successfully managing its radiation risk by integrating it into a single management system. This is illustrated by the reduction of doses observed when comparing the results for 2012 with those for 2008. An interesting observation is that the integration allows for non-specialists to audit the system to a very reasonable extent, providing very often a different perspective on a particular process and how it is to be addressed. A more simplified but internationally recognized management system will strengthen these controls as it would help to achieve system simplification and thus more effective compliance, it would provide for even better third party audits and, just as importantly, it would create a better understanding and acceptance by the general public.

6. CONCLUSION

The Palabora Mining Company has managed the radiation exposure of its workforce for more than twenty years. The recent assessment confirmed that no areas needed to be classified above the level of supervised area and has shown a general reduction in doses when compared with the situation in 2008. The reasons for this dose reduction require further investigation and quantification. The combination of the assessment strategy followed and the resulting monitoring strategy is pragmatic, as it balances efficient utilization of resources while still retaining conservatism. While the supporting integrated management system complies with the various management system requirements, a simplification of international standards would further enhance the protection programme.

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NORM MEASUREMENT METHODS AND STRATEGIES

(Topical Session 5)

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APPLICATION OF THE GAMMA SPECTROMETRY SOURCELESS EFFICIENCY CALIBRATION METHOD TO THE MEASUREMENT OF RADIONUCLIDES IN RARE EARTH RESIDUES

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Abstract

The paper investigates and analyses NORM residues from rare earth smelting and separation plants in Jiangsu Province using the high purity germanium gamma spectrometry sourceless efficiency calibration method which was verified by IAEA reference materials. The results show that in the rare earth residues the radioactive equilibrium of uranium and thorium decay series has been broken and the activity concentrations in the samples have obvious differences. Based on the results, the paper makes some suggestions and proposes some protective measures for the disposal of rare earth residues.

1. INTRODUCTION

China is the world's largest producer and exporter of rare earths. As an important mineral resource, rare earths have a high strategic position and a broad development prospect. However, a certain degree of radioactive environmental contamination from the hydrometallurgical processing of rare earths is inevitable. The United Nations Scientific Committee on the Effects of Atomic Radiation and the IAEA have paid much attention to exposures associated with NORM. The large amounts of NORM residues generated in the rare earths production process, containing various amounts of thorium, uranium and radium, are a big problem that needs to be solved urgently. Jiangsu has a large industry associated with rare

earth smelting, separation and consumption with about ten rare earth plants, most of which were established in the 1980s and 1990s. Processing of the raw material produces between 10 and 100 t of moderately active NORM waste annually. The latest statistics show that the accumulation of these residues in the rare earths separation plants amounts to 30 000 t, comprising about 5000 t of acid soluble residues and 25 000 t of neutralizing slag. As there is no clear way of dealing with these residues, the separation plants have to store them on site in large slag repositories. However, most of the slag repositories are close to saturation. In addition, the design and construction of the repositories may not meet relevant standards, so protection against natural disasters is quite weak. When natural disasters such as floods and earthquakes occur, various types of pollution may influence the surrounding environment. This could include various degrees of contamination of the surrounding water and environment by NORM residues that have been poorly disposed of, resulting in additional exposures of nearby workers and members of the public.

Radioactive residues, including NORM residues, are to be either approved for clearance or subject to ongoing regulatory control, depending on whether they meet the clearance criteria specified in national and international regulations and standards. These include Chinese regulations on basic protection and safety [1], radioactive waste classification [2] and management of radioactive waste [3], as well as IAEA standards on exclusion, exemption and clearance [4]. There is no doubt that accurate measurement of the activity concentration of NORM residues is a prerequisite for all treatment work.

1.1. Investigation of rare earth smelting and separation plants in Jiangsu Province

The main sources of raw materials for the rare earth smelting and separation plants in Jiangsu Province are from Guangdong, Guangxi, Ganzhou in the provinces of Jiangxi and Sichuan. The raw materials can be divided into two categories: concentrates and intermediate products. The concentrates are derived mainly from southern weathering crust ionic rare earth ore (referred to as south mine or heavy rare earth mine ore) categorized as high yttrium ore, middle yttrium ore or rich europium ore. Intermediate products include rare earth oxides, rare earth chlorides, enriched yttrium and samarium–europium–gadolinium feed solution.

The NORM residues from rare earth smelting and separation plants in Jiangsu Province are mainly divided into acid soluble residues and neutralization slags. Acid soluble residues are from the filter press slag which is generated in the dissolution of the rare earth concentrates in hydrochloric acid. They contain, among other things, iron, aluminum, silicon, rare earth oxides, barium sulphate,

radioactive elements, insoluble rare earth double salts. Rare earth smelting and separation plants in Jiangsu generally use a single rare earth feed solution which is produced from the processes of precipitation, extraction and separation with excess oxalic acid. The treatment procedure can separate rare earth elements from impurities, leaving large amounts of hydrochloric and oxalic acid in the wastewater. The wastewater is neutralized with lime, thereby generating a large amount of neutralization slag. The main ingredients of the slag are calcium oxalates and entrained water. Furthermore, small amounts of rare earth oxalate, radioactive elements, insoluble salts of heavy metals, calcium chloride and calcium carbonate will be generated during long term storage due to weathering.

1.2. The sourceless efficiency calibration method for gamma spectrometry

The calibration method is described in Refs [5, 6]. High pressure germanium gamma spectrometry analysis is an important method in rare earth NORM waste analysis because it is non-destructive and has a high resolution. In the measurement of activity concentration, efficiency calibration is critical. At present, the efficiency curve method with standard sources is a regular efficiency calibration method. The efficiency is calibrated by using a standard gamma source with known activity, giving the full energy peak efficiency curve of such kind of standard sample. However, for routine analysis, the domestic laboratory uses a standard soil material as a gamma calibration source. Rare earth residue and soil are different in terms of their density and material composition, as shown in Table 1. If standard soil material is used for efficiency calibration, the analysis of rare earth residue will be subject to greater uncertainty.

TABLE 1. DENSITY AND COMPOSITION DIFFERENCES BETWEEN RARE EARTH RESIDUES AND SOIL

	No. of samples	Density (g/cm ³)		Composition
		Range	Av.	
Acid soluble residue	10	0.61–1.31	1.04	70% BaO
Neutralization slag	10	0.52–0.88	0.72	76% CaO
Soil	22	1.25–1.70	1.40	82% SiO

2. VERIFICATION OF THE SOURCELESS EFFICIENCY CALIBRATION METHOD

2.1. Gamma spectrometry

Canberra high purity germanium gamma spectrometry was used in this experiment. Its standard characteristics include the following:

- (a) Wide range of energies;
- (b) 49% relative efficiency;
- (c) 0.78 cps of background of the full energy peak (20keV–2MeV);
- (d) 1.48 energy resolution (^{60}Co);
- (e) 84:1 peak to Compton ratio (^{60}Co);
- (f) Less than 0.24 channels of peak drift (100 h).

Genie 2000 software was used for spectrum analysis. According to the verification report of the detector, the relative uncertainties (1 standard deviation) of the LabSOCS efficiency calibration for 50–150 keV, 150–400 keV and 400–700 keV are less than 7.1%, 6.0% and 4.3%, respectively.

2.2. Efficiency calibration

The efficiency calibration for gamma spectrometry uses Canberra LabSOCS software. The detector is characterized by using MCNP software. Then, combined with LabSOCS software, the result of the efficiency calibration of samples with different components and geometry is obtained by calculation.

2.3. Analysis method of elemental composition

First, rare earth slag is prepared in accordance with the standard sample preparation method. Second, the prepared sample is put into the standard sample container, making sure it covers the bottom of the container completely after passing through an 80 mesh sieve. Third, the sample is analysed using an S2 Ranger energy dispersive X ray fluorescence spectrometer to obtain the oxide content of the element. Finally, the amounts of elements contained in the sample are obtained by calculation.

2.4. Method for activity calculation

The sample activity, A (in Bq/kg) is given by:

$$A = \left(\frac{S}{T} - \frac{S_0}{T_0} \right) \cdot \frac{F \cdot F_2 \cdot \text{HXB}}{E \cdot Q \cdot F_1 \cdot P} \quad (1)$$

where

- S is the net area of the full energy peak;
- T is the sample measuring time(s);
- S_0 is the net area of the background full energy peak;
- T_0 is the background measuring time(s);
- F is the correction of the sampling time;
- F_1 is the correction of the shelf time;
- F_2 is the correction of the measuring time;
- HXB is the ratio of ash and fresh;
- E is the full energy peak efficiency;
- Q is the sample volume (kg);

and P is the branching ratio of the full energy peak.

2.5. Verification using IAEA reference material

Three reference materials supplied by the IAEA [7] are chosen — uranium ore (RGU-1), thorium ore (RGTh-1) and soil (IAEA-2010-03). Their composition is given in Table 2. The samples are put into polyethylene plastic boxes of size $\Phi 75 \text{ mm} \times 35 \text{ mm}$. The three standard reference materials are then placed on top of the detector. There is a cap in the detector, which gives an accurate description of the measuring sample, the shape, size and composition of container, as shown in Table 3 and Fig. 1. The results of the measurement are given in Table 4.

TABLE 2. CHEMICAL COMPOSITION OF THE STANDARD MATERIALS

	Composition (%)						
	Si	Ca	P	Al	O	Fe	H
U ore	99.5	0.1	0.1	0.1	—	—	—
Th ore	98.0	1.0	0.5	0.2	—	—	—
Soil	26.2	—	—	8.5	57.5	5.6	2.2

TABLE 3. LabSOCS MEASURING PARAMETERS OF THE SAMPLES AND SAMPLE CONTAINERS

	d1.1 (mm)	d1.2 (mm)	Material	Density (g/cm ³)
Side walls	2.5	70	Polyethylene	0.95
Bottom wall	2.5	—	Polyethylene	0.95
Sample	29	—	U ore	1.5
			Th ore	1.5
			Soil	0.8
Absorber 1	5	—	Polyethylene	0.95
Absorber 2	2	—	Dry air	0.001 29
Source detector	7	—	—	—

3. ANALYSIS OF RARE EARTH RESIDUE SAMPLES

3.1. Collection and treatment of rare earth residue samples

At each of the ten rare earth plants investigated, about 500 g of material was collected in five parts by sampling from the centre and the four corners of the rare earth residue stack. The five parts were mixed thoroughly to provide a homogenous sample. The samples were dried at 105°C until a constant mass was obtained. Each sample was then crushed and sieved through a 60 mesh screen before introducing into the $\Phi 75$ mm \times 35 mm polyethylene sampling container and sealing with paraffin not less than 20 days before measurement.

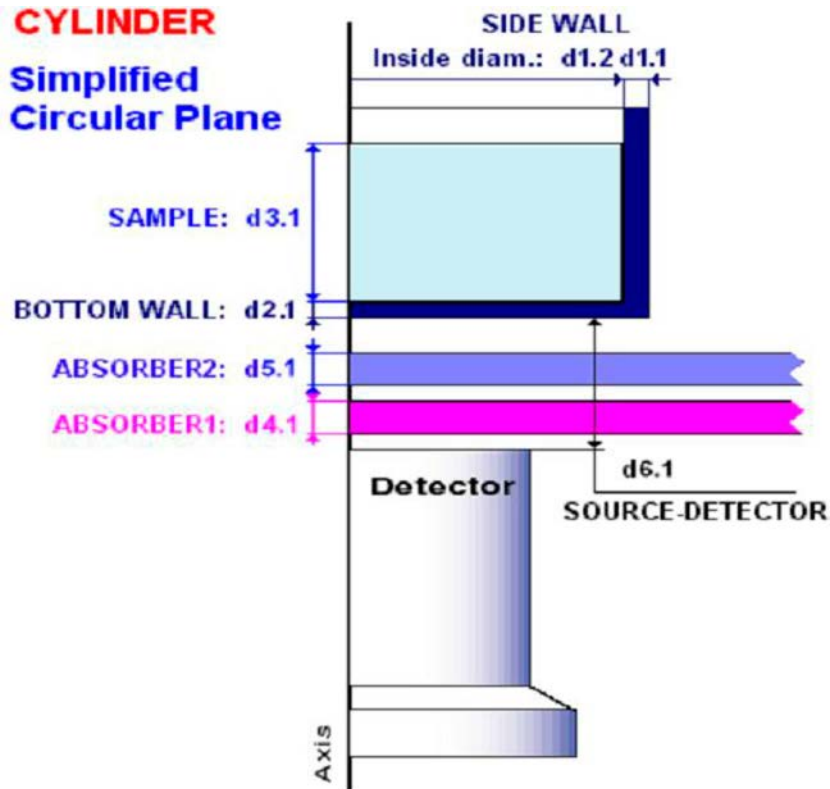


FIG. 1. LabSOCS description of measuring samples.

3.2. Sourceless efficiency calculation of the residues

The samples were analysed by the X ray fluorescence spectrometer (S2 Ranger), thus obtaining the element oxide composition. The element composition was obtained by calculation. The compositions of the acid soluble residues and neutralization slags from the ten rare earth plants are shown in Figs 2 and 3. To describe the 20 samples of acid soluble residue and neutralization slag with the sourceless efficiency calibration, the full peak efficiency (EI) curve of each sample could be plotted by the LabSOCS software.

3.3. Radionuclide analysis of the rare earth residues in Jingsu Province

The radionuclide compositions of the residue samples, determined from the results of the efficiency fitting using the sourceless efficiency calibration method, are given in Tables 5 and 6.

TABLE 4. RESULT OF LabSOCS ANALYSIS OF THE STANDARD SOURCES

	IAEA		Measured		Relative bias (%)	Z score	U score	Ratio, measured to IAEA	Trueness	Precision
	Value	Uncertainty	Value	Uncertainty						
U-238	4 960	25	5 388	259	8.6	0.43	1.64	1.09	A	A
Th-232	3 250	78	3 087	161	-5.0	-0.25	0.91	0.95	A	A
Ra-226	19 050	260	18 071	1 122	-5.1	-0.26	0.85	0.95	A	A

Source: See Ref. [8].

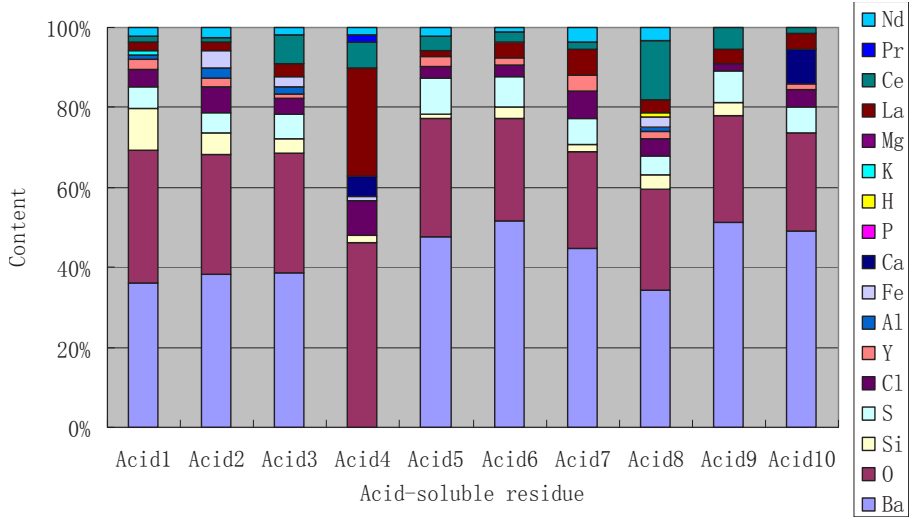


FIG. 2. Composition of acid soluble residue from ten rare earth plants.

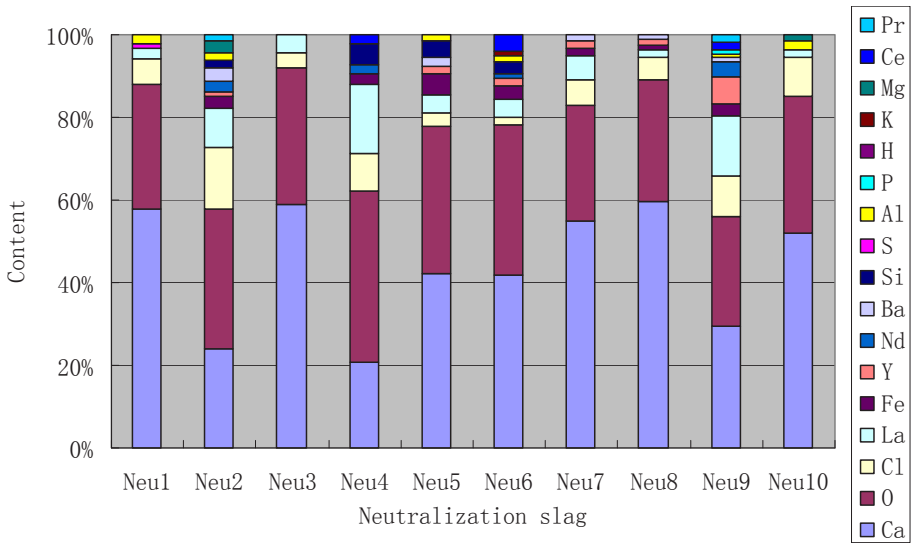


FIG. 3. Composition of neutralization slag from ten rare earth plants.

TABLE 5. RADIONUCLIDE COMPOSITION OF ACID SOLUBLE RESIDUE SAMPLES FROM TEN RARE EARTH PLANTS

	Activity concentration (Bq/g)									
	Sample 1 78 g	Sample 2 143 g	Sample 3 143 g	Sample 4 108 g	Sample 5 162 g	Sample 6 147 g	Sample 7 168 g	Sample 8 118 g	Sample 9 122 g	Sample 10 141 g
Th-232 series										
Tl-208	1.1±0.1	2.3±0.3	25.5±3.8	0.006±0.001	4.2±0.5	2.0±0.3	1.4±0.2	5.8±0.6	12.9±1.5	0.3±0.1
Pb-212	4.5±0.8	9.11±1.22	77.6±8.5	1.2±0.2	13.3±1.3	7.2±0.8	5.0±0.6	25.1±3.2	40.7±5.3	2.6±0.3
Bi-212	4.6±0.5	7.47±0.98	96.9±13.5	0.02±0.003	13.9±1.5	7.5±0.9	4.9±0.6	18.2±2.5	45.5±5.6	1.2±0.2
Ra-224	8.9±1.2	8.0±0.9	66.8±8.1	0.3±0.05	13.6±2.4	6.8±0.8	4.7±0.6	19.0±2.3	64.8±7.9	2.1±0.3
Ac-228	12.3±1.5	3.6±0.4	141±9.2	0.7±0.1	5.1±0.7	8.8±1.1	4.1±0.5	3.6±0.4	36.1±4.5	2.8±0.3
U-238 series										
Pb-214	4.8±0.6	0.5±0.1	35.4±5.2	0.1±0.02	3.1±0.5	1.5±0.3	0.8±0.1	0.4±0.1	33.3±4.5	1.1±0.3
Bi-214	4.4±0.6	0.5±0.1	35.1±4.5	0.03±0.01	2.9±0.3	1.3±0.2	0.7±0.2	0.4±0.1	31.7±5.2	1.0±0.2
Th-234	1.2±0.6	1.9±0.3	2.3±0.3	0.1±0.02	4.8±0.6	0.9±0.2	0.9±0.2	3.7±0.5	1.4±0.2	0.4±0.1
U-235 series										
Th-227	2.2±0.3	6.6±0.7	1.13±0.2	3.8±0.5	2.6±0.4	1.6±0.3	1.1±0.2	13.1±2.1	2.4±0.4	2.9±0.5
La-138	0.04±0.01	0.02±0.004	<0.01	0.2±0.04	0.02±0.004	0.03±0.005	0.02±0.004	<0.01	0.06±0.01	0.01±0.001

TABLE 6. RADIONUCLIDE COMPOSITION OF NEUTRALIZATION SLAG SAMPLES FROM TEN RARE EARTH PLANTS

	Activity concentration (Bq/g)									
	Sample 1 93 g	Sample 2 98 g	Sample 3 108 g	Sample 4 113 g	Sample 5 93 g	Sample 6 85 g	Sample 7 74 g	Sample 8 66 g	Sample 9 84 g	Sample 10 105 g
Th-232 series										
Tl-208	0.2 ± 0.03	0.02 ± 0.003	0.4 ± 0.1	0.01 ± 0.002	0.04 ± 0.01	0.01 ± 0.003	0.03 ± 0.01	0.009 ± 0.002	0.06 ± 0.01	0.03 ± 0.01
Pb-212	0.7 ± 0.1	2.2 ± 0.3	1.6 ± 0.2	0.8 ± 0.1	0.7 ± 0.1	0.09 ± 0.01	0.5 ± 0.06	0.4 ± 0.05	1.8 ± 0.2	0.4 ± 0.04
Bi-212	0.6 ± 0.08	0.07 ± 0.01	1.6 ± 0.2	0.03 ± 0.003	0.2 ± 0.02	0.04 ± 0.005	0.08 ± 0.01	0.03 ± 0.003	0.2 ± 0.02	0.1 ± 0.01
Ra-224	0.5 ± 0.06	1.0 ± 0.1	1.7 ± 0.3	0.4 ± 0.05	0.3 ± 0.03	0.04 ± 0.004	0.1 ± 0.01	0.4 ± 0.04	0.2 ± 0.02	0.1 ± 0.01
Ac-228	0.08 ± 0.01	1.1 ± 0.1	4.8 ± 0.6	0.5 ± 0.05	0.3 ± 0.03	0.05 ± 0.005	0.2 ± 0.02	0.2 ± 0.03	0.4 ± 0.04	0.3 ± 0.03
U-238 series										
Pb-214	0.01 ± 0.001	0.1 ± 0.01	0.9 ± 0.1	0.03 ± 0.003	0.09 ± 0.01	0.02 ± 0.002	0.04 ± 0.005	0.03 ± 0.003	0.08 ± 0.01	0.04 ± 0.004
Bi-214	0.01 ± 0.001	0.1 ± 0.01	0.8 ± 0.1	0.03 ± 0.003	0.09 ± 0.01	0.02 ± 0.002	0.04 ± 0.005	0.03 ± 0.003	0.08 ± 0.01	0.04 ± 0.004
Th-234	0.5 ± 0.06	0.5 ± 0.05	0.4 ± 0.04	0.2 ± 0.02	2.0 ± 0.3	0.1 ± 0.01	2.8 ± 0.4	0.3 ± 0.03	1.3 ± 0.2	0.5 ± 0.05
U-235 series										
Th-227	0.5 ± 0.06	4.8 ± 0.6	0.8 ± 0.1	2.7 ± 0.3	0.9 ± 0.1	0.08 ± 0.01	1.4 ± 0.2	0.8 ± 0.1	2.6 ± 0.3	0.05 ± 0.005
La-138	<0.01	0.04 ± 0.005	0.009 ± 0.001	0.07 ± 0.01	0.01 ± 0.001	0.006 ± 0.000 8	0.006 ± 0.000 8	0.003 ± 0.000 3	0.03 ± 0.003	0.002 ± 0.000 2

4. SUGGESTIONS AND CONCLUSIONS

- (1) According to the rule of IAEA Proficiency Testing, the results of sourceless efficiency calibration for uranium ore (RGU-1), thorium ore (RTh-1) and soil (IAEA-2010-03) have been validated by using the methods of U test and Z score, among others. Meanwhile, the precision and accuracy are considered acceptable.
- (2) The monitoring results show that the radionuclide concentrations of acid soluble residues are <0.01–141 Bq/g, while the radionuclide concentrations of neutralization slag residues are <0.01–4.8 Bq/g. The equilibrium in the uranium and thorium series has been destroyed. Under this state of disequilibrium, the activity concentrations of ^{238}U or ^{232}Th are not to be derived from the activity concentrations of their progeny but are to be measured individually.
- (3) In the residues from the rare earth smelting and separation plants of Jiangsu Province, the difference between the maximum and minimum radionuclide concentrations is more than 4000 times in acid soluble residues and almost 100 times in neutralization slag. This is due to differences in raw materials, production processes and waste management methods used by the rare earth producers. The main raw material is divided into two categories, rare earth concentrates and intermediate products. The activity concentrations of rare earth concentrates will also show differences in activity concentration, even if they come from the same source, because of the different purification technologies used during the ore beneficiation process. In intermediate products, such as rare earth oxides recovered from NdFeB scrap, which contains rare earth elements only, the acid soluble residue generated after removing impurities with acidic water is non-radioactive.
- (4) Most of the plants did not carry out any category management of the radioactive and non-radioactive residues according to the raw materials and production technology, which directly led to the huge differences of radioactivity levels of acid soluble residues. Similarly, most plants did not perform category management on radioactive and non-radioactive wastewater either, which led to the significant differences in the radioactivity levels of neutralization slag produced by adding lime to acidic wastewater. At the same time, the mismanagement of residues and wastewater, on the other hand, is largely due to the expansion of the amounts of radioactive solid waste from the rare earth smelting plants in Jiangsu Province.
- (5) For the rare earth producers of Jiangsu Province, they could first of all consider excluding highly radioactive rare earth concentrates from the range of raw materials used in the production process. Second, they could consider carrying out category management on radioactive residues according

to the production process and prevent the mixing of non-radioactive and radioactive residues, especially wastewater. The residues should ideally be classified and mixed only after treating separately. In addition, radioactive wastewater should ideally be treated separately.

ACKNOWLEDGEMENTS

This work has been supported as a sub-project of the national nuclear and radiation safety surveillance project by the Ministry of Environmental Protection and a sub-project of the Department of Jiangsu Environmental Protection (No. 201150).

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EVALUATION OF NORM MEASUREMENT RESULTS BY MEANS OF STATISTICAL METHODS

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Abstract

A common task is the assessment of random samples, drawn from a large population, in order to assess whether radiological criteria are met. When making measurements of NORM (e.g. residues and contaminated areas), usually by gamma spectrometry, a sample of measurement results (i.e. in becquerels per gram or becquerels per square centimetre) is generated. A test statistic (e.g. median, mean, 95th percentile and 95% upper confidence limit of the mean) is determined from the data by means of a statistical evaluation algorithm. There are the following possibilities to apply this test statistic. It can be compared with the current control limits of NORM regulations or it can be used as a basis for dose assessments. Different statistical methods for the calculation of the test statistic will be presented. The classical statistical methods are compared with the modern numerical methods. A robust, efficient and non-parametric numerical procedure was found: the so called bootstrap method with a modification: application of the bias correction and acceleration method. The various methods (classical and numerical) are applied to measured results from the practice of radiation protection. As the relevant test statistic the upper confidence limit (UCL) of the mean was defined. The UCL was estimated from several random samples of size 20 drawn from a population (244 measured values of ^{228}Ra mass related activity of a contaminated ground area). The results of application of classical and numerical statistical methods on the test examples are compared and discussed.

1. INTRODUCTION

Because of the further progressive differentiation of the comprehensive body of legislation regarding NORM, the radiological assessment of residues, contaminated ground areas and building structures, among other things, comes more and more to the fore. Figure 1 shows the principle of such a radiological assessment. There exists a NORM object which has to be evaluated, for instance in the form of a red mud deposit as can be seen in Fig. 1. In order to simplify the procedure in connection with the dose criterion (in millisieverts per year),

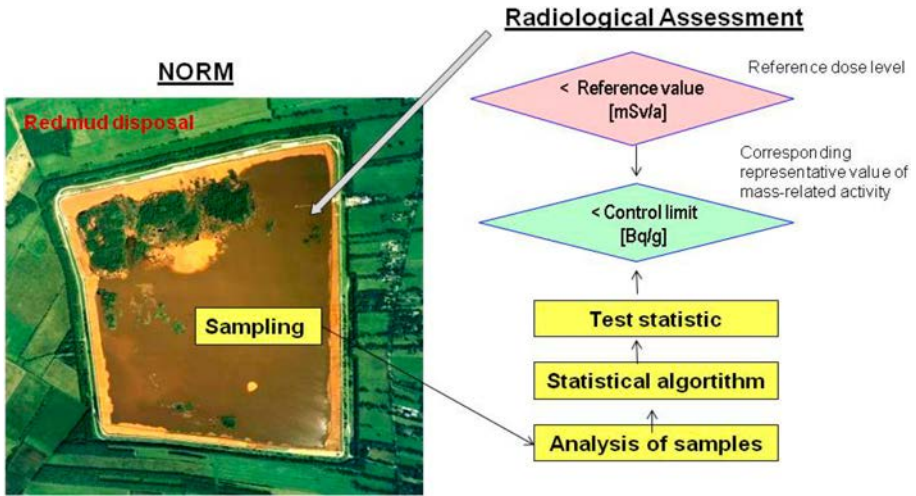


FIG. 1. Principle of the radiological assessment of NORM.

a control or monitoring limit (in becquerels per gram) is derived from it. This control limit (also exemption or clearance criteria) can be interpreted differently. For instance:

- (a) The sum of the activity concentrations;
- (b) The mean or weighted mean of the activity concentrations within a decay chain;
- (c) The sum of the activity concentrations of the maximum values of radionuclides within the respective uranium and thorium decay chain.

In a first step, values of the activity (mass related or site related) must be determined with a measuring method. That means that we have to take samples of residues or of other materials (or we measure in situ), prepare the samples, measure them and evaluate the results. From the obtained measurement results by means of a statistical algorithm, we estimate a test statistic, which is compared with the control limit. The statistical algorithm is the focus of this paper.

2. GENERAL OVERVIEW

In this section, we show some basic connections for the assessment of NORM. Figure 2 shows the basic relationships.

EVALUATION OF NORM MEASUREMENT RESULTS

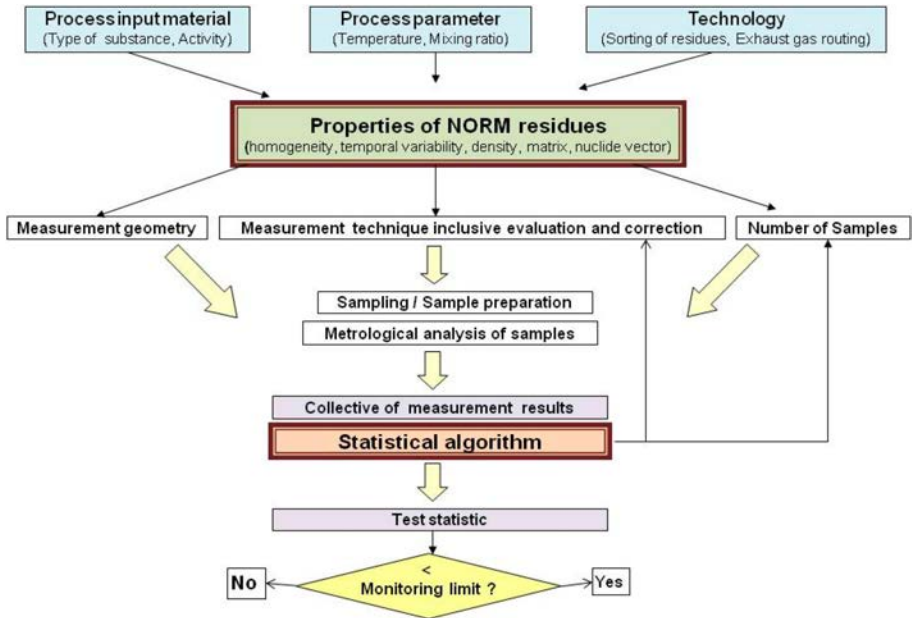


FIG. 2. Basic relationships of the radiological assessment of NORM.

The properties of NORM play a central role. These properties specify the sampling and sample preparation procedures, and there follows a simple example: the number of sampling campaigns is determined by the temporal variability and the number of samples per campaign is determined by the homogeneity. The measurement geometry (e.g. the maximum sample thickness) is determined by the matrix of residues, their density and the gamma ray lines. The measurement geometry in turn determines the required sample preparation, such as milling and homogenizing.

After sampling and sample preparation, we obtain through measurement, usually by gamma spectrometry, a collection of measurement results. A test statistic from the collective of measured values is determined by a statistical evaluation algorithm. At the end, the test statistic is compared with the control limits. For the statistical evaluation, the temporal variability and homogeneity of the charge and the nuclide vector are especially relevant. In Fig. 3, several kinds of radionuclide vectors are compared. There are examples of relatively constant vectors such as, in Fig. 3, the sludge from the copper shale smelting and the dust from the smelting of iron. Exactly the opposite is seen at the scale samples where we can observe highly variable vectors. In the last diagram in Fig. 3, we see unusual activity ratios, shown here in an example from the niobium–tantalum industry, with very high ^{227}Ac activity. The radionuclide ratios of different types

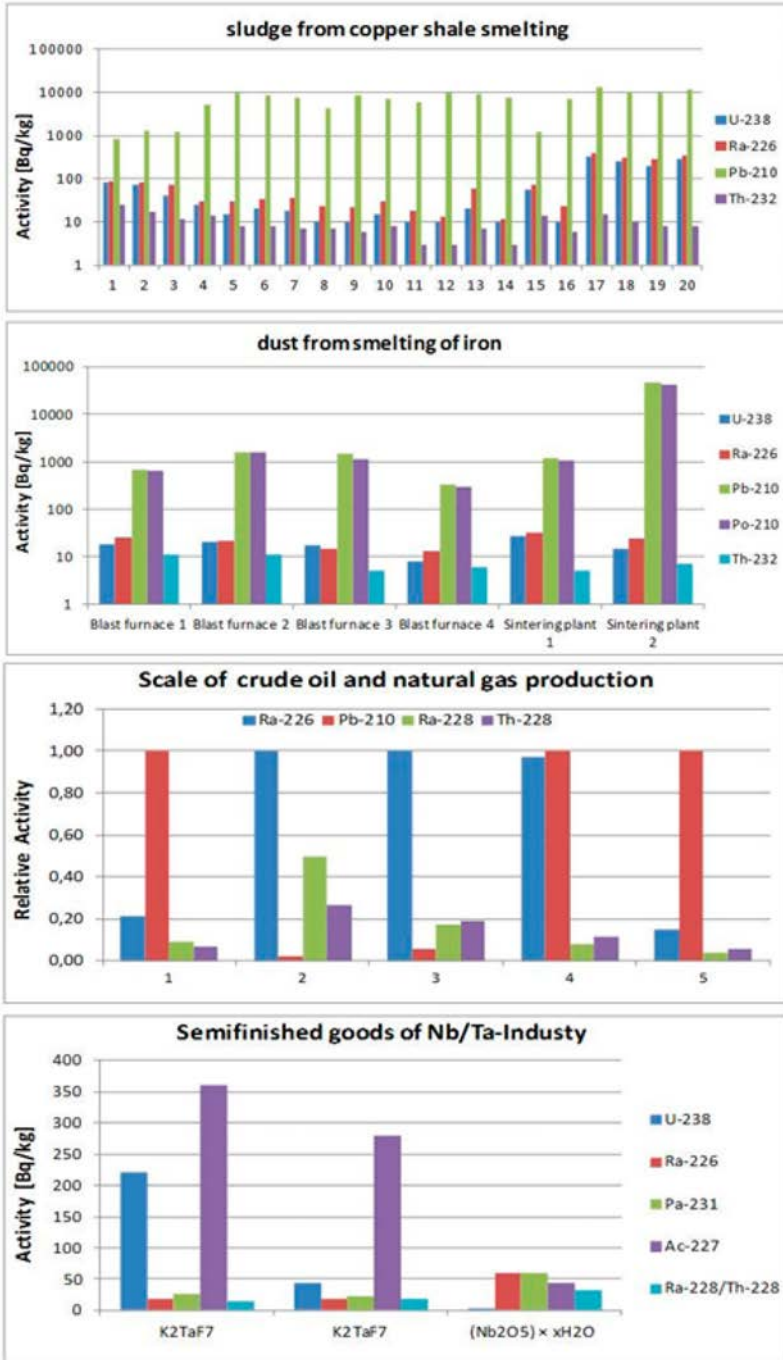


FIG. 3. Examples of several kinds of radionuclide vectors [1–4].

of NORM have a direct influence on the evaluation of NORM. If a constant vector is given, only one radionuclide in all samples need be measured and estimated and then all the other activities can be calculated. If the vector is not constant, several radionuclides must be measured and estimated.

3. PRINCIPLES FOR THE DETERMINATION OF REPRESENTATIVE VALUES

For the determination of representative values in the field of radiation protection there exist different practices. The test statistic, for instance, can be the 95th percentile, the median or the mean (see Fig. 4). Dose evaluations are mostly based on the mathematical expectation of a population. The expected value or mean of the activity concentration of a radionuclide, for example in a quantity of residues, is the relation of the whole activity of this radionuclide to the whole mass. The expected value (or expectation, mathematical expectation, mean, first moment) of a population characterizes the mean of this population. It is the weighted average of all possible values that a random variable can take (integral of the random variable with respect to its probability measure). With an infinite quantity of random variables, the expected value is described by the arithmetic mean. A very realistic, and at the same time conservative enough, estimate of the mathematical expectation of the specific activity is the upper confidence limit (UCL) for the mean. Figure 5 shows possible positions of the confidence intervals to the control limit. Case A is significantly below the limit; case D is significantly above. The use of the UCL as a test statistic has the advantage that by increasing the sample size or the measurement accuracy, the UCL can be reduced and the limit undercut (as in cases B and C).

4. STATISTICAL METHODS

Several methods can be used to estimate the mean and the confidence limits for the mean of normal or two parameter log-normal distributions. The log-normal distribution is the most commonly used probability density model for environmental contaminant data. Figure 6 shows how to obtain the mean and the upper one-sided $(1 - \alpha)$ confidence limit for the mean by the classical approach (UCL_{class}). The confidence level we usually choose is $P = 95\%$. The formulas were taken from Ref. [5].

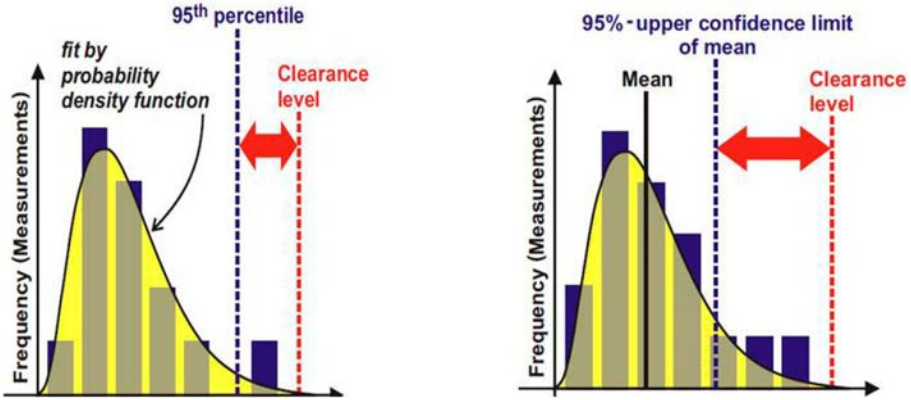


FIG. 4. Examples of several kinds of test statistics.

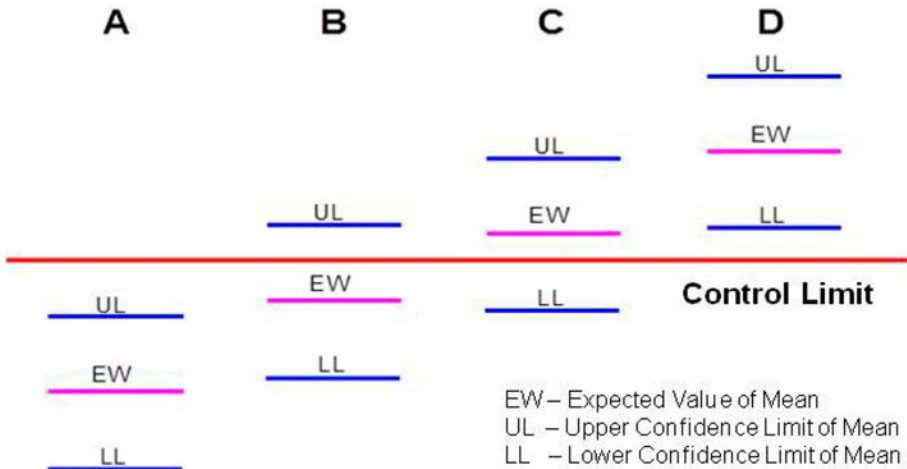


FIG. 5. Possible positions of the confidence intervals of the expected value of the mass related activity to the control limit.

In addition to classical approaches, numerical methods also exist. Bootstrapping is a numerical method for assigning measures of accuracy to sample estimates. It is particularly useful when the theoretical distribution of a statistic of interest is complicated or unknown. The bootstrap method was only recently developed because it requires modern computer power to simplify the often intricate calculations of traditional statistical theory. The bootstrap algorithm (see Fig. 7) draws many independent bootstrap samples (values drawn with replacement from x), evaluating the corresponding bootstrap replications,

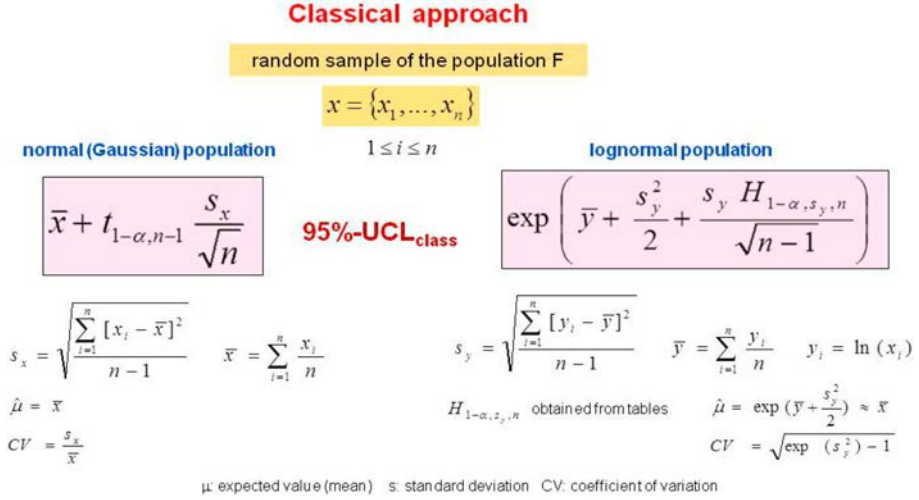


FIG. 6. Classical approach for estimating the mean and the upper confidence limit of the mean (UCL_{class}).

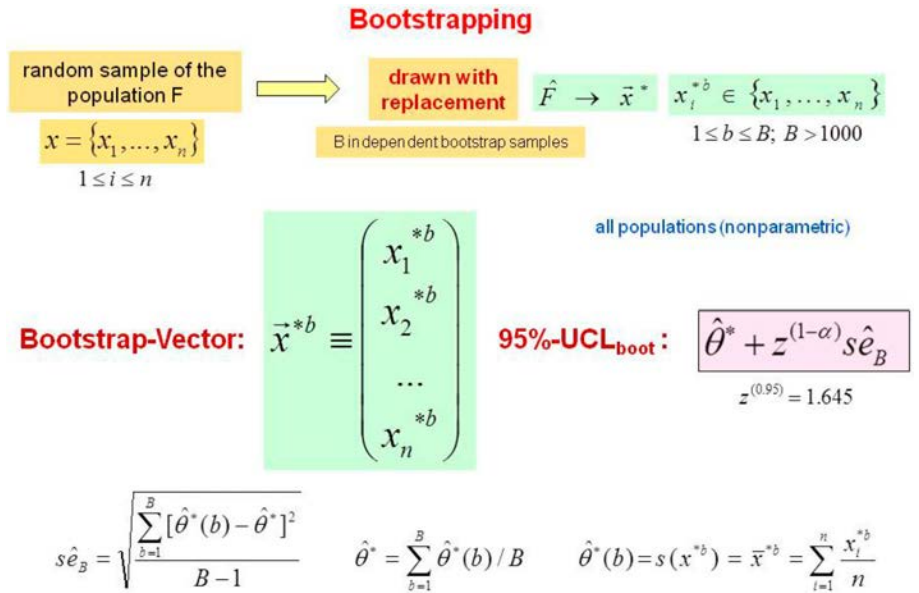


FIG. 7. Bootstrapping algorithm for estimating the mean and the upper confidence limit of the mean (UCL_{boot}).

and estimating the standard error θ by the empirical standard deviation of the replications. The result is called the bootstrap estimate of standard error, denoted by se_B , where B is the number of bootstrap samples used. The formulas were taken from Ref. [6]. In addition, the bias correction and acceleration (BCa) method introduced in Ref. [6] was used to modify the results (UCL_{BCa}).

5. APPLICATION OF THE METHOD

The bootstrap statistical algorithm was applied to a test example: 244 measured values of ^{228}Ra activity concentrations in thorium contaminated ground around an industrial building. The almost area wide population is described by 244 measurements using collimated in situ gamma spectrometry. The measuring surface amounted to 5.1 m^2 by analysis of the 911 keV gamma energy line (see Fig. 8).

All measured ^{228}Ra activities are assumed in a histogram (see Fig. 9). Because the measurements were carried out nearly area wide, we present these data as the basis of a statistical population. In Fig. 9, it can be seen that it is nearly a log-normal distribution.

From the population, ten datasets were generated. Each dataset consists of 20 data each drawn at random from a two parameter log-normal distribution with the parameter $\hat{m} \approx \bar{x} = 231 \text{ Bq/kg}$ and the coefficient of variation $VC = 1.3$. The first six datasets were generated with a random generator. The last four datasets were generated with a simulated sampling technique: random sampling within segments. The site is divided into blocks of equal size (or segments), and one or more samples are taken at random locations within each block (segment). This procedure combines systematic and random sampling and allows for a more uniform coverage than simple random sampling [5]. Areas near the building were weighted more heavily due to the simulated sampling. On each of the ten datasets, the classical and the numerical bootstrap algorithm (with $B = 1000$), modified with the BCa method was applied.

6. RESULTS

The results of application of different statistical algorithms (classical and numerical) are shown in Fig. 10. It is presented using the 95% UCL of the estimation of the expected value (mean) as a representative value of the ^{228}Ra activity concentration. It should be noted that a random sample of 20 is

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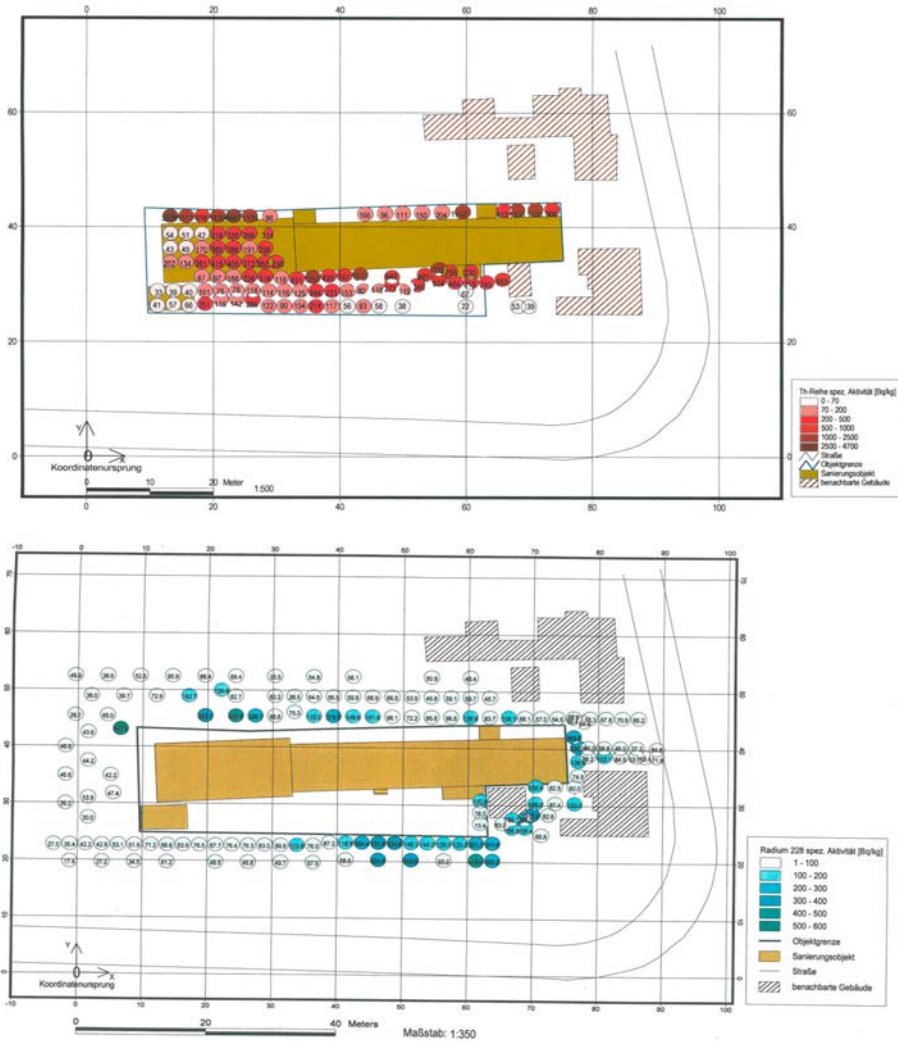


FIG. 8. Results of measurements on a thorium contaminated ground area.

too small for a population with a high coefficient of variation ($CV = 1.3$) and therefore the results have a high variance. Nevertheless, the results using the bootstrap algorithm are closer to the ‘true value’ (231 Bq/kg). They also have a lower variance. It is statistically tolerable that one confidence level (#1) of $P = 95\%$ is lower than the mean.

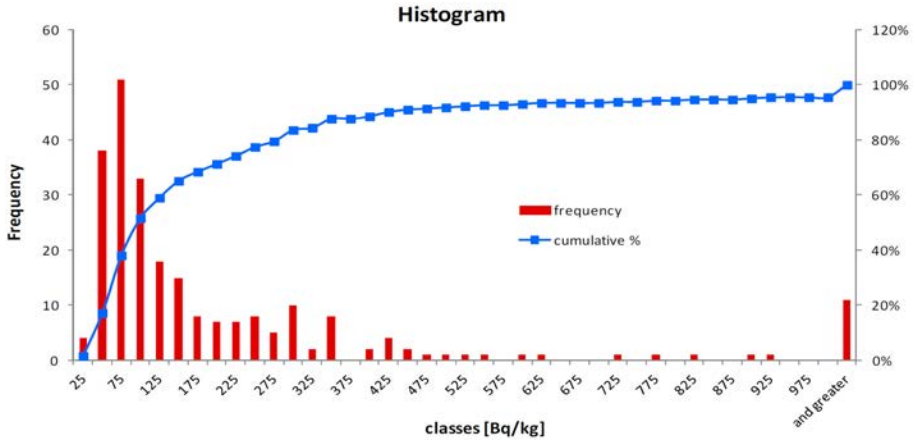


FIG. 9. Histogram of measurement results on a thorium contaminated ground area (population).

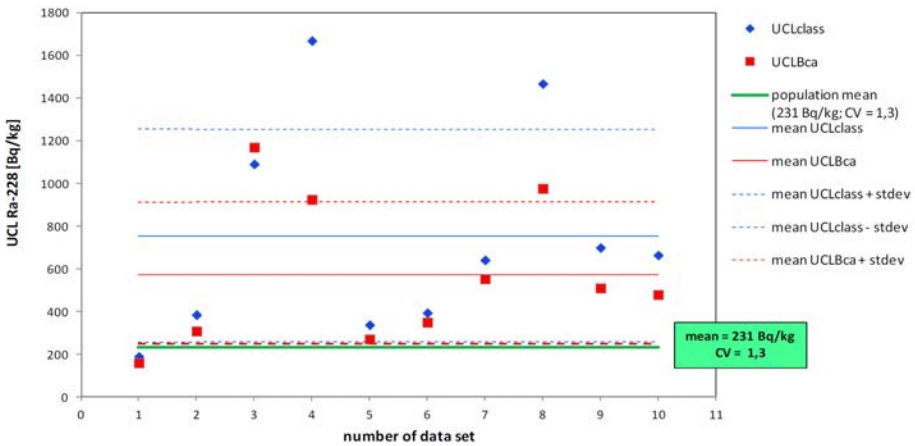


FIG. 10. Results of application of different statistical methods (classical method and bootstrapping) for generating a test statistic (upper confidence limit of mean).

7. CONCLUSIONS

The main disadvantage of the classical method is that it finds application only to known distributions, such as normal and log-normal distributed populations. Through this work, a statistical method with the following properties was found:

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- (a) Consistent procedure for all applications (even independent of the coefficient of variation);
- (b) Fundamental suitability to test the 95% VG;
- (c) Parameter free method (no proof of the assumed probability density function is necessary);
- (d) Efficient method, which means fast convergence of the calculated estimates to the actual parameters;
- (e) Robust method that generates plausible results;
- (f) Type B uncertainties can be taken into account.

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ANOMALIES IN NATURAL BACKGROUND LEVELS ASSOCIATED WITH MINERALS WITH ELEVATED RADIONUCLIDE CONCENTRATIONS

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Abstract

In the paper, the results of terrestrial natural background measurements in seven Chinese provinces are discussed. They have been generated as part of a mapping system based on sampling grids of 4 and 16 km² to provide geochemical data for uranium, thorium and potassium, and complemented by gamma dose rate measurements 1 m above the ground. Points of elevated dose rates were identified as being due to human activities involving minerals with elevated activity concentrations. The number of these instances, and thus the general level of background radiation, is increasing. The result of the analysis indicates that control of activities involving such minerals needs to be considered.

1. INTRODUCTION

Human exposure to radiation from both natural and artificial background radiation varies by location in the world. Natural radiation is ubiquitous. According to 1990 survey data in China, natural background radiation contributes 92.8% to the total dose received by humans. If the impact of human activities involving minerals with elevated activity concentrations is taken into account, the contribution of radiation from natural sources to total exposure reaches 95.62% [1]. If the concentrations of radionuclides of natural origin in a material

exceed certain levels, control of exposure needs to be considered in order to protect against possible health effects and such material might then have to be regulated as NORM. Enhanced radiation exposure from natural sources arises from industries associated with the exploitation and use of minerals and raw materials. In recent years, human activities involving minerals are increasing, so some effective rules and regulations should ideally be set up and brought into the regulatory system.

According to the ‘technical regulations on monitoring based on radioactive sources’, so called ‘associated minerals’ are defined as follows [2]:

- (a) The concentration of uranium and thorium radionuclides in the mineral or main raw material is more than 0.1 Bq/g.
- (b) The gamma dose rate 1 m from the surface of the mineral, raw material or solid waste is more than 50 nGy/h above the local natural background level.

As long as one of the above conditions is satisfied, the mineral is an ‘associated mineral’, the development and utilization of which can cause the radionuclides in the ground to be brought to the surface. Enrichment and migration of the radionuclides may increase the radiation dose received by workers and nearby residents by introducing an additional radiation dose from natural sources. In China, the natural background level is the terrestrial gamma radiation dose rate before any ‘associated mineral’ is developed and used. Natural background data are recorded in the Regional Geochemistry — National Reconnaissance (RGNR) project. The RGNR project (also called China’s National Geochemical Mapping Project) was initiated in 1978. It has covered 6.8 million km² of China’s territory during the past 32 years. It has turned out to be one of the most successful geochemical mapping projects in the world. The RGNR project provides the geochemical data for uranium, thorium and potassium — one sample data for every 4 km² in general, 1–2 original samples in 1 km² and rock samples collected in rock outcrop regions. In addition, related information at sampling sites and quality control data for resampling and standard sample control in the laboratory are analysed. This research will estimate the terrestrial gamma dose rate in Guangdong as well as Guangxi provinces.

2. PRELIMINARY MAPPING OF TERRESTRIAL GAMMA DOSE RATE IN SOME PROVINCES

The terrestrial gamma dose rate was calculated from the geochemical data of uranium, thorium and potassium, with one data sample chosen in 4 km².

(At least one sample is taken in 1 km^2 and then the data are combined into a sample). The analysis results of the sample compositions are compiled into a map. The sample numbering method in the field, based on a large grid (including four small actual sample grids), is shown in Fig. 1. The processing and analysis flow chart of the sample combination (including four small actual sample grids) is shown in Fig. 2. The average value and the background value of terrestrial gamma dose rate obtained from the low density grid ($2 \text{ km} \times 2 \text{ km}$) and the very low density grid ($4 \text{ km} \times 4 \text{ km}$) gave similar results, showing that the analysis technique for regional geochemical exploration is reliable (see Table 1).

High background dose rates are defined as 300 nGy/h in Ref. [3]. This value is about five times the world average value of background radiation. Some locations with a dose rate exceeding this level were found in the very low density grid ($4 \text{ km} \times 4 \text{ km}$) and more locations were found in the low density grid ($2 \text{ km} \times 2 \text{ km}$). The results are shown in Table 1.

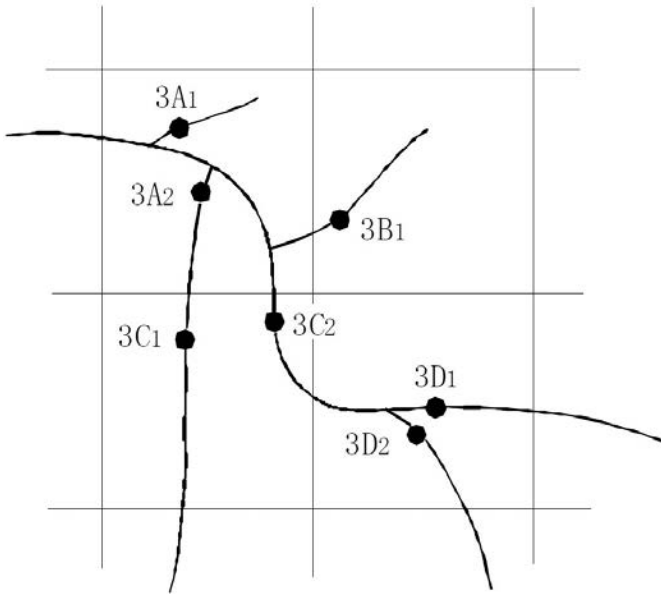


FIG. 1. The sample numbering method in the field based on the large grid (including four small actual sample grids).

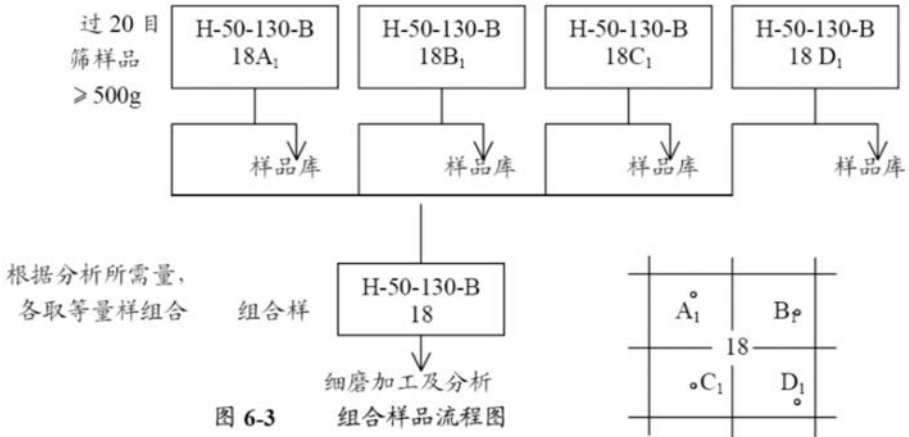


FIG. 2. The processing and analysis flow chart of sample combination (including four small actual sample grids).

3. RELATIONSHIP BETWEEN THE ANOMALY DOSE RATE AND GEOLOGIC UNITS IN GUANGXI

Depending on the source of the mineral, the presence of elevated background radiation levels in Guangxi can be due to three possible causes:

- (a) The mining of local ‘associated minerals’;
- (b) The use of local ‘associated minerals’;
- (c) The use of non-local ‘associated minerals’.

The geological map shown in Fig. 3 indicates that 131 anomaly points of ‘associated minerals’ exist in Guangxi [4]. Ninety percent of them are not included among the very low density grid (4 km × 4 km) and low density grid (2 km × 2 km) anomaly points. That is to say, 117 anomaly points of ‘associated minerals’ are newly found. The finding that, due to the emergence of new anomaly points, Guangxi and the other six provinces showed higher natural background levels than those in other areas, is not unexpected. However, it is worth mentioning that the increase in new anomaly points is more than 70%. The increase in the terrestrial natural background levels is caused mainly by the use of non-local ‘associated minerals’.

TABLE 1. DOSE RATES DETERMINED FROM REGIONAL GEOCHEMICAL DATA AND DIRECT MEASUREMENTS

Province	Method	No. of samples	Dose rate (nGy/h)				No. of anomaly points		
			Min.	Max.	Av.	Standard deviation	Anomaly threshold ^a	Total	>300 nGy/h
Guang Dong	Calculated (4 km × 4 km) ^b	10 941	15	558.8	114.77	59.43	290	134	105
	Calculated (2 km × 2 km)	45 289	8.2	9 436.9	115.49	89.73	385	343	343
	Measured ^c	900	17.1	281.4	97.14	39.62	215	12	0
Guang Xi	Calculated (4 km × 4 km)	14 770	21.3	447.1	71.13	30.80	165	275	42
	Calculated (2 km × 2 km)	60 767	5.8	1 373.7	73.32	41.78	200	1 017	341
	Measured	882	6.9	276.5	64.29	32.61	160	16	0
Gui Zhou	Calculated (4 km × 4 km)	10 884	30.3	140.3	76.40	12.22	113	40	0
	Calculated (2 km × 2 km)	46 004	9.6	263.3	68.06	16.34	120	108	0
	Measured	248	13.1	142.2	63.67	20.26	125	1	0
Hu Bei	Calculated (4 km × 4 km)	9 397	27.9	238.5	64.43	15.62	110	67	0
	Calculated (2 km × 2 km)	35 275	9.7	437.4	64.86	21.39	130	321	13
	Measured	559	10.9	170.5	60.24	17.34	110	9	0

TABLE 1. DOSE RATES DETERMINED FROM REGIONAL GEOCHEMICAL DATA AND DIRECT MEASUREMENTS (cont.)

Province	Method	No. of samples	Dose rate (nGy/h)				No. of anomaly points		
			Min.	Max.	Av.	Standard deviation	Anomaly threshold ^a	Total	>300 nGy/h
Hu Nan	Calculated (4 km × 4 km)	12 954	43.5	326.3	78.15	27.25	160	323	4
	Calculated (2 km × 2 km)	51 893	5.6	748.0	79.51	35.76	190	1 163	150
	Measured	1 321	21	271.2	69.14	21.87	135	19	0
Shan Dong	Calculated (4 km × 4 km)	5 750	35	169.4	58.30	13.4	100	137	0
	Calculated (2 km × 2 km)	19 968	21.7	353.1	59.18	19.39	120	469	3
	Measured	1 856	16.9	162.6	56.40	13.9	100	25	0
Shan Xi	Calculated (4 km × 4 km)	4 979	43.5	140.6	59.70	8	85	71	0
	Calculated (2 km × 2 km)	13 468	21.3	411.3	60.75	13.07	100	186	2
	Measured	359	31.1	85.7	53.09	9.60	80	3	0

^a The anomaly threshold is the arithmetic mean plus three standard deviations (rounded).

^b Calculated using the Beck formula recommended by UNSCEAR in 1982 in Guanxi and Shangxi provinces.

^c Obtained from the Radiation Monitoring Technology Centre, Ministry of Environmental Protection.

ANOMALIES IN NATURAL BACKGROUND LEVELS



Note: The field measured gamma data of abnormal points was obtained from the Guangxi radiation environmental monitoring station.

FIG. 3. The relationship between the anomaly sites of terrestrial gamma radiation dose rate and geologic units in Guangxi.

4. CONCLUSION

Natural background data in the form of terrestrial gamma radiation dose rates have been gathered since before the emergence of activities involving minerals with elevated activity concentrations ('associated minerals'). The present situation in Guangxi and six other provinces, showing higher levels of background radiation than in other areas is not unexpected. However, it is worth mentioning that the increase in the number of anomaly points due to these 'associated minerals' is more than 70%. This implies that there are still many problems with the management of these minerals, requiring central government policy and supporting scientific research. The regulatory body should consider paying more attention to the problem and strengthening its supervision. A comprehensive work programme should ideally be carried out to ensure the

reliability of local monitoring data and avoid the increasing trend of regions with highly elevated radiation levels.

ACKNOWLEDGMENTS

Thanks go to projects 41273101/D0309, 40773050/D0309 and 40573053 /D0309 supported by NSFC and the project Geological Information Public Service Product Development (Grant No. 1212011120434) supported by the Development and Research Centre, China Geological Survey. Thanks also go to the Radiation Monitoring Technology Centre of the Ministry of Environmental Protection for their field measured gamma data and to the Guangxi radiation environmental monitoring station for their field measured gamma data of anomaly points.

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NATURAL RADIOACTIVITY SOURCE TERM BASED ON REMOTE SENSING DATA

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Abstract

The paper describes the basic principles for applying satellite remote sensing technology to the investigation of natural radioactivity. The relationship between areas of natural background anomalies and geological characteristics is analysed systematically. The supervised classification method and spectral angle mapping are used for the extraction of remote sensing information. Geological features with elevated levels of gamma radiation can be identified on small scale maps. On-site inspections have been launched. The relationship between natural radiation level and radiation source term is becoming clearer. The study provides exact locations and targets for protection and control in areas with elevated levels of gamma radiation. The project has the potential for expanding the range of services in environmental geochemistry and remote sensing geology. It opens up a new approach for conducting research on natural radioactivity.

1. INTRODUCTION

Human exposure to radiation comes mainly from natural sources which contribute 92.8% of the total dose. If the impact of human activities involving

minerals with elevated radioactivity levels is taken into account, the contribution of natural sources is 95.62% [1]. However, raising awareness of the need to control radiation hazards may give rise to unwarranted concern among members of the public. For example, the Fukushima nuclear accident in 2011 provoked fear and an improper response from society. So the choice of method for studying natural sources is becoming increasingly important.

In the field of remote sensing, different types of ground covering have different emission characteristics. Investigations of the natural radioactivity source term, using remote sensing data applied to complex geological features, and remote sensing images of various targets [2, 3] show a series of geological regularities and geological phenomena. This allows in-depth systematic mapping of geological information and remote sensing images using digital image processing techniques, mathematical statistical analysis and other techniques. This form of investigation allows dynamic monitoring, forecasting, simulation and examination of complicated geological phenomena. Current geochemical data, based on one sample per 4 km², do not allow the exact locations of areas of elevated activity concentration to be easily identified. Using the spectral characteristics of remote sensing images, such locations can be determined. Under the present quality assurance system, geochemistry data for uranium, thorium and potassium are used for the extraction and identification of remote sensing information. Spectral angle mapping is used as a study technique and the geological information extracted is relevant to gamma radiation. Using satellite remote sensing technology, geological features with elevated levels of gamma radiation can be identified on small scale maps. On-site inspections have been launched. The relationship between natural radiation levels and radiation source item is becoming clearer. The study provides exact locations and targets for protection and control in areas with high levels of gamma radiation.

This paper describes the basic principles for applying satellite remote sensing technology to the investigation of natural radioactivity and describes some of the results of a study to evaluate environmental radioactivity with geochemical survey data (uranium, thorium and potassium) and remote sensing images.

2. RELATIONSHIP BETWEEN AREAS OF ABNORMAL RADIOACTIVITY AND ROCK FORMATIONS IN GUANGXI

Throughout Guangxi, rock formations exhibiting relatively high elevations of radioactivity levels are mainly composed of large batholiths, as shown in Fig. 1. On the other hand, large batholiths may not always exhibit such highly elevated elevations in radioactivity levels. It is difficult to determine the causes of elevated radioactivity only from a geological map. Through the analysis of

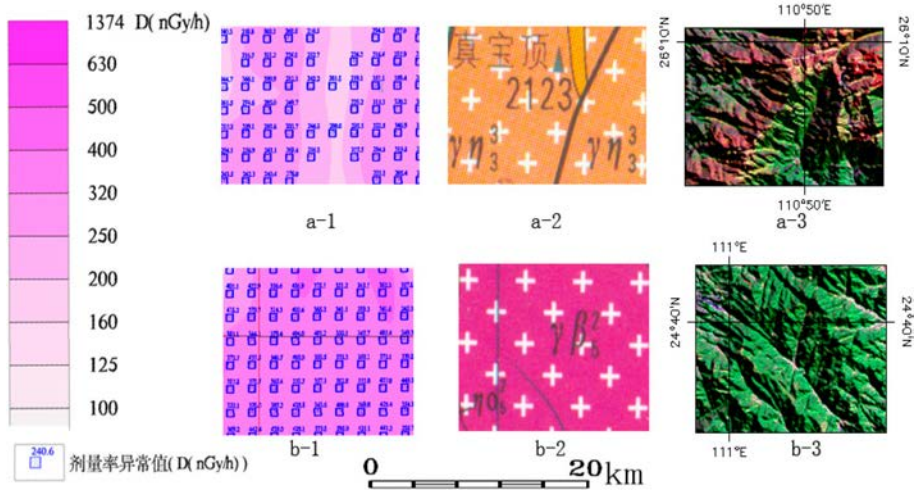


FIG. 1. Magmatic formations in Guangxi exhibiting relatively high elevations of radioactivity levels.

remote sensing information, rock formations with relatively highly elevated radioactivity levels have been shown to have the following features:

- They are located in high mountain regions.
- They are generally associated with strong tectonic movements.

In both situations, terrestrial gamma dose rates can exceed 300 nGy/h.

Rock formations with relatively moderately elevated levels of radioactivity contain, for example, variscan biotite adamellite in Pubei and Yulin ($\gamma\eta_4$) and Yanshanian biotite adamellite in the north-east of Nanning ($\gamma\eta_5^3$) (see Fig. 2). Although the rock formation is biotite adamellite ($\gamma\eta$) and batholith scale is the largest rock formation in Guangxi, the terrestrial gamma dose rates are moderate (100–200 nGy/h). Through the analysis of remote sensing information, the following features of the rock formations were identified:

- (a) Although the neotectonics of granitic batholiths is intense, the terrain elevation difference and absolute height above sea level of the batholiths are smaller, this being a low mountain landscape area.
- (b) The rock formation has the characteristics of weathering erosion and nesting of landscapes in the remote sensing image. The terrestrial gamma dose rate is about 150 nGy/h.

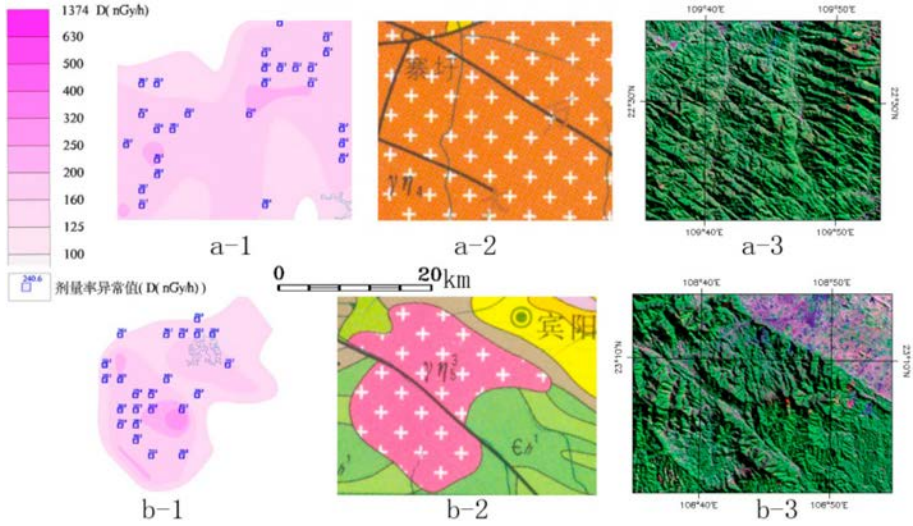


FIG. 2. Magmatic formations in Guangxi exhibiting relatively moderate elevations of radioactivity levels.

The type and distribution of rock formations exhibiting relatively low elevations of radioactivity levels have a certain regularity. Proterozoic granite ($\gamma_2^{2(2)}$) and early Yanshanian granite porphyry ($\gamma\pi_5^1$) both have low elevations of radioactivity levels, with a terrestrial gamma dose rate of about 150 nGy/h. The old granite body ($\gamma_2^{2(2)}$) is in a region characterized by high altitudes and large differences in altitude, topographic and geomorphic (see Fig. 3).

The most important factors affecting the level of radioactivity in magmatic formations in Guangxi are, firstly, the rock type and, secondly, the exposed location. The regions consisting of biotite adamellite or with intense neotectonics exhibit relatively high elevations of radioactivity levels. The radioactivity levels of other granites are not so high. The foregoing discussion relates to the general characteristics of intermediate acidity magmatic rocks in Guangxi.

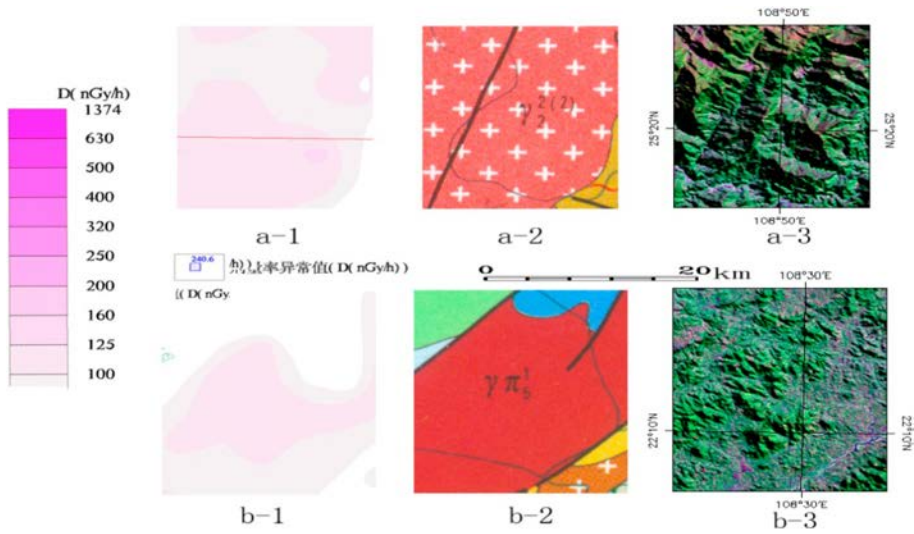


FIG. 3. Magmatic formations in Guangxi exhibiting relatively low elevations of radioactivity levels.

3. RESULTS OF ON-SITE INSPECTIONS

From the on-site inspections, dose rates were determined at locations of elevated radioactivity levels and are shown in Table 1. A comparison of the remote sensing data with the on-site inspection information verified the remote sensing anomaly characteristics at all sampling sites. It also verified that the relative degrees of elevation of radioactivity levels in the rock were as follows:

- Gxfy001 and Gxfy002: high;
- Gxfy007: moderate;
- Gxfy008, Gxfy009 and Gxfy010: low to medium;
- Gxfy004, Gxfy005 and Gxfy006: low.

The results also revealed that alteration phenomena were caused by hidden igneous rock.

TABLE 1. TERRESTRIAL GAMMA DOSE RATES AT LOCATIONS WITH ELEVATED RADIOACTIVITY LEVELS

	Longitude	Latitude	Location	Dose rate (nSv/h)	
				Mean	Standard deviation
Gxfy001	110°53'02.00"	26°03'32.50"	30 km NNW of Quanzhou	231.25	14.34
Gxfy002	110°57'30.40"	24°35'25.90"	4 km E of Tong An Zhen	202.20	9.42
Gxfy004	108°07'6.00"	24°34'40.74"	1.5 km W of Bai Tu Xiang	40.09	4.24
Gxfy005	108°08'8.70"	24°35'26.46"	1 km W of Bai Tu Xiang	42.70	2.33
Gxfy006	108°09'45.30"	24°27'6.12"	14.2 km SSE of Bai Tu Xiang	57.20	2.68
Gxfy007	106°36'2.80"	23°5'37.60"	3.6 km NE of Tong De Xiang	183.00	6.68
Gxfy008	106°26'52.20"	23°04'32.70"	Five Ridges	107.00	3.83
Gxfy009	109°34'48.50"	22°30'14.20"	25 km N of Pu Bei County	132.50	4.27
Gxfy010	110°58'54.70"	22°56'42.00"	2.6 km NW of Cen Xi	152.70	7.94

4. CONCLUSION

The results verify the rationality and reliability of the investigation of the natural radioactivity source item based on remote sensing data. Through this technique, areas with elevated radioactivity levels can be rapidly identified and accurate targets can be provided for the effective implementation of any protection and control measures needed. The project has the potential for expanding the range of services in environmental geochemistry and remote sensing geology. It opens up a new approach for conducting research on natural radioactivity.

ACKNOWLEDGMENTS

Thanks go to the projects 41273101/D0309, 40773050/D0309 and 40573053/D0309 supported by the NSFC and the project Geological Information Public Service Product Development (Grant No. 1212011120434) supported by the Development and Research Centre, China Geological Survey.

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NORM RESIDUES

(Topical Session 6)

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Invited Paper

TOWARDS A SUSTAINABLE SOLUTION TO NORM RESIDUE MANAGEMENT SAFETY

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Abstract

NORM residues arise inevitably from a broad range of mining and mineral processing activities. The need for radiological safety of NORM residues has long been recognized; however, no harmonious solution has been found internationally. Recent work on NORM residues has improved the understanding of NORM residues considerably, but much remains to be done. A systematic investigation of NORM residues is still needed in order to acquire a comprehensive understanding of NORM residues. Stakeholder awareness of the potential radiological hazards to health and the environment needs to be raised. A graded approach needs to be explored in order to achieve a practical and reasonable level of control over NORM residues. Management options for NORM residues need to be further explored and evaluated, including the options for recycling, use as by-products and disposal as waste. For those residues for which use or recycling is not feasible, careful consideration needs to be given to possible exemption and clearance in order to reduce the amount requiring disposal as waste.

1. INTRODUCTION

Owing to the ubiquity of radionuclides of natural origin in the environment, it is natural and inevitable that human activities such as mining and mineral processing can change the concentration of these radionuclides in products, by-products, residues and wastes. Such changes in concentration can result in enhanced exposure of workers, the public and the environment. NORM residues were addressed in 1997 in the first of this series of international symposia on NORM [1]. The IAEA International Conference on Topical Issues in Nuclear, Radiation and Radioactive Waste Safety, held in 1998, concluded that further guidance was needed on the issues of control of exposure to natural radiation [2]. In 2002, the IAEA published IAEA Safety Standards Series No. WS-G-1.2, Management of Radioactive Waste from the Mining and Milling of Ores [3]. An IAEA publication in 2006, Safety Reports Series No. 49, Assessing the

Need for Radiation Protection Measures in Work Involving Minerals and Raw Materials [4], identified 12 industries which may require consideration in terms of radiation protection and NORM residue management. In addition to the mining and processing of uranium ore, the other 11 industry sectors are as follows:

- (1) Extraction of rare earth elements;
- (2) Production and use of thorium and its compounds;
- (3) Production of niobium and ferroniobium;
- (4) Mining of ores other than uranium ore;
- (5) Production of oil and gas;
- (6) Manufacture of titanium dioxide pigments;
- (7) The phosphate industry;
- (8) The zircon and zirconia industries;
- (9) Production of tin, copper, aluminium, zinc, lead, iron and steel;
- (10) Combustion of coal;
- (11) Water treatment.

This list brings challenges to the management of NORM residues, not only in technical aspects but also in social aspects. At the NORM V symposium in 2007, ten years after NORM I, the question was raised as to whether a consensus on the management of exposure to NORM was being reached, or whether there was still a state of chaos [5]. It was concluded that some consensus still needed to be reached, particularly on the scope of regulatory control of NORM. In the meantime, the IAEA was continuing to develop safety reports for specific industry sectors such as rare earths extraction [6], the oil and gas industry [7], the zircon and zirconia industry [8], titanium dioxide production [9] and the phosphate industry [10]. This paper reviews the work of the IAEA in developing a safety guide for NORM residue management. Current challenges in the management of NORM residues are summarized and potential solutions are proposed.

2. DEVELOPMENT OF IAEA SAFETY STANDARDS FOR NORM RESIDUE MANAGEMENT

The IAEA safety standards framework comprises safety fundamentals, safety requirements and safety guides. These are supported by other publications including safety reports. Shortly after publishing IAEA Safety Standards Series No. WS-G-1.2 [3], the IAEA started to prepare a more general safety guide dealing with the management of NORM waste that did not focus specifically on uranium mining and processing waste. Concurrently, the IAEA was developing a safety guide on the protection of the public against exposure to natural sources.

After some time, efforts were made to merge these two documents into a single safety guide, but it was eventually decided to keep them as separate documents.

The term ‘NORM residue’ is defined in the IAEA Safety Glossary [11] as:

“Material that remains from a *process* and comprises or is contaminated by *naturally occurring radioactive material (NORM)*” (original emphasis).

The related term ‘NORM waste’ is defined as:

“*Naturally occurring radioactive material* for which no further use is foreseen” [11] (original emphasis).

In terms of these definitions, therefore, a NORM residue may or may not be waste. There has been relatively little specific guidance on the management of NORM residues (including NORM residues designated for disposal as waste) arising from mining and raw material processing operations other than those associated with the exploitation of uranium and thorium. Such industries include titanium dioxide pigment industries, phosphate fertilizer production, mineral sands exploitation and water treatment. To fill this gap and also to meet the increasing demands from IAEA Member States in dealing with issues associated with NORM residues, the IAEA started to revise IAEA Safety Standards Series No. WS-G-1.2 [3] to cover residues from NORM activities other than the mining and mineral processing of uranium ore. This safety guide would focus on the identification and implementation of appropriate measures for protection of members of the public and the environment against radiological hazards associated with the management of all types of NORM residue encountered in industrial operations. The developing guidance will serve as life cycle guidance on the site selection and evaluation and design of management facilities, and on their construction, operation and closure, decommissioning and termination, including organizational and regulatory requirements. The guidance under development is expected to provide regulatory bodies with the necessary knowledge for authorizing industries to continue to operate safely and effectively while ensuring that the desired goals of long term safety and protection for the workforce, the public and the environment are achieved.

3. CHALLENGES IN NORM RESIDUE MANAGEMENT

The management of NORM residues is facing quite significant challenges, in particular the challenges of awareness and risk communication with concerned parties, the identification and characterization of NORM residues and the

management strategy for the use, recycling and disposal of NORM residues. The risks associated with NORM residues have not been well recognized in the past. Some of the industries on the list in Section 1 are not usually associated with the additional need for radioactive waste management. However, some protection measures are still needed to protect the worker. The level of awareness of the issues of radiation exposure in various working activities is quite low. Large volumes of NORM residues have been produced by industries around the world. The potential risk to human health and the environment have not received a commensurate level of attention. Proper actions need to be taken to improve knowledge and understanding. People may be concerned that many of the industries and processes that generate NORM residues have not traditionally been associated with radioactivity. Thus, the introduction of radiation protection requirements, in compliance with international standards, can potentially cause considerable public concern, especially where industries have been located in, or near, areas of high population. Equally, the introduction of such requirements can have a significant impact on the industries themselves. This undoubtedly presents a communication challenge.

Increasingly, NORM residues have been recognized as a resource rather than a waste. Recycling of NORM residues or their use as by-products is increasingly being catered for (and encouraged) in national legislation. However, there is a lack of uniformity in the regulatory approach to NORM in building materials, which is more conservative in highly developed countries and more pragmatic in countries with developing economies, while still respecting the need for safety. The IAEA is developing a safety guide to provide more relevant information on the recycling and use of NORM as building materials. In practice, such recycling and use need a prior comprehensive evaluation with consideration of various factors, including non-radiological contaminants contained in the NORM residues concerned.

Disposal options seem to be clear. However, the disposal of NORM residues in practice is challenged by the need to decide between the basic approaches of dilution/dispersion and isolation/containment. It is also challenged by issues such as long term management and access to landfill sites. When disposal is the only option for NORM residue management, the strategy of disposal is to be carefully evaluated. Exemption of NORM residues can be a pragmatic solution when the potential risk to the public and environment is sufficiently low. Ideally, a safety assessment should be conducted, based on reasonable assumptions rather than on very conservative scenarios. An overconservative assessment may result in the wrong message to the stakeholder, who may then challenge the appropriateness of the disposal actions for such residues. Eventually, this could result in a greater risk rather than greater safety.

4. VISION FOR A SUSTAINABLE SOLUTION

A sustainable solution to the safety of NORM residue management needs to be established to protect people and the environment from the harmful effects of radiation without unduly limiting NORM activities. Such a solution should ideally be holistic and pragmatic, comprehensive, consistent with the graded approach to regulation and founded on a solid scientific and technological basis. The solution should ideally be developed with a broad awareness among, and acceptance of, the various concerned parties. It should ideally be separated rather than integrated with non-radiological control.

To develop such solution, a coordinated and well prepared awareness programme needs to be established and implemented by seeking synergies between current international, regional and national activities and resources, and by attracting attention at the international and regional level such as through the NORM Symposia and similar events. Correct and reasonable information and knowledge need to be explored using recognized technical approaches, methodologies and procedures. The IAEA is seeking to develop thematic documents supporting the identification and characterization of NORM residues and safety assessment approaches for screening management options. The IAEA is intending to develop a safety report to review existing good practices in the management of NORM residues among its Member States and to promote the sharing of knowledge and information.

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Invited Paper

EXPERIENCE FROM REMEDIATION OF THORIUM CONTAMINATED SITES: RADIATION PROTECTION AND WASTE MANAGEMENT ISSUES

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Abstract

The paper describes the practical radiation protection and waste management issues arising from the decommissioning and demolition of sites contaminated with thorium from past industrial practices. This includes a description of the radiological objectives established at the start of decommissioning and the practical radiation protection procedures used to deliver these objectives, as well as the strategies and methods used for the management of radioactive waste.

1. INTRODUCTION

In addition to its national radiation protection functions, the Centre for Radiation, Chemical and Environmental Hazards of Public Health England (PHE)¹ provides advice and technical services to organizations using radiation sources, including NORM industries, and has accumulated a significant amount of practical experience in establishing and operating radiation protection and waste management programmes. This paper summarizes the experience gained from the decommissioning and remediation of several (former) industrial sites contaminated with ²³²Th and its progeny. The industrial activities involving NORM that were conducted previously at these sites include the following:

- (a) Processing of mineral sands containing ²³²Th series radionuclides in equilibrium;

¹ Formerly, the Health Protection Agency.

- (b) Manufacture of thorium gas mantles;
- (c) Production and recycling of magnesium–thorium alloy.

This paper describes the methods used to establish the on-site radiological conditions prior to decommissioning and remediation and the setting of post-remediation radiological end points. It then summarizes the main operational features of the radiation protection and waste management programmes implemented to achieve these end points.

2. ESTABLISHING THE PRIOR RADIOLOGICAL CONDITIONS

For all the sites considered in this paper, it was necessary to undertake a comprehensive site survey (or surveys) to gain an understanding of the location, quantity and distribution of the radioactive contamination, and thus facilitate the planning of the decommissioning and remediation process. However, before this survey was undertaken, it was important to review any available historical information about the site relating to the materials used and how they were processed. For example:

- (a) Feed materials: It is useful to know the quantities that were previously used, the thorium activity concentration and current decay chain equilibrium (because this may have changed since the operations ceased) and the storage arrangements on site. The physical and chemical properties are also important, not just for internal dose calculations (e.g. to assign an appropriate lung absorption type), but also for considering how contamination might have been spread at the site during operations and how it might have subsequently migrated through the ground. The visual properties of materials may also be useful, especially when remediating mineral sand sites where layers of buried materials may be easier to see than to identify by radiological monitoring.
- (b) Processes: It is important to understand the effect of any industrial processes on the radionuclides in the feed materials and to consider where these radionuclides might be deposited in the plant and where they might appear in either the products or residues. In older processes, this may not have ever been considered, but it is still very useful to consider this retrospectively prior to remediation.
- (c) Original waste management practices: It is always important to know how residues and waste materials (such as those from plant cleaning and refurbishment) were dealt with and in particular whether there was any on-site disposal (usually by shallow burial).

This historical information will be useful for the planning of pre-remediation activities, for example in terms of the areas to be surveyed, possible depth of ground contamination and selection and calibration of survey equipment. However, it is often difficult to obtain reliable historical information on waste management practices, especially for processes that operated prior to the imposition of regulatory controls on such NORM industries. In almost all cases, the actual extent of contamination has been found to be larger than originally expected and the initial surveys are always to be designed with this in mind. For thorium contaminated sites, the initial survey has typically consisted of:

- (a) A 'walkover' survey with portable NaI probes to detect the gamma emissions from ^{232}Th series radionuclides. This has generally proved to have sufficient sensitivity for locating contamination, even when concealed under surfaces or buried below ground level. For large outdoor areas, GPS logging is used to facilitate the mapping the results. Where significant readings are obtained, gamma dose rate measurements are also made for risk assessment purposes.
- (b) Surface contamination measurements (dual detection of alpha and beta radiation) may also be undertaken, where appropriate. Normally, this is just in, or on, buildings in locations where the contamination is likely to be confined to the immediate surface. Care is needed in interpreting the results, especially for alpha contamination, which is often completely absorbed by the surface), but in the right circumstances it can accurately delineate contaminated material. This has proved especially useful where uncontaminated building materials are to be reused or recycled.
- (c) Where practicable, trial excavations in the form of boreholes or pits are undertaken. In practice, buried contamination has been found on every site, even where the historical records would suggest otherwise. Surface gamma surveys can detect buried contamination, in some cases up to 2 m deep, but at depths greater than about 30 cm, they are unlikely to be sensitive enough for clearance surveys.
- (d) The analysis of samples by gamma spectrometry confirms the degree of decay chain equilibrium and also provides an empirical calibration for the on-site gamma measurements. Final clearance surveys always include sample analysis by gamma spectrometry to support the walkover survey.

In the contaminated sites considered in this paper, contamination levels have typically been found to be in the range of 0.1–100 Bq/g for ^{232}Th series radionuclides, with a maximum value of 500 Bq/g (12% thorium by weight). Dose rates have typically been found to be up to 20 $\mu\text{Sv/h}$ at surfaces and up to 5 $\mu\text{Sv/h}$ at positions representative of whole body exposure.

3. ESTABLISHING THE END POINT OF REMEDIATION

In the United Kingdom, the basic reference level applied to the release of contaminated land for public use is an incremental dose of 300 $\mu\text{Sv/a}$ to a member of the public. Below this reference level, the remediation should be optimized so that predicted future exposures will be as low as reasonably achievable. For contamination by radionuclides of artificial origin, the ambition is to reduce doses to below 20 $\mu\text{Sv/a}$ where practicable. This is not usually practicable for contamination by radionuclides of natural origin, normally because of difficulties in distinguishing the contamination from the normal background level, and 300 $\mu\text{Sv/a}$ is often adopted as the target value.

For practical purposes, the remediation end point needs to be defined in terms of a residual thorium activity concentration, and guidance has been published in the United Kingdom on the methodology for determining this [1]. In addition, in UK legislation there are generic 'out of scope' activity concentration levels for radionuclides of natural origin, which are based on the concept of exclusion from the scope of legislation. As a result, different end points (in terms of the residual activity concentration for ^{232}Th series radionuclides) have been agreed in recent years. Examples include the following:

- (a) Redevelopment, for residential use, of sites contaminated by past practices: <0.1 Bq/g;
- (b) Transfer of a contaminated site to a different industrial use: <0.5 Bq/g;
- (c) Partial remediation of a site at which industrial operations are to continue: <0.5 Bq/g.

In practice, it was found that an end point of <0.1 Bq/g (above a natural background level of 0.03 Bq/g) was achievable but only just, and then only if a reasonable degree of averaging over several square metres of ground could be applied. Indeed, the degree of averaging is always a factor (although often not explicitly stated) in all site remediation activities.

4. RADIATION PROTECTION PROGRAMME

The specific details of the radiation protection programme vary from site to site. However, they are all underpinned by a prior radiation risk assessment, which is required by UK regulations [2]. In such an assessment, the radiation risks to employees and other persons from normal operations and from reasonably foreseeable accidents have to be considered. This assessment is then used to determine the need for specific radiation protection measures, such as

designated areas and individual monitoring. Typical conclusions for the thorium contaminated sites considered in this paper are as follows:

- (a) Predicted doses (remediation workers):
 - (i) External radiation 0.1–1 mSv/a;
 - (ii) Internal radiation from inhalation, assuming no respiratory protective equipment 0.2–2 mSv/a.
- (b) Dose restriction measures:
 - (i) Written procedures for all operations;
 - (ii) Simple dust suppression measures such as extraction and damping down;
 - (iii) Temporary containment or ventilation where required;
 - (iv) Entry and exit facilities as appropriate (e.g. barrier, change, wash and monitor);
 - (v) Containment of accumulated NORM waste;
 - (vi) Training of workers;
 - (vii) Personal protective equipment (PPE), including respiratory protective equipment (RPE) for defined dusty tasks;
 - (viii) Regular ‘in-progress’ surveys;
 - (ix) Contingency plans for cuts and wounds.
- (c) An investigation level of 1 mSv (annual effective dose).
- (d) Designation of controlled areas for remediation of thorium processing areas and supervised areas for remediation of areas such as those for mineral sand processing to prevent the spread of contamination.
- (e) Designation, as radiation workers, of remediation workers (‘Category B’ workers).
- (f) Individual assessments of external radiation doses using passive dosimeters of the thermoluminescent type, mostly for reassurance purposes.
- (g) Periodic air sampling to provide an estimate of the level of internal exposure, although internal doses are not routinely assessed.

On-site radiation protection supervision has been provided by PHE or, where appropriate, delegated to trained site personnel. On-site precautions such as entry and exit procedures were designed to fit in with familiar site procedures, and were generally successful. For example, existing containment tents, procedures and PPE used for asbestos removal were readily adapted for work with radioactive contamination.

A review of the radiological information obtained from the radiation protection programme for the largest site remediation project undertaken revealed the following:

- (a) External doses were less than 0.1 mSv compared with predictions of up to 1 mSv.
- (b) Internal doses were less than 0.1 mSv compared with predictions of up to 2 mSv with no RPE.
- (c) Exit monitoring — from a total of 1300 person exits:
 - (i) There were 66 instances of contaminated gloves (5%);
 - (ii) There were 2 instances of contaminated skin (0.2%);
 - (iii) There was 1 instance of contaminated shoes (<0.1%);
 - (iv) All of the above were successfully decontaminated at the exit.
- (d) Concentrations of airborne thorium at the site were at background levels and no surface contamination was detected outside the designated areas.
- (e) There were four minor wounds, but no contamination was detected.

5. WASTE MANAGEMENT PROGRAMME

All the sites referred to in this paper were authorized for the accumulation and disposal of radioactive waste in accordance with an authorization granted by the regulatory body [3]. This authorization was either already in existence (i.e. to cover the site operation) or granted specifically for the site decommissioning and remediation project. A condition of such an authorization is that the amount of radioactive waste (in terms of activity and volume) is minimized via a process referred to as ‘best available techniques’ (BAT). Thus, there is a requirement to efficiently segregate radioactive and non-radioactive waste material. Consequently, all waste arising from the site remediation needed to be subject to radiological screening measurements and then categorized according to regulatory requirements. In the United Kingdom, the current approach for waste containing ^{232}Th series radionuclides is typically as follows:

- (a) If the radionuclide activity concentration exceeds 5 Bq/g, the waste is disposed of at an authorized disposal site as radioactive waste.
- (b) If the radionuclide concentration is 0.5–5 Bq/g, the waste is designated as exempt NORM waste [4] and disposed of at a conventional landfill facility.
- (c) If the radionuclide concentration is below 0.5 Bq/g, the waste is ‘out-of-scope’ and can be left on site and recycled, among other things.

The volume of waste over which the activity concentration can be averaged needs to be considered when designing the waste management programme. In practice, the averaging volume has ranged from individual bags containing a few kilograms of waste stripped from buildings, through excavator bucket volumes, up to 10 t skip volumes. Smaller waste containers enable better BAT

waste segregation, but are not always an optimum option, especially for ground excavations where a significant degree of mixing is unavoidable.

Waste screening is normally done by in situ external gamma radiation measurement (rather than sampling and gamma spectrometry), supported by a geometry specific, computer calculated calibration to estimate the activity concentration. In some cases (normally as a result of waste acceptance criteria), fixed geometry rotating drum gamma spectrometers have been used. In practice, handheld monitors have proved equally accurate (and often more reliable). In either case, it is important to cross-check the accuracy of the measurement using standards containing unevenly distributed thorium with a known activity concentration.

The waste management programme needs to include rigorous record keeping arrangements for authorized and exempt waste. On the biggest remediation project, this involved keeping records of over 6000 individual waste bags, 40 low level waste drums and 150 lorries and skips of exempt waste. Spreadsheets were developed to facilitate the record keeping and to be able to readily demonstrate compliance with regulatory limits.

6. CONCLUSIONS

From practical experience in planning, implementing and completing the remediation of thorium contaminated sites, key lessons have been learned and the following conclusions can be drawn:

- (a) The end point of remediation needs to be agreed in advance with the relevant authorities, including the arrangements for averaging activity concentrations over defined waste volumes.
- (b) A thorough site survey is needed to establish the radiological conditions on site prior remediation. Equally important, however, is the gathering and review of historical data about the site, the feed materials and how they were processed, and any previous waste management arrangements.
- (c) The initial survey should ideally be guided by the historical information, but be based on the expectation that the extent of any residual contamination will be greater than initially indicated, with more contamination being discovered as the remediation progresses.
- (d) Gamma monitoring is the best all-round tool for the initial survey, especially where contamination is not directly on the surface. However, it is to be supported by other monitoring techniques or sampling to give a full picture.

- (e) Buried contamination is common, even when it is not indicated by historical records. Trial excavations or pits are always to be considered as part of the initial survey.
- (f) Radiation protection programmes are to be based on a realistic prior risk assessment. In practice, precautions based on normal good industrial hygiene are effective. A dose constraint of 1 mSv/a has been appropriate for most sites.
- (g) A waste management strategy needs to be developed from the start, based on national criteria for different categories of waste. A waste screening programme will be required, for which a variety of gamma measurement methods can be utilized, provided that they are supported by geometry specific calibration factors and tested with samples of known activity.

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DISPOSAL OF RARE EARTH NORM RESIDUES IN JIANGSU PROVINCE, CHINA

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Abstract

The present situation of NORM residues arising from rare earths extraction and separation in Jiangsu Province has been investigated comprehensively. The management of these residues is discussed and analysed with respect to the separation technology, the activity concentrations, the amounts of residue generated and the regional eco-environmental features. Some conclusions and suggestions on the disposal of these NORM residues are given, with reference to the relevant national laws, regulations and standards.

1. INTRODUCTION

Rare earth elements are a group of 17 elements, 15 within the chemical group called lanthanides, plus yttrium and scandium, which play an important role in civilian and military high technology applications owing to their magnetic and conductive properties. China, as the largest global rare earths producer, has to face the domestic problems of industry including, among other things, possible smuggling, environmental damage and technology-for-resources initiatives. The mining and processing of rare earths have produced millions of tonnes of wastewater, harmful chemical runoff and radioactive residues which, if not properly disposed or managed, will contaminate surrounding waters and farmlands and will also, to a varying degree, give rise to significant exposures of workers and members of the public. Mineral processing operations give rise to the concentration of uranium, thorium and their progeny radionuclides in sludge and tailings to levels significantly higher than natural background levels.

Activities involving rare earths in China can be divided into three categories: mining, processing and use of products. Jiangsu Province has about 11 rare earth smelting and separation plants and thus is an important national player. Most of these plants were built during the 1980s and 1990s and have produced tens or hundreds of tonnes of NORM waste per year. Currently these rare earth separation plants have accumulated up to 30 000 t of these NORM wastes which, in the absence of a clear disposal strategy, have to be kept in

storage at the plants concerned. A few of these storage facilities are almost full because they were not designed to meet present demands. Their ability to retain their integrity in the event of natural disasters is very limited, which could result in pollution of the surrounding environment. The possibility of illegal dumping of these wastes may also cause wide ranging pollution or environmental damage, including radioactive contamination of the environment. Monitoring data show that the soil and surface water have become polluted by radionuclides around some plants [1–5].

2. INVESTIGATION OF RARE EARTH PROCESS PLANTS IN JIANGSU

2.1. Process description

Rare earth materials produced in Jiangsu are primarily in two categories: concentrates and intermediate products. Rare earth concentrates are mainly in the form of weathered minerals containing heavy rare earths and are rich in yttrium and europium. Intermediate products include, among others, rare earth oxides, chlorides and samarium, europium and gadolinium. Mixed rare earth chlorides are produced by dissolving in acid and removing impurities, following which light and heavy rare earths are extracted as different components of an intermediate product solution. This is then processed by oxalic acid or soda precipitation, washing and dehydration, to achieve separation of the rare earths from the impurities as a highly pure chloride and eventually as oxides by high temperature processing. The process is shown in Fig. 1 [1, 2]. Wastewater treatment in the rare earth separation process is shown in Fig. 2 [3].

2.2. NORM residues

There are two main categories of moderately active NORM residue produced in the rare earth separation plants: acid dissolution residue and neutralization residue. Acid dissolution residue is generated by filtration of the residue produced by dissolving rare earth concentrate in hydrochloric acid (see Fig. 1) and contains mainly iron, aluminum, silicon, rare earth oxides, barium sulphate, radionuclides and insoluble complex rare earth salts. Rare earth separation mainly involves a single rare earth liquid produced by excessive extraction and separation of oxalic acid precipitate to achieve the separation of the rare earth element and non-rare-earth impurities in a high purity rare earth chloride. This leads to the generation of wastewater with a high percentage of hydrochloric acid and oxalic acid. Applying lime directly to the wastewater produces a large amount of neutralization residue (see Fig. 2), the main components of which are oxalic acid,

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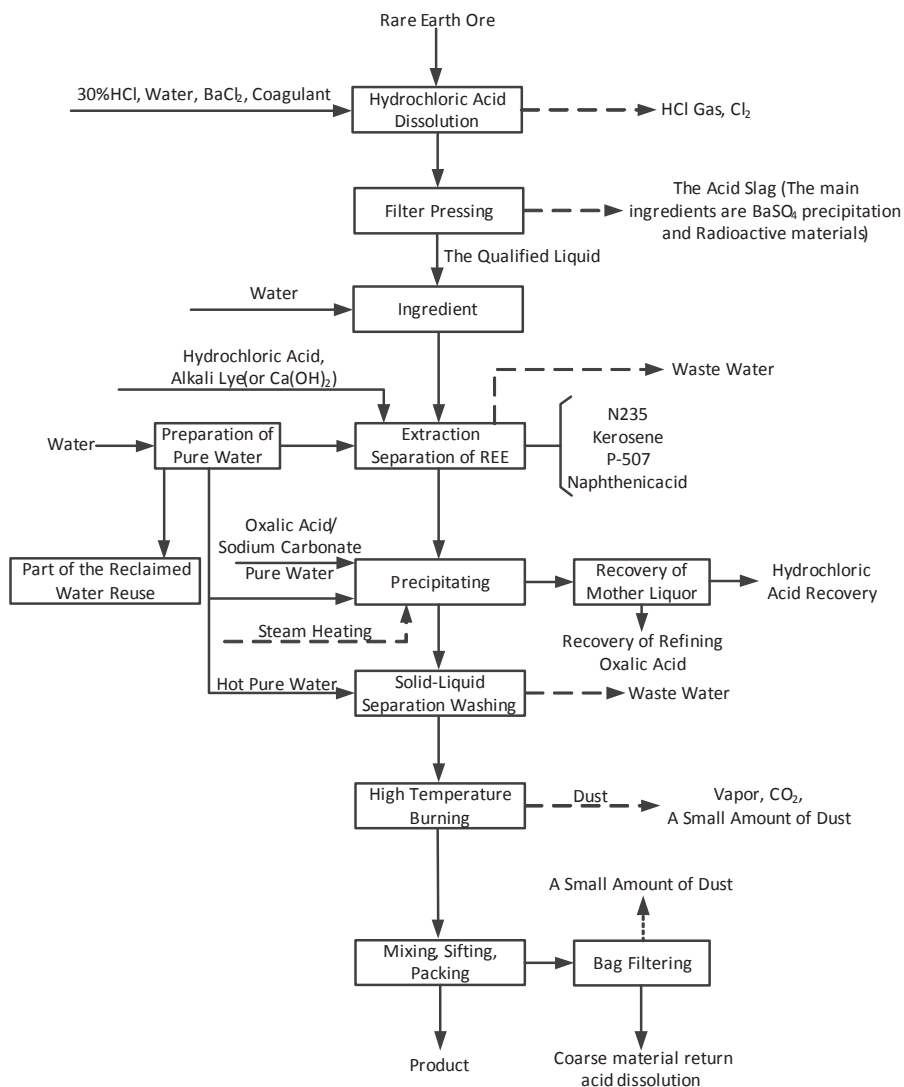


FIG. 1. Rare earth hydrometallurgical production process in Jiangsu.

calcium and water, with a small amount of oxalic acid, rare earths, radionuclides, heavy metal insoluble salts and calcium chloride. Of the 30 000 t of moderately active NORM waste produced by the plants, 5000 t are acid dissolution residue and up to 25 000 t are neutralization residue. The waste containing various amounts of uranium, thorium and their progeny will inevitably cause radioactive pollution of the environment. According to the latest (2011–2012) sample

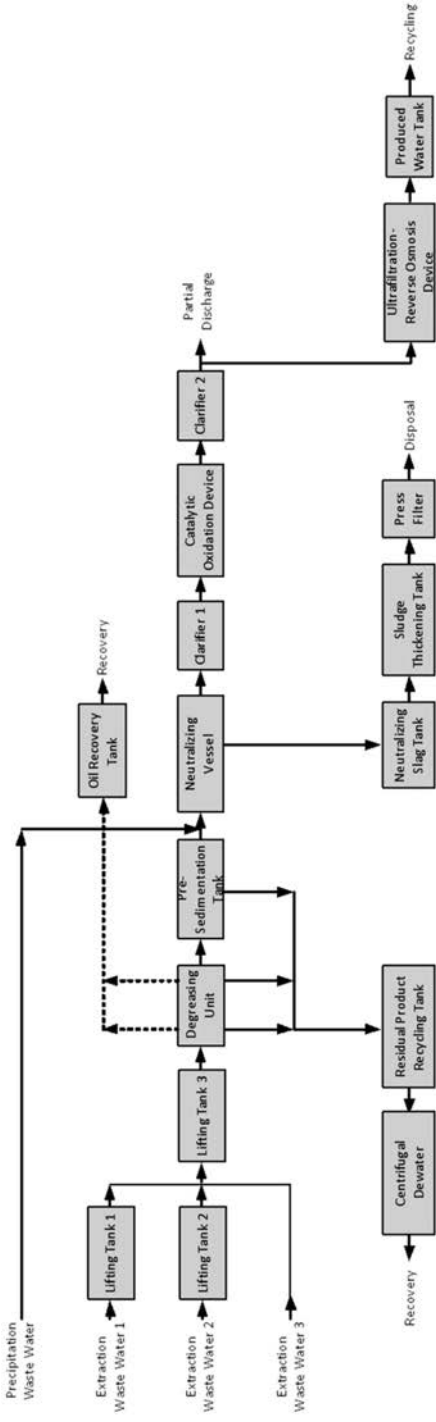


FIG. 2. Wastewater treatment in the rare earth separation process.

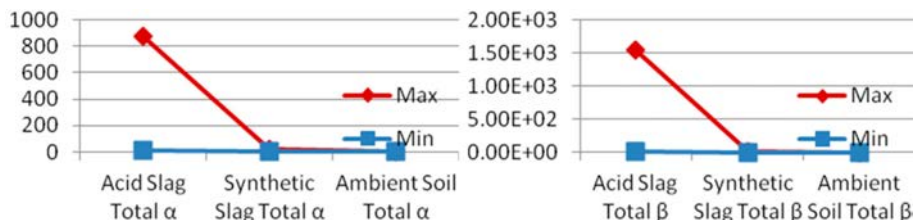


FIG. 3. Gross alpha and beta activity in acid dissolution residue, neutralization residue and environmental solids (in Bq/kg).

analysis results produced by the Jiangsu Radiation Environmental Management and Monitoring Centre (JSRE) [4, 5], acid dissolution residue has a high activity concentration, with a gross alpha activity concentration of 26–870 Bq/g and a gross beta activity concentration of 9.4–1600 Bq/g. The neutralization residue has a gross alpha activity concentration of 2.7–47 Bq/g and a gross beta activity concentration of 1.9–58 Bq/g. According to monitoring data for more than 60 environmental soil samples analysed by the JSRE, the environmental soil has a gross alpha activity concentration of 0.068–2.2 Bq/g and a gross beta activity concentration of 0.35–1.95 Bq/g. A summary of the activity concentration data is given in Fig. 3.

2.3. Management of NORM residues

Options for NORM residue management are mainly the following: temporary storage at the plant; returning to the producer of the raw material; reselling to other qualified companies. Most of the plants have built temporary storage facilities for the residues, but the overall situation is unsatisfactory owing to differences in the standard of construction and management approach, for instance inadequate storage capacity, a lack of effective surveillance and handling instructions, and a lack of control over disposal, leading to additional exposure of workers and members of the public.

3. PROBLEMS WITH NATIONAL REGULATIONS AND STANDARDS

Regulations and standards for the disposal of rare earth residues need to be improved in China. Although the foundation has been laid for radiation safety management in the development and use of mineral resources [6], there are also practical problems such as uncertain areas of responsibility, management programmes and requirements, which make it difficult for the implementation of

the regulations and standards. One problem is the lack of practical regulations and standards, although the mining of radioactive ores has already been brought under specific areas of responsibility [6, 7], with specific provision having been made for the associated radioactive waste. The other problem is the low operational level of hierarchical management. For low activity but massive amounts of mineral waste, the provisions for exclusion, exemption and clearance will be particularly important. Problems with the relevant standards specifications are as follows:

- (a) Exemption levels have been specified for specific radionuclides [8], but these levels can only be applied to small amounts of material. In addition, the exemption value for various types of radiation and radionuclide composition cannot be applied with the confidence that the balance of natural radioactivity will not be disturbed. It is difficult to use one or more weighted mean values of the exemption values for uranium or thorium series radionuclides to make a suitable judgement.
- (b) The categories of radioactive waste are clear in Ref. [9] — waste containing radionuclides of natural origin at activity concentrations greater than 74 Bq/g should be treated as radioactive waste. Residues with activity concentrations less than this value are nevertheless to be properly disposed of. Mines associated with uranium or thorium generate large amounts of NORM. It is not clear whether this activity criterion refers to the weighted mean of all individual radionuclide concentration measurements or to the total activity of any single uranium and thorium series radionuclide or to the total alpha–beta activity.
- (c) Although three categories of radioactive solid waste — low, medium and high level waste — are defined in Ref. [10], with specific provision for exemption of waste, the standard is very impractical for managing NORM waste associated with the mining of radioactive ores.

4. SUGGESTIONS AND CONCLUSIONS

4.1. Improving laws and regulations for NORM residues

Laws or regulations for the treatment, disposal and recycling of rare earth residues including relevant standards or rules for their implementation need to be

published instead of having to adopt the single approach of temporary storage. NORM waste is to be treated properly and strictly. It is not to be dumped in open places like normal industrial waste. Plants producing large amount of NORM waste are responsible for establishing temporary storage facilities. Plants need to put in place detailed management methods for temporary storage and introduce stricter supervision of rare earth residues.

4.2. Determining exemption and clearance levels

Classification of radioactive waste is an important principle in Ref. [8], which emphasizes the coherence between the management requirements implemented and the characteristics of the source, and also its size and hazard potential. It is critical to have defined criteria for exclusion, exemption and clearance for large amounts of residues with low activity concentrations. International organizations including the IAEA and the European Commission give standards and guidance on a classification system of protection and optimization for administrating NORM residues. There are two approaches in determining clearance levels: one is based on the ranges of natural levels of radioactivity in the environment, while the other is based on incremental dose. In Ref. [11], a clearance level of 1 Bq/g is given for individual radionuclides of natural origin. Clearance levels for individual radionuclides recommended by the European Commission are listed in Table 1. Canadian unconditional derived release limits are shown in Table 2 [11–14].

Safety based on the control of individual dose is a basic principle in radiation protection, but it is hard to obtain the data. On the other hand, the activity concentration of NORM residue is easy to measure, making this approach more feasible in practice. But the issue is that, if the activity concentration of a certain residue exceeds the exemption value, it may still not represent a risk to workers or members of the public. In such cases, it is generally to be exempted from regulatory supervision. Decision making based on dose may involve assumptions in the dose assessment which brings some uncertainty, but it is more scientific and acceptable.

TABLE 1. RECOMMENDED CLEARANCE CRITERIA:
EUROPEAN COMMISSION

	Activity concentration (Bq/g)
U-238 sec, U-235 sec	0.5
U nat	5
Th-230	10
Ra-226 ⁺	0.5
Pb-210 ⁺	5
Po-210	5
U-235 sec	1
U-235 ⁺	5
Pa-231	5
Ac-227 ⁺	1
Th-232 sec	0.5
Th-232	5
Ra-228 ⁺	1
Th-228 ⁺	0.5

TABLE 2. RECOMMENDED CLEARANCE CRITERIA: CANADA

	Activity concentration		
	Aqueous (Bq/L)	Solid (Bq/g)	Air (Bq/m ³)
U-238 (with progeny in equilibrium)	1	0.3	0.003
U-238 (U-238, Th-234, Pa-234m, U-234)	10	10	0.05
Th-230	5	10	0.01
Ra-226 (with progeny in equilibrium)	5	0.3	0.05
Pb-210 (with Bi-210, Po-210 in equilibrium)	1	0.3	0.05
Th-232 (with progeny in equilibrium)	1	0.3	0.002
Th-232	1	10	0.006
Ra-228 (with Ac-228 in equilibrium)	5	0.3	0.005
Th-228 (with progeny in equilibrium)	1	0.3	0.003
K-40	Not applicable	17	Not applicable

So, combined with national criteria for the use of minerals and referring to experience from Canada, the European Commission and the IAEA, the activity concentration seems to be the best way to classify the material as being exempt or subject to regulatory control, whether or not the dose from associated products, raw materials or residual material exceeds the exemption level. For example, a residue which satisfies national requirements [15] can be used as construction material for a roadbed, otherwise it is to be treated as normal solid waste. Specifications of sampling and monitoring which will instruct the practical operations for the enterprises or the management agencies are to be formulated and published by the relevant departments as soon as possible. If the radioactive waste index is greater than the clearance level, it should ideally be supervised by the environmental protection agency.

4.3. NORM waste repository

A repository for low activity NORM waste needs to be built according to the national and regional laws or regulations whose sites are chosen provincially or regionally considering geological safety and natural conditions. Reference [16] also requires that the provincial environment protection agency would be responsible for establishing such a repository.

4.4. Waste reduction and recycling in the rare earths separation process

According to the investigation and monitoring data of rare earth plants in Jiangsu, the activity concentrations of acid dissolution residue vary by three orders of magnitude, while those of the neutralization residue vary by two orders of magnitude. These variations are due to differences in raw materials, manufacturing technology and waste management approaches. Different impurity removal processes, even for the same rare earth ore, will lead to significant differences in residue activity concentrations. Most of the plants have not classified the radioactive and non-radioactive residues in terms of how the different raw material and manufacturing technologies will result in higher activities in acid dissolution waste. If the wastewater is not classified into radioactive and non-radioactive streams, or if it is not treated to remove radioactivity, the amount of radioactive neutralization residue production will increase by many times. It is worth mentioning that the annual production and storage amount of neutralization residue takes up more than 85% of all NORM waste. Applying the process of radioactivity removal (i.e. through adding aluminum sulphate and barium chloride to precipitate radionuclides in the extraction and precipitation processes) could reduce radioactivity levels by 80%. In addition, an investigation into the recycling of thorium, which is present at about 1–2% in the acid dissolution residue, and its industrial application, could reduce the amounts NORM residues generated.

ACKNOWLEDGEMENTS

This work was supported as a sub-project of a national nuclear and radiation safety surveillance project by the Ministry of Environmental Protection and as a sub-project from the Province of Jiangsu Environmental Protection (No. 201150).

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DISPOSAL OF SOME TYPES OF NORM THAT EXCEED 10 Bq/g IN SWEDEN

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Abstract

The Swedish Radiation Safety Authority (SSM, Strålsäkerhetsmyndigheten) has issued regulation SSMFS 2011:4, which specifies exemption levels for NORM and regulates how NORM are to be handled. However, the disposal of NORM with activity concentrations higher than 10 Bq/g for ^{238}U or ^{232}Th decay series radionuclides is not included in this regulation. Examples of such material are scrap metal with radioactive scale contamination found in metal recycling facilities and uranium contaminated filters derived from water treatment. This study was performed with a view to providing a basis for SSM's judgment on how the final disposal of this NORM contaminated material is to be handled in a radiologically safe manner in Sweden.

1. INTRODUCTION

Large quantities of NORM from past activities in Sweden include residues coming from industries such as the phosphate industry, alum shale exploitation for energy production and the production and use of shale based lightweight concrete for building construction. Besides these large amounts of existing waste there are other, smaller amounts of NORM waste. Presently, these are collected and stored at several places in Sweden. This accumulated waste is both metallic and non-metallic, with the metallic fraction being the most prevalent. One common feature of both types of waste is that the activity concentration exceeds the upper level in the current legislation for regulating the management of the waste, including disposal. Another common feature is that it often comes from activities that are not regulated under the Radiation Protection Act and furthermore, once this waste is detected, its handling occurs without formal authorization under the Act.

2. LEGISLATION

When the activity concentrations of ^{238}U and ^{232}Th decay series radionuclides in NORM waste are above 1 Bq/g but do not exceed 10 Bq/g, the waste can be

handled and disposed of according to the guidelines in regulation SSMFS 2011:4. For ⁴⁰K, the corresponding lower and upper activity concentrations are 10 and 100 Bq/g, respectively. There is also a special regulation, SSMFS 2012:3, on the management of peat ash and wood ash from energy production facilities. The ²³⁸U activity concentration in peat used for energy production may not exceed 2.5 Bq/g. The ash is regarded as contaminated if the activity index I_2 is greater than 1 or if the ¹³⁷Cs activity concentration is greater than 1 Bq/g and the ash may be recycled only if the activity index I_1 is less than 1 and the ¹³⁷Cs activity is less than 10 Bq/g, where:

$$I_1 = \frac{C_{232Th}}{1} + \frac{C_{238U}}{1} + \frac{C_{40K}}{20} \quad (1)$$

and

$$I_2 = \frac{C_{232Th}}{0.2} + \frac{C_{226Ra}}{0.3} + \frac{C_{40K}}{3} \quad (2)$$

and C_{232Th} , C_{238U} , C_{226Ra} and C_{40K} are the activity concentrations of ²³²Th, ²³⁸U, ²²⁶Ra and ⁴⁰K, respectively, in dry matter (Bq/g). The requirements for NORM waste disposal are summarized in Table 1.

TABLE 1. REGULATORY REQUIREMENTS FOR DISPOSAL OF NORM WASTE

U-238 and Th-232 series activity concentrations (Bq/g)	Requirements for disposal	Post-disposal radiation protection requirements
≤1	General exemption from the RPA	General exemption from the RPA
>1 and ≤10	No licence required but the general responsibilities under Section 6 of the RPA remain	General exemption from the RPA
>10	Licence required	Licence required for the disposal site (alternatively: exemption after individual regulatory review)

Note: RPA — Radiation Protection Act.

3. WASTE TYPES AND THE WASTE OWNER

The waste consists of objects usually coming from a variety of activities where they have come into contact with liquids or gases containing radionuclides of natural origin at moderate activity concentrations. The waste may comprise, among other things, containers, heat exchangers, pipes and valves of various origins and sizes. Many of those objects were demolished which hinders decontamination by conventional methods such as blast cleaning. Another type of NORM waste of interest in this study is ion exchange resin from water filters coming from water treatment facilities and private householders. The industrial facilities at which the waste is discovered, usually metal recycling companies equipped with portal systems for radiation detection, store the material at their yards pending further action. When the waste generator cannot be traced, the finder of the waste becomes the new owner.

4. ACTIONS

Since the amount of waste will normally increase over time and since the relevant industrial activities are managing and storing the waste without a licence, it is important that a radiological evaluation be made and an appropriate solution be found. With the intention of acquiring more knowledge of the NORM waste that has been detected in a number of scrap recycling facilities, the Swedish Radiation Protection Authority made several visits during 2009–2011 to examine the scrap that had been collected. Meanwhile, a number of samples of material were taken for analysis.

5. METHODS

The material samples were analysed by gamma spectrometry. A fictitious radioactive source term was created with the aim of using it in some exposure scenarios for the estimation of the radiation hazards from the contaminated material. To determine the source term, the activity concentration of the 75th percentile of the values from the analysed material was used. The exposure scenarios included external gamma exposure to scale attached to metal and ion exchange resins from water filters. Internal exposure from inhalation was also considered. Furthermore, a scenario involving the spread of radioactivity as a result of a fire was considered in order to study impacts on the surrounding environment. Finally, the ion exchange resin was assumed to have been disposed

of after various degrees of dilution in areas of various sizes and depths to study the risks due to external radiation and the radiological impacts to the environment.

6. RESULTS

About 90% of the waste appeared to be metallic. About 65% of material in the form of scale and water filters had activity concentrations exceeding 10 Bq/g. Radium-226 was the dominant radionuclide, with a mean activity concentration of 55 Bq/g. In one scenario, it was calculated that a small pile of contaminated scrap metal and ion exchange resin could raise the local natural background radiation level by 1–2 mSv/a. The handling of the waste in the pile could cause workers to receive a dose of about 0.5 mSv/a via the inhalation pathway. A scenario involving an extensive fire in the pile indicated that the migration of radionuclides would give rise to enhanced radiation levels in the environment. Finally, assessments based on the disposal scenario showed that the estimated absorbed dose rates to biota were higher than the screening dose rate value of 10 μ Gy/h [1].

7. DISCUSSION

The radiological hazards associated with accumulated metallic and non-metallic NORM waste have been examined. The waste, which is presently accumulated in several places in Sweden, has activity concentrations exceeding the upper bound of the scope of application of current legislation. Regarding the metallic waste, the investigation suggests that specific actions need to be taken to reduce the waste volume and to dispose of the radioactive constituents in a safe manner. The study also points out that facilities such as those for metal recycling need to be subject to an authorization to handle and to store NORM waste. From a radiation protection point of view, this will lead to safer workplaces and environment.

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LIMITATIONS ON THE ACTIVITY CONCENTRATION OF MINERAL PROCESSING RESIDUES USED AS BUILDING MATERIAL

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Abstract

The use of mineral processing residue as a building material accounts for a significant proportion of the various uses of such material. The activity concentrations of radionuclides of natural origin in the material can be significantly elevated, resulting in enhanced exposure of occupants of the buildings concerned unless some restriction is placed on the radioactivity content. Accordingly, in order to comply with the dose limitation requirements of the relevant laws and standards, the activity concentration in the material has to be restricted. The use of mineral processing residues in building material is controlled in three categories according to a parameter known as the equivalent concentration, which can guide the product specification of the building material and control the dose received by a building occupant.

1. INTRODUCTION

With the expansion of industrial development, various amounts of mineral processing residue are used as building material. Some of these residues have a significant radioactivity content, which can cause an increase in the exposure of building occupants, and the associated health impacts have attracted the attention of the public [1, 2]. Limitation of the activity concentration of mineral processing residues is an effective way of controlling the exposure of building occupants.

2. DOSE MODEL

2.1. Model design

A representative dwelling is shown in Fig. 1 [3]. The internal dimensions of the dwelling are 4 m × 3 m × 2.8 m, with a window in one wall and a door facing the window. In order to estimate the effective dose, the following assumptions are made: (a) the dwelling is made of the same building material throughout; (b) the surface structure of the building material is similar throughout; and (c) the window and door are kept closed.

2.2. Activity index

The activity index for internal exposure is calculated by considering the contributions of ^{222}Rn and ^{222}Rn . The index, I_{in} , is defined as follows:

$$I_{\text{in}} = \frac{C_{\text{Ra}}}{200} + \frac{C_{\text{Th}}}{650} \quad (1)$$

where C_{Ra} and C_{Th} are the activity concentrations of ^{226}Ra and ^{232}Th in the building material, respectively (Bq/kg).

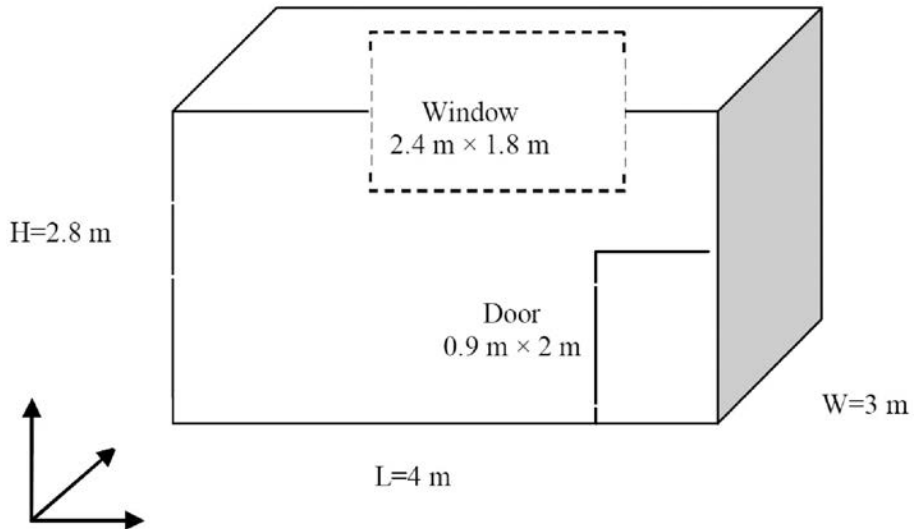


FIG. 1. The model house.

For external exposure, the index, I_γ , of national standard GB 6566-2001 was used, namely:

$$I_\gamma = \frac{C_{Ra}}{370} + \frac{C_{Th}}{260} + \frac{C_K}{4200} \quad (2)$$

where C_K is activity concentration of ^{40}K (Bq/kg). According to European Union guidance, the permissible incremental gamma dose is 0.3–1 mSv/a [4], so the maximum value, 1 mSv/a, was chosen, and then calculated for $I_\gamma = 4$.

Assuming that the building material is a mixture of mineral processing residue and a non-radioactive component and that the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the non-radioactive component are 41, 48 and 666 Bq/kg, respectively, the following expressions are obtained from Eqs (1) and (2):

$$\frac{fC_{Ra_1} + 41(1-f)}{370} + \frac{fC_{Th_1} + 48(1-f)}{260} + \frac{fC_{K_1} + 666(1-f)}{4200} \leq 4 \quad (3)$$

and

$$\frac{fC_{Ra_1} + 41(1-f)}{200} + \frac{fC_{Th_1} + 48(1-f)}{650} \leq 1 \quad (4)$$

where f is the fraction of mineral processing residue in the building material and C_{Ra_1} , C_{Th_1} and C_{K_1} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively, in the mineral processing residue. Solving the inequalities for $f \leq 15.4\%$, external exposure is the limiting parameter, giving:

$$3.25C_{Ra_1} + C_{Th_1} \leq 3300 \text{ Bq/kg} \quad (5)$$

At the same time, the limit value is consistent with national standard GB 18871-2002, so the activity concentration of ^{226}Ra is no more than 1000 Bq/kg [5].

3. LIMIT ON ACTIVITY CONCENTRATION

It is important to limit the activity concentration of mineral processing residue when used in building material, and the radiation risk is mainly from ^{226}Ra , ^{232}Th and ^{40}K . The ^{40}K contribution was ignored because the effective dose from this radionuclide is very small (often 10^{-5} mSv/a). Considering internal and external exposure pathways, the equivalent activity concentration C_e is used as the limiting parameter, where:

$$C_e = C_{\text{Ra}} + 1.3C_{\text{Th}} \quad (6)$$

To avoid confusion, activity concentration limitation was calculated under different conditions, based on regulations and laws such as those on radioactivity limits in building material and on basic standards for protection against ionizing radiation and for the safety of radiation sources.

3.1. Unrestricted use

When the activity concentrations in mineral processing residue are $C_e < 350$ Bq/kg and $C_{\text{Ra}} < 200$ Bq/kg, its use and production is not subject to any restriction. The incremental annual dose from external gamma exposure is less than 0.5 mSv, which corresponds to the dose associated with living in soil dwellings. Building material containing this type of mineral processing residue can be used for the construction of, among other things, hotels, hospitals, schools, offices, markets and exhibition rooms.

3.2. Restricted use

When the activity concentrations in mineral processing residue are $350 \leq C_e \leq 1350$ Bq/kg or $200 \leq C_{\text{Ra}} \leq 1000$ Bq/kg, its use needs to be restricted to the following:

- (a) Structures around which people rarely spend much time, such as roadbeds, bridges and dams, where it can be used directly (i.e. without mixing with non-radioactive material);
- (b) Buildings with a low occupancy factor, such as gymnasiums, stadiums and storerooms, where it can be used only when mixed with non-radioactive material.

3.3. Prohibited use

When the activity concentrations in mineral processing residue are $C_e \geq 1350$ Bq/kg or $C_{Ra} \geq 1000$ Bq/kg, its use as a building material is to be prohibited. The material is to be stored according to standard GB 18599 or used only for special purposes under supervision or management in terms of an appropriate authorization.

4. POSSIBILITIES FOR THE USE OF RESIDUES

Mineral processing residues were analysed in terms of activity concentration and amounts generated, as found in the literature and the database of the China Institute of Atomic Energy. Several types of residue were investigated, including coal gangue, bone coal slag, coal ash, smelting slag and industrial gangue. Results for representative residues are given in Table 1 and show that 97.2% of mineral processing residue can be freely used, 2.3% is subject to restrictions on use and 0.5% is prohibited for use. This control measure is consistent with the national law on reuse and recycling.

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TABLE 1. AMOUNTS OF MINERAL PROCESSING RESIDUES GENERATED AND RESTRICTIONS ON USE

Type of residue	Total	Amount generated (Mt)		
		Unrestricted use $C_e < 350$ Bq/kg	Restricted use $350 < C_e < 1350$ Bq/kg	Prohibited use $C_e < 1350$ Bq/kg
Mining (45%)	873	864 (99%)	8.5 (1%)	0.6 (0.07%)
	1.2	0.53 (44%)	0.26 (21%)	0.41 (35%)
Electricity and gas (19%)	37.4	34.5 (92%)	2.2 (6%)	0.7 (2%)
Melting industry (17%)	42.5	37.7 (89%)	2.2 (5%)	2.6 (6%)
	42.8	33 (77%)	9.8 (23%)	0
Chemicals and coloured metal (12.5%)	2.9	2.5 (86%)	0.3 (12%)	0.06 (2%)
Total	999.8	972 (97.2%)	23.2 (2.3%)	4.4 (0.5%)

CHARACTERISTICS AND DISPOSAL CATEGORIZATION OF SOLID RADIOACTIVE WASTE FROM THE FRONT END OF THE URANIUM FUEL CYCLE

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Abstract

The proper categorization of radioactive waste forms the basis for defining its disposal method. In particular, it is the basis for defining the disposal policy for solid radioactive waste from the front end of the uranium fuel cycle to identify scientifically its characteristics, in view of the differences in regulatory approach between artificial radioactive waste and NORM waste. The paper examines the disposal policy and practice in China and other countries for solid radioactive waste from the front end of the uranium fuel cycle and discusses the confusion in disposal of the waste as artificial radioactive waste. The radionuclide composition and characteristics of the solid radioactive waste from the front end of the uranium fuel cycle are investigated in detail and a new idea that such waste needs to be disposed of and categorized as NORM waste is proposed.

1. INTRODUCTION

As of 31 December 2010, approximately 900 m³ of solid radioactive waste containing uranium (with a ²³⁵U abundance greater than that in natural uranium) had been accumulated in China as scrapped filter cores, resins and residues from front end uranium fuel cycle facilities (for the purification, conversion, enrichment of uranium and fuel element fabrication). With the expansion of nuclear power and the associated demand on nuclear fuel fabrication, the quantity of such waste is expected to increase steadily. At present, such waste is kept in temporary storage at the facilities owing to a lack of a proper disposal strategy, representing a potential hazard to human health and the environment. Proper waste categorization is the key to determining the disposal strategy for radioactive waste. When compared with natural uranium, the uranium in the solid radioactive waste from the front end of the uranium fuel cycle ('UF waste') differs

only in the abundance of uranium isotopes as a result of physical processing. Is it reasonable to require such waste to be disposed of in the same manner as for artificial radioactive waste just because it comes from nuclear facilities? Would there be a problem if it were to be managed as NORM waste according to its characteristics? In view of the above questions, the differences in regulatory requirements for artificial radioactivity and natural radioactivity and the isotope composition and intrinsic properties of UF waste are investigated in this paper. It is proposed that, for purposes of disposal, such waste needs to be categorized as NORM waste, potentially allowing it to be disposed of with tailings from the mining and processing of uranium ore.

2. DIFFERENCES IN REGULATORY REQUIREMENTS BETWEEN ARTIFICIAL RADIOACTIVITY AND NATURAL RADIOACTIVITY

Neither IAEA Safety Standards No. GSR Part 3, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards [1], nor the Basic Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (GB18871-2002) of China [2] make any explicit distinction between regulatory requirements for artificial radioactivity and those for natural radioactivity in terms of the system of radiation protection. In practice, however, differences in regulatory approach have emerged gradually as more and more attention has been focused on NORM associated with industrial activities.

2.1. Differences in exemption criteria

Based on the regulations in GB18871-2002 [2], a practice or a source within a practice may be exempted without further consideration provided that the following criteria are met in all feasible situations:

- (a) The effective dose expected to be incurred by any member of the public due to the exempted practice or source is of the order of 10 μ Sv or less in a year.
- (b) Either the collective effective dose committed by one year of performance of the practice is no more than about 1 man Sv or an assessment in terms of the optimization of protection shows that exemption is the optimum option.

It is recommended in IAEA Safety Standards Series No. RS-G-1.7, Application of the Concepts of Exclusion, Exclusion Clearance [3], that, for material containing only radionuclides of natural origin, regulation as a practice is unnecessary if the concentrations of radionuclides in the uranium and thorium

decay series do not exceed 1 Bq/g. It is noted in this regard that doses to individuals as a consequence of these activity concentrations would be unlikely to exceed about 1 mSv/a.

2.2. Entry level for regulation of NORM

In China, the regulatory starting point for natural radionuclides is an exemption level which is the same as that for artificial radionuclides. Similarly, the activity level for releasing regulatory control is the clearance level. However, it has been highlighted that exemption levels for NORM can be significantly higher than 1 Bq/g. The IAEA criterion for exemption of NORM is based on dose (i.e. a dose of the order of 1 mSv/a [1]). The European Commission has also established a criterion of 1 mSv/a for workers (for members of the public it is 0.3 mSv/a) [4]. Values of exempt activity concentration consistent with this dose criterion are generally much higher than 1 Bq/g, particularly for ²¹⁰Pb and ²¹⁰Po, as shown in Table 1.

TABLE 1. ACTIVITY CONCENTRATIONS OF RADIONUCLIDES OF NATURAL ORIGIN BELOW WHICH REGULATORY CONTROL IS UNLIKELY TO BE NECESSARY

	Activity concentration (Bq/g)
European Commission exemption values [4]	
U nat, Pb-210+, Po-210	5
Th-230	10
Ra-226+	0.5
U-238, Th-232 series activity concentrations corresponding to 2 mSv/a [5]	
Bulk material (e.g. ore body and mineral stockpile)	5 or more
Material in small quantities (e.g. mineral concentrates, scale and sludge)	50 or more
Volatilized furnace fume and precipitator dust (Pb-210, Po-210)	500 or more

2.3. Differences in disposal requirements for radioactive waste

Based on IAEA Safety Standards Series No. GSG-1, Classification of Radioactive Waste [6], very low level waste (VLLW) includes waste arising from the operation and decommissioning of nuclear facilities with levels of activity concentration in the region of or slightly above the levels specified for the clearance of material from regulatory control and waste containing naturally occurring radionuclides, which originate from the mining or processing of ores and minerals. An adequate level of safety for VLLW may be achieved by its disposal in engineered surface landfill type facilities. This is the usual practice for waste from some mining operations and for other waste containing naturally occurring radionuclides from various operations involving mineral processing and other activities. Some countries also use this disposal method for waste with low levels of activity concentration arising from nuclear installations.

It is expected that with a moderate level of engineering and controls, a landfill facility can safely accommodate waste containing artificial radionuclides with levels of activity concentrations one or two orders of magnitude above the levels for exempt waste, for waste containing short lived radionuclides and with limited total activity. In general, for waste containing radionuclides of natural origin, acceptable levels of activity concentration will be expected to be lower than those for waste containing artificial radionuclides, in view of the long half-lives of such radionuclides.

Mine tailings resulting from the processing of uranium and thorium ores generally contain elevated levels of radionuclides of natural origin and are required to be managed as radioactive waste for radiation protection purposes and safety reasons. These tailings contain most of the decay progeny of the parent radionuclide. Some of the progeny may be more susceptible to leaching and emanation from the tailings than from the original ore. In addition, the tailings contain significant amounts of hazardous chemicals, including heavy metals such as copper, arsenic, molybdenum and vanadium. These need to be considered in assessing the safety of planned management options.

Similar types and quantities of radioactive waste containing radionuclides of natural origin also arise from the extraction and/or processing of certain other materials [5]. These materials include phosphate minerals, mineral sands, some gold bearing rocks, coal and hydrocarbons, and contain long lived radionuclides at relatively low concentrations. The concentrations of the radionuclides in these waste streams may exceed the levels for exempt waste. Some waste, such as scales arising in the oil and gas industry, may have very high activity concentrations. These may necessitate the management of waste in the same way as low level waste (LLW) or, in some cases, intermediate level waste (ILW).

CATEGORIZATION OF SOLID RADIOACTIVE WASTE

Sources and types of radioactive wastes have been listed in annex III of IAEA Safety Standards Series No. GSG-1 [6], which includes wastes from mining and milling (containing high levels of natural radionuclides) and wastes from nuclear power plants. However, there is no description of solid UF waste. The categorization of such waste in No. GSG-1 is unclear.

It can be concluded from above analysis that:

- (a) NORM waste, including that from the mining and processing of uranium ore, belongs to the VLLW category, except in a few circumstances where it has particularly high activity concentrations.
- (b) Disposal of NORM waste can generally be accomplished in a landfill facility for VLLW.
- (c) The upper limits for VLLW are one or two orders of magnitude higher than exemption levels for artificial radionuclides.
- (d) The appropriate category for UF waste is unclear.

3. DOUBTS ABOUT THE CATEGORIZATION OF UF WASTE FOR PURPOSES OF DISPOSAL

The current radioactive waste classification standard in China (GB9133-1995) was developed in accordance with the radioactive waste classification guideline published by the IAEA in 1981, which is applicable to pre-disposal management. According to the classification standard and the Law of the People's Republic of China on Prevention and Control of Radioactive Pollution, UF waste is categorized as LLW, which should be disposed of in a local near surface disposal facility. However, with the improvement of people's living standards and the opportunities for public participation, it is very difficult to select sites for near surface disposal facilities for LLW and ILW in China. Thus, UF waste has not been disposed of in a timely manner.

The categorization of UF waste in the radioactive waste classification guide of the IAEA is unclear. In the management policies and practices for UF waste in the United States of America, France, Japan and other countries, however, it is common practice for such waste to be managed and disposed of as LLW. But this concept of waste classification brings difficulties for the implementation of waste disposal.

In addition to the fact that UF waste has not been disposed of in a timely manner in China, as stated in above, there is no agreed practice for the disposal of

such waste elsewhere in the world. The main reason is that there are doubts of the categorization of UF waste, as shown by the following discussion:

- (a) It would be logical for UF to be categorized as LLW based on its activity level if such waste had to be managed as artificial radioactive waste. However, compared with natural uranium and its progeny in NORM, which may have been subjected to both physical and chemical processing, the uranium in UF waste is changed only in the abundance of uranium isotopes by physical methods.
- (b) Is it reasonable to require that such waste be disposed of as an artificial radioactive waste just because it comes from nuclear facilities?
- (c) Can UF waste be managed and disposed as VLLW in the same way as NORM waste?
- (d) Would there be problems if it is managed as NORM waste according to its characteristics? Would the level of safety be compromised?

How to solve these questions? It is considered that the characteristics of the waste should form the basis for its disposal categorization. To address this issue, it is useful to analyse the characteristics of UF waste.

4. COMPARATIVE ANALYSIS OF THE CHARACTERISTICS OF UF WASTE

UF waste mainly includes waste filter cores, resins, equipment and residues containing ^{238}U , ^{235}U and ^{234}U and other radionuclides of natural origin, mostly as triuranium octaoxide, uranium dioxide and uranium hexafluoride. Its main characteristics are long half-life, low activity and natural radioactivity. The hazards associated with UF waste come mainly from internal exposure to alpha emitters. However, some progeny radionuclides such as ^{234}Th , ^{234}Pa and ^{231}Th are beta emitters. The radioactivity of UF waste is mainly from low enriched uranium. The activity concentration of low enriched uranium can reach 10 000 Bq/g, mainly ^{238}U , ^{235}U and ^{234}U , the mass percentages of which are approximately 94.95%, 5% and 0.05%, respectively. However, the activity concentrations are generally lower than this because of the low concentration of uranium in the waste. On the other hand, the activity concentration of ^{226}Ra in uranium tailings can reach 100 Bq/g and radionuclide activity concentrations in some NORM facilities are even higher. Therefore, the activity concentration of UF waste is at the same level as that of wastes from some NORM facilities.

From the perspective of radionuclide composition, the radionuclides in UF waste are mainly three isotopes of uranium and almost no progeny. Radionuclides in uranium mining waste rock are associated with the complete decay chains of the uranium series. Tailings from the processing of uranium ore mainly contain ^{226}Ra , a relatively long lived progeny of ^{238}U , together with its own progeny. NORM waste can contain the whole decay chains of the uranium or thorium series, or only progeny such as ^{226}Ra , ^{210}Pb and ^{210}Po . Two radionuclides of particular importance are ^{222}Rn and ^{220}Rn from uranium waste and other NORM waste, which can cause prolonged internal exposure of the lung. Therefore, the hazard posed by UF waste to the environment would be smaller than that of NORM waste if the activity concentrations were at the same level.

From the perspective of radionuclide accumulation mechanisms, ^{235}U and ^{234}U in UF waste are transferred from one part of natural uranium to another by diffusion or centrifuging forming depleted uranium and enriched uranium. Materials associated with uranium mining and processing remain as natural uranium, which simply becomes more concentrated. The progeny end up in enhanced concentrations in the tailings. Thus, the radionuclides in NORM wastes may well be physically and chemically concentrated, but without the occurrence of a nuclear reaction, without the production of new radionuclides, or without the production of any additional radioactivity. The radionuclides are simply transferred from one part of the material to another part during the whole process and redistributed by human activity.

It can be concluded from the above analysis that UF waste needs to belong to the category of NORM waste.

5. COMPARATIVE ANALYSIS OF DISPOSAL SAFETY OF UF WASTE

As stated above, UF waste contains only radionuclides with long half-lives, which is the same as for NORM waste. Due to the long timescale, there will be an ingrowth of a large number of progeny such as ^{226}Ra and ^{222}Rn . The activity of low enriched uranium is determined by the number and characteristics of the progeny. The activity of low enriched uranium will increase during a 1000 to 1 million year period as a result of the large ingrowth of ^{222}Rn , ^{210}Pb and other hazardous radionuclides.

Short term effects and long term effects are both considered in the safety assessment of radioactive waste disposal. The effects on short term safety are almost the same for landfill facilities for VLLW and disposal facilities for LLW and ILW. There is essentially no difference between the effects on long term safety of landfill facilities for VLLW and the effects on disposal facilities for LLW and ILW because these two kinds of disposal facilities are both near surface

facilities. Thus, the management of UF waste as NORM waste, for instance by disposing it along with uranium mining and processing waste, does not affect the safety of disposal.

6. CONCLUSION

UF waste has been managed in accordance with regulations for artificial radioactive waste in China. The possible reason is that the facilities for the front end of the uranium fuel cycle belong to nuclear facilities and the wastes from them need to be managed in accordance with unified radiation protection requirements. However, the choice of disposal methods for radioactive waste are to be based on the characteristics of the waste, not the sources of the waste. It is concluded that the UF waste belongs to the category of NORM waste and it is scientific and safe to define the disposal strategy on this basis. Thus, uranium tailings repositories can be potential disposal sites for UF waste.

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RAPPORTEUR SUMMARY OF TOPICAL SESSIONS 4, 5 AND 6

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1. INTRODUCTION

This report briefly summarizes Sessions 4, 5 and 6 of the Seventh International Symposium on Naturally Occurring Radioactive Material (NORM VII) in Beijing, China. Short accounts of each of the 19 papers presented in these sessions are outlined.

2. SITE SPECIFIC MEASUREMENTS, DOSE ASSESSMENT AND APPLICATION OF THE ALARA PRINCIPLE

2.1. Exposure assessment of workers handling industrial NORM in Japan

A survey carried out in Japan on NORM industries such as monazite, zirconium and titanium to establish a database in reference to Japan's 'NORM Guideline' was presented. Minerals are mainly imported in Japan, except limestone, and include monazite, zircon, bastnasite, titanium, phosphate and coal. Exposure to natural radionuclides in radon spas and consumer products are also observed. Exemption levels are established for nuclear fuel or source material, but no such values are available for NORM. A report on the regulation of NORM published in 2003 provides a 1 mSv/a dose criteria for exemption. The 2009 Guideline for ensuring safety of raw materials containing uranium and thorium requires detailed investigation of worker exposures, such as dose rate measurements and airborne dust at workplaces, among other things. The effective doses to workers in NORM industries are estimated to be less than 1 mSv/a in most cases with a highest dose at about 0.5 mSv/a in a zircon electro cast plant.

2.2. Radionuclide release from the combustion of coal: A case study

The paper provided information on the radionuclide release from the combustion of coal where coal-uranium-polymetallic ores are processed using blast furnaces in Yunnan Province in China. China's coal consumption was 3.2 billion t in 2010 with 500–800 million t of ash generated. The activity concentrations of ^{238}U and ^{232}Th in coal are mostly less than 0.1 Bq/g, but some

sources contained up to 5.6 Bq/g ^{238}U and 29 Bq/g ^{232}Th . Some of the lignite based coal sources showed radionuclide concentrations above 1 Bq/g. Activity concentrations of ^{238}U , ^{226}Ra , ^{210}Pb , ^{210}Po , ^{232}Th and ^{40}K are enhanced in fly ash (especially lead and polonium). In contrast, enhancements are also observed in bottom ash except for lead and polonium. Activity concentrations in ash are higher than exemption levels. Some radioactivity (^{238}U , ^{226}Ra , ^{210}Pb , ^{210}Po and ^{232}Th) is released to the atmosphere, ranging from 1.25×10^8 Bq/a ^{232}Th to 5.08×10^{10} Bq/a ^{210}Po . Nearly 71% of lead and 89% of polonium are discharged to atmosphere and releases via off-gases greatly exceed losses from fly ash. Activity concentrations in the ambient environment are ten times background, requiring regulatory control.

2.3. Occupational exposure in ammonium phosphate fertilizer plants

The study initiated to close knowledge gaps concerning activity concentrations and occupational exposures in two plants producing ammonium phosphate fertilizers (mono-ammonium phosphate and di-ammonium phosphate) in Spain. These plants use mostly Moroccan rock averaging 1.5 Bq/g ^{238}U and 0.02 Bq/g ^{232}Th . The annual effective doses estimated are less than 1 mSv/a and the external and inhalation components are similar. The dose due to inhalation of airborne dust is about 0.1 mSv/a (radon is at background levels). No additional measures for radiation protection need be considered.

2.4. Remediation of a NORM contaminated site in Austria

This paper was on a small contaminated area that was detected and remediated around a school near Vienna with ^{226}Ra contamination. Between 1910 and 1917, a chemical plant produced radium bearing products but ceased all related activities in 1925. Description of the strategy to control the area, applied measurement methods, results and their consequences were provided. Dose rates ranged 100–250 nSv/h with hotspots up to 800 nSv/h in workshops. Elevated radon up to 20 500 Bq/m³ in offices was measured, and offices were relocated and long term radon monitoring was started. Demolition plan was put into operation including aerosol measurements, personal dosimetry and remediation of hotspots and on-site high purity germanium gamma analysis of soil filled barrels. Some of the activities include detection of subsurface contamination, numerous on-site measurements, applied thresholds for clearance levels and the methodology to derive them and RESRAD simulations on dump sites. The contaminated soil up to 2 Bq/g was covered by 2–4 m of fresh soil and monitoring has continued after the school was established in 2010.

2.5. Mobile unit for site characterization in environmental remediation projects

Site characterization is an important step in environmental remediation and is typically expensive and time consuming. On-site real time analysis is always preferred in such situations. A data acquisition and control module is developed to support the characterization. The system is field tested at remediation sites with IAEA cooperation. Components can be customized to site requirements with easy sample scheduling, and the system is useful for environmental gamma, soil analysis, radon/thoron, some nuclide specific evaluations and the measurement of radioactive aerosols. Some of the features include chemical sensors because sites frequently contain contaminants other than NORM, vacuum/detector system for mop test, simultaneous operation of all detectors and remote data transmission.

2.6. ^{222}Rn , ^{220}Rn and progeny measured in a limestone cave and the associated radiation dose

This study reported ^{222}Rn , ^{220}Rn and their progeny measured at different locations inside a limestone cave using CR-39 and LR-115 Type II solid state nuclear track detectors. Attached and unattached fractions were determined and committed equivalent doses due to ^{218}Po and ^{214}Po were calculated for the respiratory tract. Inhalation doses were modelled using the ICRP HRT model, and it was found that doses increase with distance from the cave entrance. The effective dose is 17–19 μSv per hour of exposure, suggesting that visitors must spend less than 1 h/d inside the cave.

2.7. Hazard assessment at the site of a former coal fired power plant

The EU precautionary principle (2000) is to be applied where there is a reasonable suspicion of the existence of health or environmental risk at contaminated sites. The reported study is from legacy sites in Croatia: coal sludge and fly ash in a coastal impoundment. The site was transferred from the Government to new owners without clear responsibility lead to hazard characterization in order to establish a remediation plan. It is noted that “radioactivity never comes alone”, and the Ra(eq) concept with hazard indices was used for the site evaluation. The best solution was to allow nature to reclaim it. NORM measurements are underway for this colony and the adjacent marine system beyond the impoundment wall. Extensive contour mapping with identification of hotspots near seaside dam wall was carried out. Precise GPS and dose rate meter are required for such studies.

2.8. Worker exposure at a copper mining and beneficiation facility

Information is presented on occupational exposures, smelter and manufacturing at Palabora Mining Co., a copper mine in South Africa. Copper concentrate classified as NORM under national regulations and specific nuclear authorization has existed for the last twenty years. Safety assessment for worker exposure is required. The latest analysis showed ^{238}U at 1.43 Bq/g in copper concentrate and 3.52 Bq/g in extraction tailings, and 0.56 Bq/g ^{232}Th in concentrate. Recommendations were made to guide the future monitoring strategy for external gamma, inhalation of airborne dust, and radon/thoron progeny. Contaminants were homogeneously distributed in the plant, which reduces sampling density and most areas exposures were less than 1 mSv/a. Annual and biannual surveys are to be considered and ongoing monitoring strategy is not overly conservative and is flexible.

2.9. Poster presentations

There were 16 poster papers presented in the session and can be classified into energy sector (3), other mining and processing (4), instrumentation and techniques (3), environmental (3) and other occupational (3).

3. NORM: MEASUREMENT METHODS AND STRATEGIES

3.1. Application of the gamma spectrometry sourceless efficiency calibration method to the measurement of radionuclides in rare earth residues

This paper investigated and analysed NORM residues from rare earth smelting and separation plants in Jiangsu Province using the high purity germanium gamma spectrometry sourceless efficiency calibration method, which was verified by IAEA reference materials. Considerable disequilibrium in uranium and thorium decay series was observed. The study concluded that feeds should ideally be managed for NORM content and waste (water) streams be segregated.

3.2. Evaluation of NORM measurement results by means of statistical methods

A common task is the assessment of random samples, drawn from a large population, in order to assess whether radiological criteria are met at areas

contaminated with NORM. From measurements of NORM (i.e. residues and contaminated areas), usually by gamma spectrometry, a sample of measurement results (Bq/g and Bq/cm²) was generated. A test statistic (e.g. median, mean, 95th percentile and 95% upper confidence limit of mean) was determined from the data by means of a statistical evaluation algorithm for reliable information. This would be useful for comparing with the current control limits of NORM regulations or used as a basis for dose assessment.

Different statistical methods for the calculation of the test statistic were presented:

- (a) The classical statistical methods compared with the modern numerical methods;
- (b) Robust, efficient and non-parametric numerical procedure — the ‘bootstrap method’ with a modification;
- (c) A recently developed technique for making certain kinds of statistical inferences (resampling method), which requires modern computer power to simplify the often intricate calculations of traditional statistical theory and application of the bias correction and acceleration method.

The various methods (classical and numerical) were applied to measured results. As the relevant test statistic, they defined the upper confidence limit (UCL) of the mean. The UCL was estimated from several random samples of size 20 drawn from a population (244 measured values of ²²⁸Ra mass related activity of a contaminated ground area). The results of application of classical and numerical statistical methods on the test examples were compared and discussed in the paper.

3.3. Anomalies in natural background levels associated with minerals with elevated radionuclide concentrations

The results of terrestrial natural background measurements in seven Chinese provinces were discussed. They were conducted as part of China’s national geochemical mapping project, with an area of 6.8 million km². Gamma doses at 1 m above ground surface due to terrestrial radionuclides indicated locations of elevated dose rates due to human activities involving minerals with elevated activity concentrations. The study was coupled with other environmental monitoring data to define the relationship between mining pollution and terrestrial gamma dose rates. Dose rates calculated by the Beck formula and 300 nGy/h defined as high. Increased regulatory awareness and supervision are considered.

3.4. Natural radioactivity source term based on remote sensing data

This paper described the basic principles for applying satellite remote sensing technology to the investigation of natural radioactivity. The relationship between areas of natural background anomalies and geological characteristics was analysed systematically. The supervised classification method and spectral angle mapping were used for the extraction of remote sensing information. Geological deposits with higher gamma radiation levels identified and described followed by on-site investigation. Remote sensing can rapidly discover higher radiation regions which may require regulatory control.

4. NORM RESIDUES

4.1. Towards a sustainable solution to NORM residue management safety

The lack of harmonious international system for management of NORM residues and the need for graded approach to regulation was highlighted. Efforts need to be made to reach an international consensus regarding NORM residue reuse and recycle with attention to transboundary movement of NORM. It was emphasized that stakeholder awareness of radiation risk needs to be improved.

4.2. Experience from remediation of thorium contaminated sites: Radiation protection and waste management issues

This paper described the practical radiation protection and waste management issues arising from the decommissioning and demolition of sites contaminated with thorium from past industrial practices. Description of the radiological objectives established at the start of decommissioning, practical radiation protection procedures used to deliver these objectives and strategies and methods used for management of radioactive wastes were discussed.

4.3. Disposal of rare earth NORM residues in Jiangsu Province, China

The present situation of NORM residues arising from rare earths extraction and separation in Jiangsu Province was investigated and the results were presented. The management of these residues was discussed and analysed with respect to the separation technology, the activity concentrations, the total residue quantities and the regional environment and ecology features. Suggestions were made for residue disposal considering national laws, regulations and standards.

4.4. Disposal of some types of NORM that exceed 10 Bq/g in Sweden

Swedish Radiation Safety Authority regulation (SSMFS 2011:4) specifies NORM exemption levels and regulates how NORM should be handled. Disposal of NORM with activity concentration levels above 10 Bq/g per radionuclide in the ^{238}U , ^{232}Th decay chains is not included in the regulation. Examples of such items include scrap metal with radioactive scale found in metal recycling facilities and uranium contaminated filters derived from water treatment. The study provided a basis for how the final disposal of these NORM contaminated items will be safely handled.

4.5. Limitations on the activity concentration of mineral processing residues used as building material

Mineral processing residue is used as a building material in large amounts, and its use is to be limited to avoid unnecessary doses to residents when activity concentrations are relatively high. Activity concentrations in the residue are limited to meet public dose limits. Three categories of residue were defined according to activity concentrations to ensure appropriate use of the material and limit residential doses, free use, restricted use and prohibited use.

4.6. Characteristics and disposal categorization of solid radioactive waste from the front end of the uranium fuel cycle

Categorization of radioactive waste defines its disposal method and disposal policy. Difference in regulatory principle between artificial and natural radioactive waste is common. Comparison of the disposal policy and practice in China with other States lead to confusion and improper disposal of the waste as artificial radioactive waste. The nuclide composition and characteristics of the waste was investigated in detail. A new concept was proposed: the solid radioactive waste from the front end of the uranium fuel cycle is to be disposed of and categorized as natural radioactive waste.

NORM TRANSPORT AND OTHER ASPECTS

(Topical Session 7)

Chairpersons

D. CHAMBERS

Canada

YONGKANG ZHAO

China

Invited Paper

ACCURATE PREDICTION OF RADIATION EXPOSURES OF WORKERS INVOLVED IN THE TRANSPORT OF NORM

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Abstract

A study of the radiation exposures encountered by workers involved in the transport of minerals and mineral concentrates containing radionuclides of natural origin was undertaken during 2008–2012. Hundreds of measurements were made during road, rail and marine transport of NORM between mining and processing sites in Australia and within and between ports in Australia, China and Japan. The investigation was focused on minerals and mineral concentrates containing thorium and uranium (including ilmenite, rutile, zircon and monazite). It was found that the use of the ‘exclusion’ factor of 10 for the concentrations of radionuclides in natural materials in the IAEA Transport Regulations is appropriate and is to be maintained. The dose rates from all potential pathways of exposure of workers could be accurately predicted, based on the concentrations of thorium and uranium in the transported material. These dose rates remain the same, irrespective of whether the transport is by road, rail or sea. The information presented in the paper allows, by the use of simple charts, the accurate prediction of doses to workers involved in the transport of NORM. It is suggested that it can be used in any assessments of exposures of workers that may be required prior to the start of the NORM transport process, by both regulatory bodies and by the mining and mineral processing industry.

1. INTRODUCTION

The transport of minerals and concentrates is a significant component of the mining and production process. This paper summarizes the results of a four year study of exposures of workers involved in the transport of NORM in the heavy mineral sands industry within Australia and between Australia and other countries (China and Japan). The study was carried out in three stages. The first stage involved measurements in Australia and was jointly sponsored by the Australian Radiation Protection and Nuclear Safety Agency (ARPANSA), the mineral sands industry and Calytrix Consulting. The second stage was focused

on obtaining more data from Australian and international transport routes and was jointly sponsored by the mineral sands industry and Calytrix Consulting. During the third stage, additional monitoring was undertaken addressing materials shipped from Australia in bulk and in containers to overseas ports and was sponsored solely by Calytrix Consulting. The detailed results can be found in Ref. [1]. This paper builds on a report on radiation exposure in the transport of heavy mineral sands compiled by Calytrix Consulting for ARPANSA [2].¹

2. HEAVY MINERAL SANDS MINING AND PROCESSING

The mineral sands ore after its collection is typically screened (to break it down into grains no larger than 2 mm) and carried by a system of pipes and/or conveyors to the primary concentrator. At the primary concentrator, heavy sands are separated from other sands using a system of gravity separators (or occasionally using ‘wet magnetic separation’). In most cases, the primary concentrate is further processed through a series of spirals to remove tailings and excess fines. This secondary concentration process may be incorporated into the same plant or may be carried out in a separate plant constructed to treat the primary concentrate from several mine sites. The final heavy mineral concentrate (HMC) is then transported to separation plants. After additional screening, magnetic and electrostatic methods are used in the separation of the concentrate into individual minerals. The conductive minerals ilmenite and rutile are separated from the non-conductive minerals zircon and monazite using electrostatic separation techniques. Magnetic separation is then used to separate the magnetic minerals ilmenite and monazite from the non-magnetic minerals rutile and zircon. Another important industry product is synthetic rutile (an upgraded form of ilmenite with a higher titanium content) produced by thermal and chemical treatment to remove iron oxides.

Materials encountered in the mineral sands industry may fall within the definition of NORM because of the presence of thorium and uranium in the mineral grains. As a rule, the ^{232}Th and ^{238}U decay series radionuclides are in equilibrium and the activity concentration in a particular mineral can be determined from the activity concentration of the parent. Typical radionuclide concentrations are shown in Tables 1 and 2.

¹ Neither this present paper nor the more detailed report [1] should be quoted as reflecting the views of ARPANSA; the results and conclusions remain those of the author. For ARPANSA’s position, the ARPANSA report [2] should be consulted.

ACCURATE PREDICTION OF RADIATION EXPOSURES

TABLE 1. TYPICAL ACTIVITY CONCENTRATIONS, INDUSTRY DATA

	Activity concentration (Bq/g)		
	Th-232	U-238	Sum
Materials transported between mines and between mines and plants			
HMC	0.5–6.0	0.3–2.5	0.8–8.5
Intermediate products, tailings returned to the mine	2.4–7.2	0.9–2.0	3.3–9.2
Materials transported from plants to customers overseas			
Zircon	0.8–1.1	3.2–3.8	4.0–4.9
Ilmenite	0.5–1.9	0.1–0.5	0.6–2.4
Rutile	0.2–0.6	0.1–0.8	0.3–1.4
Synthetic rutile	0.4–1.9	0.1–0.5	0.5–2.4
Monazite concentrate	82–143.5	9.5–20	91.5–163.5

Note: HMC — heavy mineral concentrate.

TABLE 2. TYPICAL ACTIVITY CONCENTRATIONS, THIS STUDY

	Activity concentration (Bq/g)		
	Th-232	U-238	Sum
Materials transported between mines and between mines and plants			
HMC	1.6	0.6	2.2
Intermediate products, tailings returned to the mine	5.1	1.7	6.8
Materials transported from plants to customers overseas:			
Zircon	0.9	3.0	3.9
Ilmenite and synthetic rutile	1.2	0.2	1.4
Monazite concentrate	84–94	9–14	~100

Note: HMC — heavy mineral concentrate.

3. SCOPE OF THE STUDY

The Australian Code of Practice for the Safe Transport of Radioactive Material [3] adopts the IAEA Transport Regulations², which state:

“107. The Regulations do not apply to:

.....

- (e) natural material and ores containing naturally occurring radionuclides that are either in their natural state, or have been processed only for purposes other than for the extraction of the radionuclides, and that are not intended to be processed for use of these radionuclides, provided that the activity concentration of the material does not exceed 10 times the values specified in para. 401(b), or calculated in accordance with paras 402–406” [3].

In the latest version of the IAEA International Transport Safety Regulations [4], not yet adopted in Australia, minor amendments were made to this definition of scope:

“107. These Regulations do not apply to any of the following:

.....

- (f) Natural material and ores containing naturally occurring radionuclides, which may have been processed, provided the activity concentration of the material does not exceed 10 times the values specified in Table 2, or calculated in accordance with paras 403(a) and 404–407” [4].

Because of this ten times ‘exclusion’ factor, all the minerals listed in Tables 1 and 2 with the exception of monazite concentrate are outside the scope of application of the IAEA Transport Regulations.

Several discussions were held since the publication of the first version of the report in 2008 [2], particularly with regard to the definition of ‘transport worker’. Paragraph 106 of Ref. [4] states:

² INTERNATIONAL ATOMIC ENERGY AGENCY, Regulations for the Safe Transport of Radioactive Material, IAEA Safety Standards Series No. TS-R-1, IAEA, Vienna (2005).

“Transport comprises all operations and conditions associated with, and involved in, the movement of *radioactive material*; these include the *design*, manufacture, maintenance and repair of packaging, and the **preparation, consigning, loading**, carriage including **in-transit storage, unloading and receipt at the final destination** of loads of *radioactive material* and *packages*” (italics, original emphasis; bold, author’s emphasis).

It is understood that it may be difficult to establish an exact ‘administrative boundary’ between ‘processing’ and ‘transport’ at a particular mining or mineral processing site but, in accordance with the definition, an employee whose primary tasks are associated with loading of the material in bulk (or into bags and containers), a loader operator handling the containers at the wharf and a person unpacking the containers at the destination all need to be considered as transport workers.

The main purpose of this study was to determine whether exclusion from the IAEA Transport Regulations of materials in the heavy mineral sands industry (other than monazite concentrate) is appropriate and whether the exclusion factor of ten for natural materials is also appropriate. All stages of transport of concentrates and intermediate and final products in the heavy mineral sands industry were studied in the following manner:

- (a) Transport routes and modes of transport were identified.
- (b) Gamma radiation levels were measured using gamma radiation monitors, electronic dosimeters and thermoluminescent dosimeter badges.
- (c) Airborne dust was measured using personal and area dust samplers.
- (d) The concentrations of ^{222}Rn and ^{220}Rn were measured using a portable electronic radon/thoron monitor SARAD-RTM1688-2.
- (e) Worker exposure periods were recorded for the purpose of dose assessments.
- (f) Relevant information for the period 2009–2011 was collected from the industry.

Exposure data were obtained for 19 transport routes:

- (a) Transport of primary concentrate to a secondary concentrator — two road routes;
- (b) Transport of HMC from mine sites to the separation plants — five road routes (including three with return of the tailings to a mine site), one rail route, one marine route;
- (c) Transport of tailings from the plant back to the mine site — one road route;
- (d) Transport of final products from a separation plant to a wharf (including data on the exposure of wharf workers) — three road routes;
- (e) Transport of final products to a customer overseas — six marine routes.

Measurements were also made at several Australian and overseas sites for minerals packed in containers. Although monazite concentrate, because of its high thorium content, does not qualify for exclusion from the IAEA Transport Regulations, information on seven shipments of monazite concentrate was available and is included in the study for purposes of comparison. The following additional information is given in Ref. [1]:

- (a) A detailed description of monitoring data obtained for each transport route.
- (b) Equipment and techniques used in the monitoring.
- (c) A detailed description of the dose assessment of workers involved in the transport and handling of minerals for each transport route.
- (d) An assessment of exposures of the members of the public.
- (e) Descriptions of the following:
 - (i) Unloading of the mineral in an overseas port;
 - (ii) The system for monitoring surface gamma radiation levels on trucks;
 - (iii) Possibilities of the loss of mineral through spillage during transport;
 - (iv) Relatively high natural background levels in one of the Australian ports.

4. RESULTS

A summary of the exposures of workers involved in the transport of mineral sands products is given in Table 3 and Figs 1–4. The differences between the results for ilmenite, synthetic rutile and HMC on the one hand and zircon on the other hand are probably due to the different thorium-to-uranium mass ratios in these materials and the associated dose conversion factors for exposure to airborne dust. Also, in the case of transport of synthetic rutile, the possibility of worker exposure due to thoron inhalation needs to be considered, as it contributes significantly to the overall exposure level. Higher gamma exposures are expected from a ‘combined’ activity concentration value of HMC, ilmenite and synthetic rutile (predominantly thorium) and zircon (typically more uranium than thorium). The typical exposure period for a worker involved in the transport of materials in the mineral sands industry was estimated to be about 1200–1400 h/a. Therefore, to ensure that the annual exposure of a worker does not exceed 1 mSv, the exposure rate is not to exceed 715 nSv/h. When comparing this value with the data in Fig. 4 it is clear that the factor of ten specified in the IAEA Transport Regulations as a scope defining criterion appears to be entirely appropriate for the transport of minerals associated with the heavy mineral sands industry.

ACCURATE PREDICTION OF RADIATION EXPOSURES

TABLE 3. SUMMARY OF THE RESULTS OF THE STUDY

Route	Material	Mode of transport	Activity concentration (Bq/g) ^a	Max. dose rate	
				nSv/h	μSv/a
1	HMC	Road	2.0	89	107
2 ^b	HMC	Road	4.1	331	397
3	HMC	Rail	4.3	423	229
4 ^b	HMC tailings	Road	8.0	552	276
5 ^b	HMC	Road	1.0	126 ^e 233 ^d	253 ^c 210 ^d
6	HMC	Road	1.5	151	151
7 ^b	HMC	Road	3.9 (~6)	549	604
8 ^b	HMC	Road	1.9 (4-6)	194 ^e 213 ^e	387 ^c 426 ^e
9 ^b	HMC	Road	1.6 (2.3-3)	162	227
10	HMC	Marine	5.4	490	196
11 ^b	Zircon	Road	4.1	114 ^e 214 ^d	69 ^c 442 ^d
	Ilmenite, synthetic rutile	Road	1.8	138 ^e 228 ^d	69 ^c 442 ^d
12 ^b	Zircon	Road	3.8	98	59
13 ^b	Ilmenite, synthetic rutile	Road	1.0	108 ^e 279 ^d	54 ^c 371 ^d
14	Zircon	Marine	3.8	140	168
15	Ilmenite, synthetic rutile	Marine	1.8	112	134
16	Ilmenite, synthetic rutile	Marine	1.1	52	25
17 ^b	Synthetic rutile	Marine	1.2	62	74
18 ^b	Zircon	Marine	3.9	111	134
19 ^b	Synthetic rutile	Marine	1.4	72	86

TABLE 3. SUMMARY OF THE RESULTS OF THE STUDY (cont.)

Route	Material	Mode of transport	Activity concentration (Bq/g) ^a	Max. dose rate	
				nSv/h	μSv/a
Additional assessments					
A ^b	Zircon	Container	3.9	40 ^c 135 ^f	48 ^c 162 ^f
B ^b	Thorium mineral	Container	4.7	110 ^e 184 ^f	132 ^c 221 ^f
C ^b	Monazite, for comparison (IAEA Transport Regulations apply)		90–110	1250 ^g –12508 ^h Av. 6887	150 ^g –1406 ^h Av. 512

Source: Data from Ref. [2] except where indicated otherwise.

Note: HMC — heavy mineral concentrate.

- ^a Values in parentheses refer to tailings returned to the mine.
- ^b New and/or more accurate data obtained subsequent to Ref. [2].
- ^c Dose rate for driver.
- ^d Dose rate for wharf worker.
- ^e Dose rate for loader.
- ^f Dose rate for freight handler.
- ^g Dose rate for ship loader.
- ^h Dose rate for loader in pit.

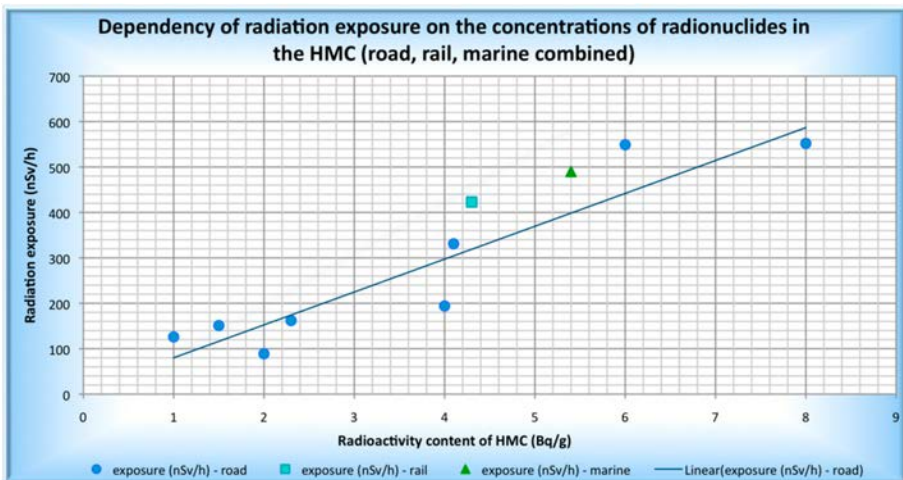


FIG. 1. Transport of HMC by road, rail and sea.

ACCURATE PREDICTION OF RADIATION EXPOSURES

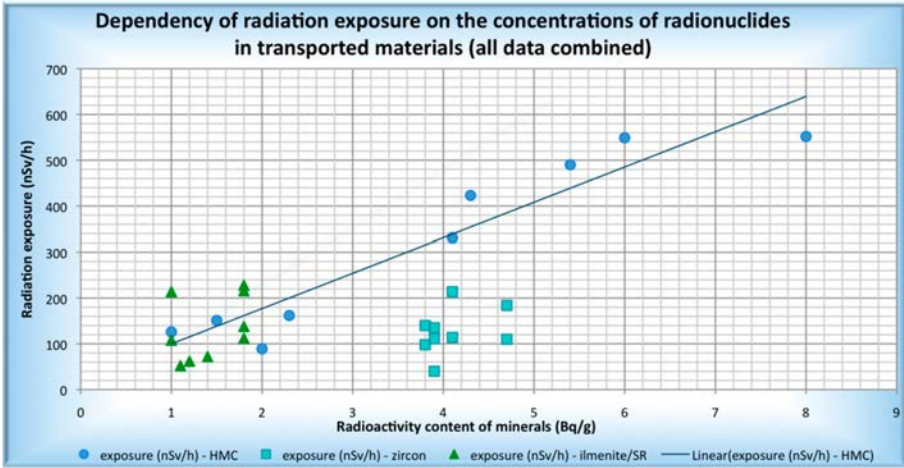


FIG. 2. All monitoring data combined.

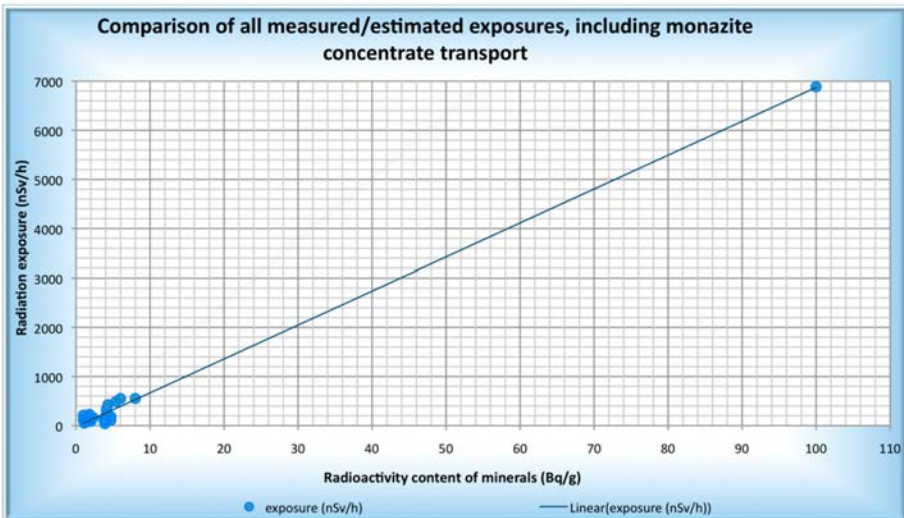


FIG. 3. All monitoring data combined (data for monazite concentrate added for comparison).

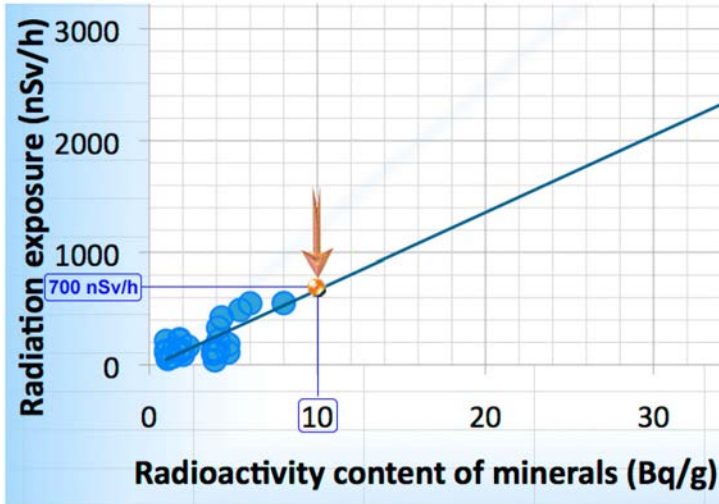


FIG. 4. A 'close-up' of the region of interest in Fig. 3.

5. CONCLUSIONS

- (1) The maximum dose received by a worker involved in the transport of minerals associated with the Australian mineral sands industry (excluding monazite concentrate) was determined to be $739 \mu\text{Sv/a}$. Therefore, the transport of such materials does not pose a significant risk to workers or members of the general public.
- (2) The use of the 'exclusion' factor of ten specified in the IAEA Transport Regulations is entirely appropriate for the heavy mineral sands industry and is to be maintained. A higher factor of, say, 15 would not be appropriate.
- (3) Exposures associated with the bulk transport of zircon are expected to be significantly lower than those associated with the bulk transport of HMC and titanium minerals with similar activity concentrations.
- (4) The highest exposure rates were those for loader operators inside the sheds at various wharves. In such situations, exposure to radon and thoron is more significant than exposure to gamma radiation and airborne dust, and the establishment of routine monitoring programmes is advisable in such situations.

ACCURATE PREDICTION OF RADIATION EXPOSURES

- (5) Relationships have been established between activity concentrations in minerals being transported and exposure rates for the workers involved. These relationships are summarized in Table 4. This information could be used to predict the exposures of workers prior to the commencement of the NORM transport process, as may be required by regulatory bodies and by the mining and mineral processing industry.
- (6) Previous publications on the transport of NORM were concerned with the application of the IAEA Transport Regulations [5] and potential problems and their solutions in international transport and trade [6, 7]. This paper and the associated report [1] complement these previous publications by providing data on **practical** measurements and assessment of **actual** radiation exposures in the transport of NORM.

TABLE 4. RELATIONSHIPS BETWEEN ACTIVITY CONCENTRATION AND EXPOSURE RATE

Activity concentration (Bq/g)	Predicted exposure rate (nSv/h)
HMC, ilmenite, synthetic rutile < 10 Bq/g (expected variance +10%)	
1	100
2	180
3	260
4	330
5	410
6	490
7	560
8	640
9	720
Typical zircon (expected variance +15 to 20%)	
3.5	140
4.0	170
4.5	200
5.0	230

TABLE 4. RELATIONSHIPS BETWEEN ACTIVITY CONCENTRATION AND EXPOSURE RATE (cont.)

Activity concentration (Bq/g)	Predicted exposure rate (nSv/h)
HMC, ilmenite, synthetic rutile, monazite ≥ 10 Bq/g (expected variance +15%)	
10	700
20	1400
30	2100
40	2700
50	3400
60	4100
70	4800
80	5500
90	6100
100	6900

Note: HMC — heavy mineral concentrate.

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ACCURATE PREDICTION OF RADIATION EXPOSURES

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RADIOACTIVITY IN ZIRCON FROM JOS, CENTRAL NIGERIA

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Abstract

Mining and processing of zircon along with other minerals, generally for export purposes, have been taking place in Jos, central Nigeria, for several decades. Over the years, there has been increasing concern about the radiological impact resulting from these activities. A previous assessment in the vicinity of processing areas reported an annual dose of 50 mSv which by far exceeds the 1 mSv dose limit for members of the public. In the study, the activity concentrations of ^{238}U and ^{232}Th in 28 samples of zircon from Jos were determined using neutron activation analysis. The results gave concentrations of 466 $\mu\text{g/g}$ (5.75 Bq/g) for ^{238}U and 7054 $\mu\text{g/g}$ (28.62 Bq/g) for ^{232}Th . These results complement the results of earlier assessments and may assist in the proper management of these activities and the choice and implementation of appropriate protective measures.

1. INTRODUCTION

Zircon (ZrSiO_4) is an orthosilicate mineral, which is usually associated with other heavy minerals such as rutile, ilmenite and monazite. Because of the geological processes that led to its formation, zircon contains within its crystal lattice trace quantities of radionuclides of natural origin, particularly those in the ^{238}U and ^{232}Th decay series [1]. In the mining activities in Jos, zircon is of radiological importance, given its reportedly high activity concentrations. The zircon recovered at Jos is an altered type which contains unusually high levels of thorium in its structure. Radionuclides in the ^{238}U and ^{232}Th decay series are omnipresent in the Earth's crust but generally at low concentrations. However, some human activities, such as mining and processing of minerals, may increase

the activity concentrations of these radionuclides in ores and mine tailings as well as in the surrounding environment, resulting in enhanced exposures to natural sources. Thus recently, NORM has received global attention and has been recognized in Nigeria as a major environmental problem in the mining and petroleum industries.

For several decades, small, medium and large scale operators in Jos have been involved in commercial mining and processing of zircon and other heavy minerals. Mining and processing sites are spread over a wide area but are concentrated around Bukuru and Ropp dome. Usually, the mining process involves the removal of large amounts of topsoil (the overburden) and then the recovery and concentration of the heavy minerals by hydrogravimetric processes, which result in bringing up the deeply buried natural radioactivity to the top soil layer. Concentrate is delivered to the processing areas (milling shades) located mostly within the settlements. Zircon is typically separated from other heavy minerals, as well as from tailings mostly using the dry separation process. While the medium and large scale mechanized operators use gravimetric, magnetic and electrostatic processes, small scale local operators in their households and backyards utilize simple tools for the panning process. These activities and the indiscriminate dumping of the tailings in the affected areas apparently cause elevated concentrations of radionuclides and hence result in occupational and environmental radiological hazards of varying magnitudes. Over the years, there has been increased and renewed concern about the hazards and earlier assessments in the vicinity of milling shades reported annual radiation exposures of 50 mSv, which by far exceeds the 1 mSv dose limit for members of the public. This shows that there is need for appropriate regulatory control. To address the radiological hazard arising from these activities, intervention and remediation measures have been recommended [2].

The implementation of regulatory control measures depends on the exact knowledge of the distribution and total radionuclide concentrations in zircon by means of reliable analytical techniques. Neutron activation analysis (NAA) is a highly sensitive technique for both qualitative and quantitative analyses of major, minor and trace elements in bulk materials. It is well established for the determination of uranium and thorium in geological matrices. In this work, the radioactivity content of zircon was assessed with particular reference to the need for implementing the graded regulatory approach for the protection of workers and members of the public against exposures associated with mining industry activities in Jos.

2. METHODOLOGY

Twenty-eight representative samples of zircon ores were collected from selected small, medium and large scale processing sites located around Jos. The samples were oven dried at 110°C, and 0.15 g of each was taken, wrapped in polyethylene, heat sealed, placed in a polyethylene vial and irradiated using the Nigeria Research Reactor-1 (NIRR-1) facility at the Centre for Energy Research and Training. NIRR-1 is a low power research reactor with a nominal thermal rating of 31 kW and maximum thermal flux of 1012 n·cm²·s⁻¹. The mass concentrations (in µg/g) of ²³⁸U and ²³²Th in the samples were determined using comparative epithermal and thermal NAA, respectively. The National Institute of Standards and Technology Standard Reference Materials coals 1632d and 1633c were used for quality control, while the Origin 7.0 program was used for data analysis. The average concentrations and standard deviation were obtained. The arithmetic mean concentrations were used to calculate the activity concentrations (in Bq/g) of ²³⁸U and ²³²Th in the samples.

3. RESULTS AND DISCUSSION

The results obtained are shown in Table 1. The ²³⁸U mass concentrations were in the range of 417–473 µg/g, while the ²³²Th mass concentrations were in the range of 6811–7207 µg/g. The arithmetic means were 466 and 7054 µg/g for ²³⁸U and ²³²Th, respectively. The results indicate that the zircon has a high concentration of ²³²Th. The calculated corresponding mean activity concentrations are 5.75 and 28.62 Bq/g for ²³⁸U and ²³²Th, respectively. These values are comparable to those in some commercial zircon [1]. However, these values obviously exceed 1 Bq/g, which is the activity concentration of ²³⁸U and ²³²Th decay series radionuclides above which regulatory control is to be considered [3].

4. CONCLUSION

This study has established that the ²³⁸U and ²³²Th activity concentrations in the sampled zircon are significant enough to warrant concern and indicative of the need to apply regulatory control over activities related to the mining of zircon and associated minerals in Jos. The result may assist both regulators and other concerned parties such as operators, in implementing the recommended graded regulatory approach for the protection of workers and members of the public against exposures in the affected area.

TABLE 1. ^{238}U AND ^{232}Th CONCENTRATIONS IN THE SAMPLED ZIRCON

Sample	Mass concentration ($\mu\text{g/g}$)		Activity concentration (Bq/g)	
	U-238	Th-232	U-238	Th-232
1	472 ± 35	7100 ± 105	5.84 ± 0.4	28.81 ± 0.4
2	469 ± 43	6964 ± 179	5.80 ± 0.5	28.25 ± 0.7
3	471 ± 50	7007 ± 201	5.82 ± 0.6	28.43 ± 0.8
4	469 ± 12	7080 ± 233	5.79 ± 0.1	28.73 ± 0.9
5	468 ± 34	7206 ± 256	5.79 ± 0.4	29.24 ± 1.0
6	472 ± 28	7207 ± 179	5.83 ± 0.3	29.24 ± 0.7
7	469 ± 37	7098 ± 209	5.80 ± 0.5	28.80 ± 0.8
8	471 ± 60	7076 ± 305	5.82 ± 0.7	28.71 ± 1.2
9	467 ± 10	7078 ± 278	5.77 ± 0.1	28.72 ± 1.1
10	469 ± 19	7197 ± 215	5.80 ± 0.2	29.20 ± 0.8
11	417 ± 43	7200 ± 235	5.15 ± 0.5	29.21 ± 0.9
12	473 ± 33	6923 ± 189	5.84 ± 0.4	28.09 ± 0.8
13	469 ± 56	6897 ± 209	5.79 ± 0.7	27.98 ± 0.8
14	473 ± 53	7178 ± 219	5.84 ± 0.7	29.12 ± 0.9
15	455 ± 41	6923 ± 177	5.62 ± 0.5	28.09 ± 0.7
16	472 ± 29	6996 ± 227	5.83 ± 0.3	28.38 ± 0.9
17	471 ± 35	6891 ± 275	5.81 ± 0.4	27.96 ± 1.1
18	467 ± 22	6923 ± 301	5.77 ± 0.3	28.09 ± 1.2
19	468 ± 53	7097 ± 223	5.79 ± 0.7	28.79 ± 0.9

RADIOACTIVITY IN ZIRCON FROM JOS, CENTRAL NIGERIA

TABLE 1. ^{238}U AND ^{232}Th CONCENTRATIONS IN THE SAMPLED ZIRCON (cont.)

Sample	Mass concentration ($\mu\text{g/g}$)		Activity concentration (Bq/g)	
	U-238	Th-232	U-238	Th-232
20	473 \pm 44	6811 \pm 317	5.85 \pm 0.5	27.63 \pm 1.3
21	467 \pm 30	7118 \pm 119	5.77 \pm 0.4	28.88 \pm 0.5
22	473 \pm 11	6993 \pm 268	5.84 \pm 0.1	28.37 \pm 1.0
23	471 \pm 21	7011 \pm 197	5.81 \pm 0.1	28.45 \pm 0.8
24	467 \pm 27	6945 \pm 189	5.77 \pm 0.2	28.18 \pm 0.8
25	470 \pm 32	7196 \pm 203	5.81 \pm 0.3	29.20 \pm 0.8
26	433 \pm 58	7139 \pm 100	5.35 \pm 0.7	28.96 \pm 0.4
27	470 \pm 40	7105 \pm 225	5.81 \pm 0.5	28.83 \pm 0.9
28	449 \pm 37	7155 \pm 205	5.55 \pm 0.5	29.03 \pm 0.8

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TEMPORAL VARIATIONS IN RADON AND METEOROLOGICAL PARAMETERS IN A MASONRY DWELLING IN ABEOKUTA, NIGERIA

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Abstract

Continuous hourly monitoring of radon activity concentrations was carried out in a masonry house in Abeokuta, Nigeria. Besides the radon activity concentrations, air temperature, relative humidity and barometric pressure were also recorded simultaneously on an hourly basis throughout the seven month exposure period, spanning June to December 2012. The average diurnal variation shows that ^{222}Rn concentrations were high during the night and early morning hours and low during afternoon hours. The monthly averages of ^{222}Rn concentration were in the range 19.9–53.5 Bq/m³, with an overall average of 32.3 Bq/m³. The indoor radon concentration correlates positively with the relative humidity (+0.4836) and negatively with both air temperature (–0.3757) and barometric pressure (–0.3463), while the daytime radon concentration and air temperature correlate positively (+0.5951).

1. INTRODUCTION

Radon and its short lived decay products are responsible for about 55% of the total background radiation exposure of the general public [1]. Following its production in the soil, radon is normally transported by diffusion or pressure

driven convective means. Radon is relatively low in outdoor air, but in indoor environments it can accumulate to concentrations which can pose significant radiological risks to the occupants. Indoor radon concentrations are influenced by several parameters, including: exhalation rates from the surfaces of the walls, roof and floor; the concentrations of ^{226}Ra in the underlying soil and building materials and their porosities; ventilation rates; and atmospheric parameters. Variability in these parameters may result in large variations in the indoor radon values. Consequently, the indoor radon concentration at a particular location may vary markedly over time.

Available data on radon in Nigeria is very limited and therefore may not be representative of the true situation. Hence, there is still a need for local, regional or national radon surveys. In an effort towards satisfying these needs, a local indoor radon measurement campaign was launched in Abeokuta in the south-western part of Nigeria. Two independent radon measurement procedures were used, comprising long term measurements (up to three months) with CR-39 alpha track detectors and continuous measurements (recorded hourly) with a solid state silicon detector in a diffusion chamber. A preliminary report of the continuous radon measurement procedure is presented in this paper.

2. MATERIALS AND METHODS

2.1. Location of radon measurements

The location of Abeokuta (7.02° N and 20.33° E) is in the south-west of Nigeria. It is a town with many rock outcrops. The monitored dwelling is a bungalow of four bedrooms and one sitting room. Only one bedroom was selected for continuous monitoring over the seven month period reported here. The room is about $3\text{ m} \times 3\text{ m} \times 4\text{ m}$ and has three large sliding glass windows for ventilation. The walls are made from cement blocks and the floor is made of polished granite stones.

2.2. Continuous radon measurements

Radon concentrations in the bedroom were monitored continuously using a Sarad Radon Scout Plus instrument that employs a solid state silicon detector to detect the alpha particles emitted by radon and its decay products following the passive diffusion of radon gas from the indoor air into the device's diffusion chamber.

3. RESULTS AND DISCUSSION

Indoor radon concentrations were measured continuously and recorded hourly from June to December 2012. Besides the radon concentration, the air temperature, relative humidity and barometric pressure were also recorded simultaneously on an hourly basis.

3.1. General distribution of radon concentrations

Altogether 3024 data points were recorded over the seven month period. The distribution is presented in Fig. 1. A minimum value of zero suggests that the value is below the detection limit of the device for an accumulation period of one hour. The maximum value recorded was 173 Bq/m³, therefore all the values encountered within the period are below the international reference level of 200 Bq/m³.

3.2. Diurnal variations

The average diurnal variation shows that radon concentrations are higher during the night and early morning than later during the day (see Fig. 2). Similar observations have been reported elsewhere (e.g. in Beijing, China) [2].

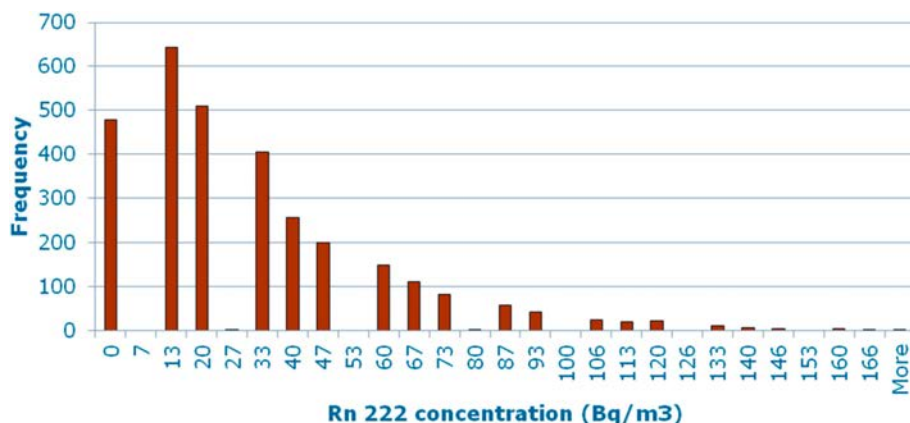


FIG. 1. Distribution of hourly radon concentrations from June to December 2012.

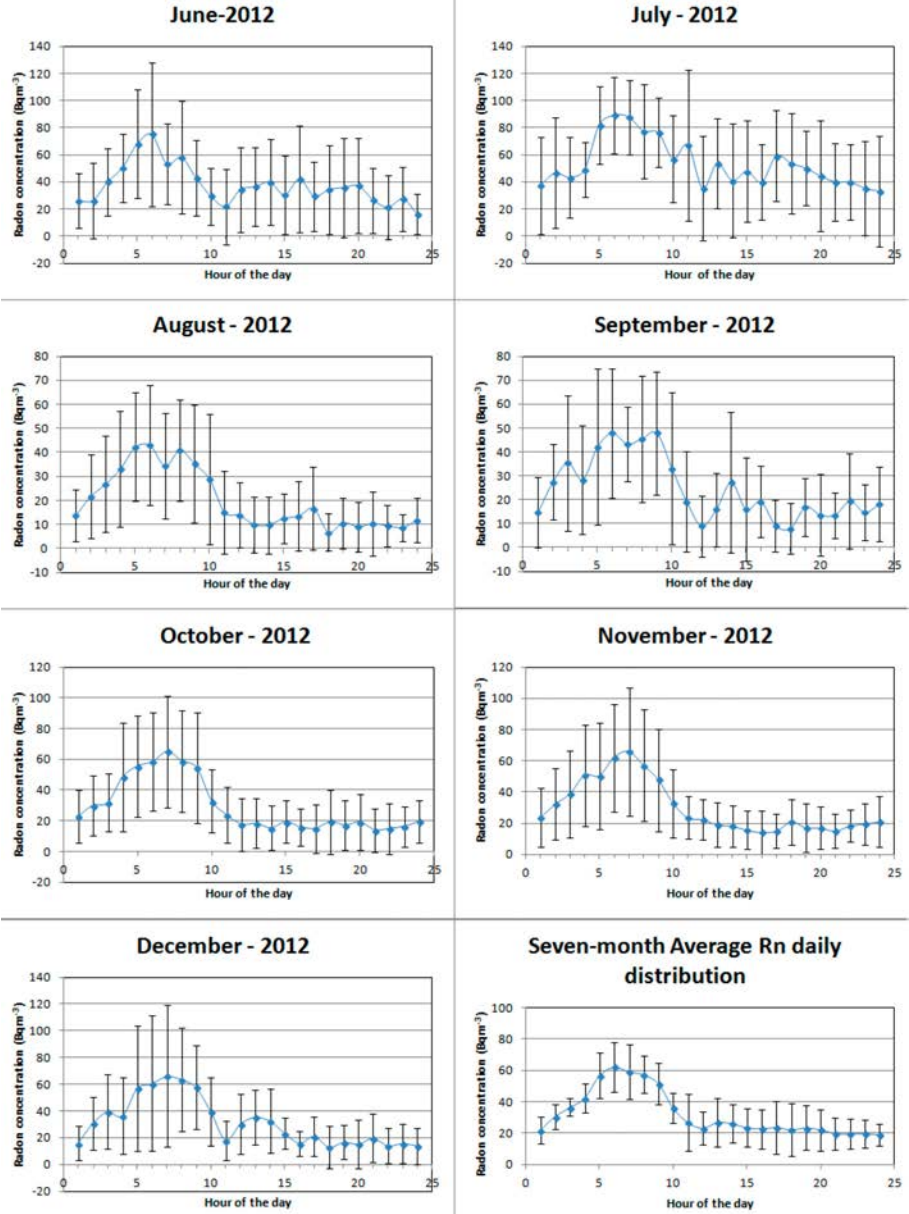


FIG. 2. Temporal variations in indoor radon concentration, by month.

3.3. Correlations between radon concentration and meteorological parameters

Simultaneous monitoring of radon concentration and meteorological parameters (temperature, relative humidity and barometric pressure) over time gives a visual impression of the relationships between these four parameters (see Fig. 3). It shows that radon correlates positively with relative humidity, but negatively with air temperature. The relationships between the barometric pressure and the other parameters are not clearly discernible from the plot.

A statistical test was performed to quantify the correlation between the radon concentration and the meteorological parameters. As shown in Table 1, the test revealed that the 24 h radon concentration (i.e. day and night) correlates positively with the relative humidity but correlates negatively with air temperature and barometric pressure. However, when only the daytime radon concentrations and meteorological parameters were considered, the correlation between radon concentration and air temperature was positive.

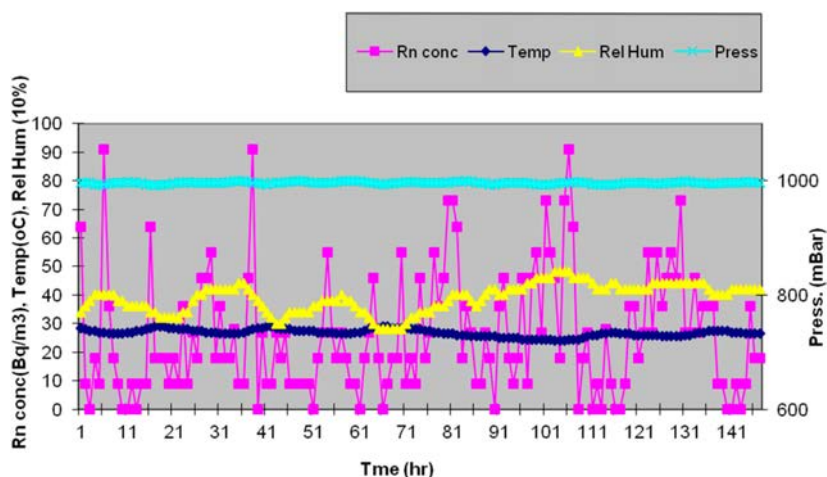


FIG. 3. Correlation between radon concentration and meteorological parameters.

TABLE 1. CORRELATION BETWEEN RADON CONCENTRATION AND METEOROLOGICAL PARAMETERS

	Correlation coefficient			
	Rn concentration	Temp.	Relative humidity	Barometric pressure
Correlation with 24 h Rn concentration				
Rn concentration	1			
Temp.	-0.375 695 031	1		
Relative humidity	+0.483 579 651	-0.838 840 202	1	
Barometric pressure	-0.346 334 346	-0.051 685 211	-0.295 719 083	1
Correlation with daytime Rn concentration				
Rn concentration	1			
Temp.	+0.595 132 419	1		
Relative humidity	+0.313 031 221	+0.039 073 233	1	
Barometric pressure	-0.280 421 204	-0.571 862 769	-0.223 445 27	1

4. CONCLUSIONS

The following conclusions were drawn from the preliminary results presented above:

- (a) The average diurnal variation shows that ²²²Rn concentrations were higher during the night and early morning hours than during the afternoon hours.
- (b) The measured radon concentrations were in the range of 9–173 Bq/m³.
- (c) The monthly average ²²²Rn concentrations were in the range of 19.9–53.5 Bq/m³, with an overall average of 32.3 ± 10.9 Bq/m³.
- (d) The indoor radon concentration correlates positively with the relative humidity (+0.4836) and negatively with both air temperature (-0.3757) and barometric pressure (-0.3463), but the daytime radon concentration and air temperature correlate positively (+0.5951).

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CONCLUDING SESSION

Chairpersons

R. GARCÍA-TENORIO

Spain

S.L. LIU

China

REVIEW OF THE SYMPOSIUM AND OVERALL CONCLUSIONS

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1. INTRODUCTION

As the final rapporteur, I was asked to give an overview of the whole symposium rather than a straight summary of the final sessions. In particular, I was asked to try and take a step back from the individual presentations and consider:

- The progress made since NORM VI (Marrakesh, 2010);
- The overall themes and conclusions from this symposium, NORM VII;
- Issues that might be relevant for NORM VIII (Brazil, 2016).

First I would like to thank the organizing committees, the symposium sponsors, the IAEA and the local hosts, all of whom helped to ensure a stimulating and successful event. Second, it is important to acknowledge the importance of holding this symposium in China, which had already been identified at previous symposia as a major international source, processor, user and supplier of industrial minerals involving exposure to natural sources. It soon became apparent that exposure to NORM and other natural sources is regarded as “an urgent concern” in China, for example, because:

- (a) A survey has identified more than 1000 mining and mineral processing sites in China that may require regulatory control.
- (b) Millions of workers in the mining and mineral processing industry are estimated to receive doses above 1 mSv/a and a significant fraction of these may be receiving doses more than 20 mSv/a, mostly from radon.
- (c) An estimated 100 million citizens live in traditional cave dwellings with elevated radon concentrations.

The symposium attracted over 150 participants from over 40 countries, and (with some exceptions) covered the global spread of NORM industries. Despite this, there were very few participants from the industries themselves — as in previous NORM symposia, most participants were from either the regulatory bodies or research institutes. It was agreed that this needs to be proactively addressed when organizing NORM VIII.

2. ISSUES FROM NORM VI (MARRAKESH, 2010)

At NORM VI, the question was asked whether we have produced a coherent and understandable system of protection for exposure to NORM. It was concluded that a number of issues needed to be resolved before making any such claims, for example:

- (a) The application, to NORM, of the International Commission on Radiological Protection (ICRP) concepts of planned exposure situations and existing exposure situations had been subject to differences in interpretation by the IAEA (in the revision of IAEA Safety Series No. 115, International Basic Safety Standards for Protection Against Ionizing Radiation and for the Safety of Radiation Sources) and by the European Commission (in the revision of European Council Directive 96/29/Euratom), and further clarification and harmonization was needed.
- (b) Derived reference levels (e.g. in the form of activity concentration criteria) are very important, especially for international NORM industries, but were often misinterpreted as limits (which they are not), or else applied to every possible use of NORM (which was never the intention).
- (c) The general concept of dose reference levels for existing exposure situations was generally understood, but their use — including the choice of values of annual effective dose to use in practice — was not.
- (d) There is a reliance on models to estimate the exposure of workers and the public. Such models are often extremely pessimistic and produce poor decision making in terms of worker and public protection and (especially) in terms of evaluating options for managing residues from NORM industries.
- (e) It was concluded that the scope of application of the IAEA Regulations for the Safe Transport of Radioactive Material to NORM was appropriate. Despite this (or even because of this), the installation of radiation monitoring equipment at national borders had resulted in an increase in the number of denials of shipment of NORM consignments.

From the above, the following issues were identified as key requirements to be brought forward to be resolved (or at least discussed and progressed) at this present symposium:

- (1) More clarity on the assignment of the most appropriate ICRP exposure situation for NORM, and more consistency between the IAEA International Basic Safety Standards and the European Directive;
- (2) Practical guidance on how to apply the ‘graded approach’ to planned exposure situations and existing exposure situations;

REVIEW OF THE SYMPOSIUM AND OVERALL CONCLUSIONS

- (3) Encouraging an industry based and industry led approach to the control of exposure to NORM;
- (4) A need to validate exposure models through confirmatory measurements and sensitivity analyses;
- (5) More practical examples of the application of the optimization principle to public and occupational exposures to NORM;
- (6) More options for, and examples of, the use and recycling of NORM residues;
- (7) More facilities for the disposal of NORM waste, in situations where this is the best residue management option;
- (8) More stakeholder involvement and more sharing of knowledge on best practices.

3. THEMES FROM THIS NORM VII SYMPOSIUM

The symposium contained oral presentations (invited and submitted) and a wide selection of poster presentations. Certain themes emerged, and I have attempted to summarize these in the following sections.

3.1. The system of protection

To understand the protection philosophy for exposure to natural sources it is necessary to understand the ICRP recommendations as they apply to the different types of exposure situation, specifically planned exposure situations and existing exposure situations. Details were given in some of the papers presented at this symposium. In essence, exposure to natural sources is by default regarded as an existing exposure situation. In certain circumstances, however, it should ideally be managed using the protective measures for planned exposure situations. The optimization of protection is required in either case and there is an emerging consensus that the question of planned exposure situation versus existing exposure situation is not as important as we might have thought. Instead, it may be more useful to move on and consider the mechanisms by which control may be exercised. Nevertheless, it is clear that this remains a source of confusion, even amongst experts in the field of natural sources, and further clarification from the ICRP through its planned task group on NORM would still be useful. It would also be useful to clarify how the concepts of (a) exclusion of exposures from regulatory instruments and (b) exemption of practices from the requirements for planned exposure situations, both of which are familiar to NORM industries, fit within the all-encompassing system of exposure situations that apply to NORM.

Deciding on the most appropriate type of exposure situation is, of course, only the first step, and the symposium highlighted a number of emerging issues with respect to implementing a system of control. For example, there is substantial

agreement on the numerical value (1 Bq/g) of the activity concentration criterion for determining whether exposure to a mineral or raw material is subject to the requirements for planned exposure situations (in which case, the practice giving rise to the exposure would require authorization (or exemption) by the regulatory body and the material would fall within the definition of NORM)¹. Despite such agreement, however, there are very diverse (and often incorrect) interpretations of what this 1 Bq/g criterion represents. The most serious misinterpretation is to consider the criterion as a limit — there were in fact examples of such a misinterpretation here in this symposium. It is also wrong to regard this criterion as a universal trigger for applying a comprehensive system of regulatory control. Such a system is to be applied using a graded approach, based on the expected level of exposure of workers and the public. It was clear from this symposium that 1 Bq/g does not automatically imply exposures of the order of 1 mSv/a — in most if not all cases, doses are very much lower. Thus, more needs to be done to (re)explain the basic concepts and a more graded structure needs to be built into regulatory systems (if necessary using different derived activity concentrations) to ensure that the degree of regulatory control is commensurate with the risks involved.

3.2. Optimization

Although the optimization principle applies to all types of exposure situation, there were relatively few examples of its practical implementation with respect to NORM. To some extent, this may be because the focus has remained on establishing the protection framework rather than on its implementation. There do remain some fundamental questions; for example:

- (a) What is an appropriate dose reference level for the remediation of contaminated sites? The ICRP recommends dose reference levels in the range of 1–20 mSv/a but, in practice, the choice is always at the lower end of this range and in some cases is even below 1 mSv/a. Furthermore, evidence from (limited) stakeholder engagement suggests that residual doses above 1 mSv/a may be regarded as unacceptable by affected communities.
- (b) The ICRP recommends that optimization should be applied below the dose reference levels, but what actual scope is there to further restrict doses below 1 mSv/a?

¹ The criterion of 1 Bq/g applies to individual radionuclides in the ²³⁸U and ²³²Th decay series.

One area where there is great scope for optimization is exposure to radon and perhaps (as a number of papers suggested) even to thoron. To date, much of the effort has been devoted to ensuring that reliable estimates of annual dose are obtained. Consequently, concerns were expressed about the proposed doubling of the risk estimate for radon and the overall reliability of the dose coefficients (i.e. risk coefficients) for both radon and thoron. Of major concern was what to do about historical dose estimates and how to communicate any such major changes in risk estimates to workers and the public.

In addition to having a knowledge of the annual dose, it is also necessary to know, for optimization purposes, “where, when and how” such exposures are received, in order that appropriate protection options can be identified. There is now a large body of scientific literature on radon remediation strategies, although further work may be needed to consider the options for protecting against radon exhaled from building materials, especially in view of the apparent increase in the use of NORM residues in such materials. In terms of NORM industries, more examples of successful ALARA (as low as reasonably achievable) programmes are needed, and best practice needs to be shared among the international NORM community, for example through networks.

3.3. Models and measurements

Although there is an increasing use of measurements to estimate individual doses (especially for workers), there is still a reliance on exposure modelling and the previous questions about the overall reliability and utility of the results of these models remain. Except for radon, high exposures are found only in a relatively few cases — in such cases workplace and environmental measurements are to be used as the basis for determining the doses received. In other cases, where doses are almost certainly low, realistic rather than pessimistic models are required.

This symposium featured several presentations and posters on NORM measurements, describing the equipment and techniques and the results obtained. There have been several recent developments in survey instrumentation, which can usefully assist in making rapid and reliable measurements on (often quite large) sites associated with NORM. Similarly, advances in radon measuring equipment can provide important input to the optimization process, such as information on the spatial and temporal variation of radon concentrations and information on task specific doses.

Models and measurements both have their limitations, and it is suggested that the optimum solution is to bring the two disciplines closer together to work in a complementary manner. Thus, models can be used to determine what types of measurements should ideally be undertaken to produce reliable indicators

of exposure (to persons and, where appropriate, non-human species). In turn, measurement results can be used to validate and improve models.

3.4. Management of NORM residues

This has always been one of the biggest issues at these symposia, and this remains the case. NORM residues range from low to medium quantities of highly active scale to vast quantities of minimally active residues such as phosphogypsum and fly ash:

- (a) For the former, there may be limited scope for recycling (or at least materials recovery), but in many cases a final disposal solution is required. We are gradually seeing new disposal facilities becoming available, which are suitable for (or even specifically designed for) NORM. This is a welcome development, but clearly more of these are still required.
- (b) For the latter, the low radiation risk and the huge physical quantities require us to think of such residues as a potential resource (i.e. for recycling or use as by-products). While this idea is gradually gaining momentum, and there are good examples of its practical implementation, it is still far from being implemented on a large scale. One reason is the concern about doses to future generations from, for example, incorporating NORM into building materials. It is necessary to proceed with caution, since such practices may ultimately contribute the largest proportion of the collective dose in the life cycle of NORM.

It is suggested that the future focus needs to be on the question of optimization, considering the full NORM life cycle and engaging more fully with concerned parties. It is easy to forget that the radioactivity in NORM is neither created nor destroyed — it is simply moved from one place to another — and all options (including doing nothing) involve some radiation exposure.

One area where there was clear evidence of progress was in the decommissioning and remediation of NORM industrial sites. There were good examples from a number of different countries of remediation projects in which the radiation protection and waste management issues were addressed and important lessons learned. There are still many sites that will need to go through a similar process, and the sharing of this information is extremely valuable. Consequently, the compilation of a NORM site remediation library or database would be very useful.

3.5. Other issues arising

NORM is a multidisciplinary field and, as always, space does not permit a summary of all the issues that arose during the course of this symposium. However, to give a flavour of the proceedings, the following examples of such issues are given:

- (a) Coal fired power stations: Most coal exhibits relatively low activity concentrations and estimated doses are correspondingly low (although collective doses may still be significant due to the large populations involved). However, it is clear that in some cases NORM concentrations, and thus radiation exposures, can be significant.
- (b) Knowledge and awareness among NORM workers: It was concluded that more needs to be done to train and inform NORM workers so as to improve their perceptions of radiological risk and also to understand why (in some cases) radiation protection controls are necessary. Also, more needs to be done to engage with workers as a key stakeholder group.
- (c) Transport of NORM: As in previous NORM symposia, the general consensus was that the IAEA Regulations for the Safe Transport of Radioactive Material, including especially the ten times ‘exclusion factor’ for NORM, are appropriate and work well in practice. However, NORM consignments continue to have problems at borders, usually because exempt material still triggers radiation detection systems. Such systems are increasingly sophisticated and allow the presence of NORM to be specifically identified. To help facilitate transport it might be useful to develop NORM ‘catalogues’ for persons operating monitoring equipment and for consignors to provide a brief information package (a ‘NORM passport’) which can accompany consignments.

4. CLOSING THOUGHTS — ISSUES FOR NORM VIII

In terms of the priorities from NORM VI listed at the start of this summary, it can be seen that there has been progress, and in some cases this has been significant. Even so, it is in fact possible to take the same list of priorities forward to NORM VIII.

Being realistic, issues such as NORM residue management cannot be expected to be completely resolved in just a few years. We can, however, expect progress in terms of preparing the way for such options to be implemented. To do this, further clarification on the system of protection for NORM and its implementation in practice is needed from international organizations. At the same time, industries themselves need to think about how they can make

convincing optimization cases for the full NORM life cycle. This in turn needs to be supported by radiation protection experts, for example by producing reliable data on exposures to underpin the decision making process.

The issue of stakeholder engagement also needs to be taken forward. Although now a common feature of the radiation protection landscape, and very relevant to many NORM issues, very few examples of it being undertaken were presented at this symposium. Experience suggests that this can be a slow and lengthy process and needs to be started now if we want to use NORM VIII in 2016 as a forum to discuss progress.

Finally, I would like to again say how valuable this symposium was and to pass my compliments to the organizers, sponsors and all those who participated to the event.

CLOSING ADDRESS

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Distinguished Mr Chair, dear friends, ladies and gentlemen, good morning.

The Seventh International Symposium on Naturally Occurring Radioactive Material is coming to an end after all the reports and discussions in these last few days. During this conference, Chinese and international experts delivered lectures and reports about NORM in industries and the environmental aspects, current status and challenges of NORM management; and they jointly discussed with all the participants topics regarding the development of NORM measurement methods and strategies, safety assessment, radiation protection of NORM, the management of NORM residues, as well as NORM related disposal and transport. Many very constructive ideas and recommendations have been put forward during the lectures and discussions, which brought us rich contents and results. Here, on behalf of the national organizing committee of the conference, I would like to thank everyone for all your active participation and hard work.

This is the first NORM conference held in Asia, which has important significance for China as the host country. Currently, systematic technical research and its application in relation to NORM is in the initial stages in China. Many problems occur during industrial processes, such as obsolete technology, improper disposal, incomplete standards and imperfect regulation; similar situations happen when recognizing and disposing of NORM in terms of its environmental impact. Therefore, there is a certain gap between China and the international counterparts. This conference has provided a very special platform; here we could fully acquire knowledge on the current status of studies and management of NORM from the international side, we could have in-depth exchange of ideas with top experts focusing on NORM from the IAEA and other countries, we could get the most updated study results and development information, and we could learn about advanced ideas, technologies, standards and good practices. The documents and ideas resulting from this conference could be helpful to further increase research and the level of regulation of NORM in China and worldwide.

The Nuclear and Radiation Safety Center (NSC) is affiliated to the Ministry of Environmental Protection and the National Nuclear Safety Administration and is a non-profit technical support organization in the field of nuclear and radiation

safety regulation. Its main mission is to provide overall and comprehensive technical support and assurance to the safety regulation and environmental administration for China's civilian nuclear facilities. After the Fukushima nuclear accident, the international society and public put more focus and sensitivity on nuclear safety and the radiation environment; simultaneously, requirements and expectations for the nuclear safety regulation institutions and technical organizations are increasing.

To adapt to the requirements of nuclear power construction and safety assurance, the NSC made great developments in the areas of institutional functions, organizational scale, human resource building and technical capability in recent years. Now we have 484 official staff working in 4 managing departments and 17 operating divisions. We have six main functions including: providing technical support on nuclear and radiation safety review and regulation; making reaction to and assessment of nuclear accidents; studying nuclear supervision policy, law and regulations; making scientific research on nuclear and radiation safety; providing technical consultation; as well as providing an information service. The work fields of the NSC refer to technical review and assessment, licence application, qualification management of operators, as well as fundamental studies in the stages of siting, construction, commissioning, operation and decommissioning of nuclear facilities, which could provide overall technical support and guarantee the safety of China's civilian nuclear facilities, nuclear technical applications as well as nuclear and radiation safety regulation work.

To keep up with the NSC's rapid development requirement that I have already mentioned, the NSC is positively promoting a project to build a national research and development base of nuclear and radiation safety regulation. This project has been approved in the government document of the 12th Five Year Plan and Vision 2020 of Nuclear Safety and Radioactive Pollution Prevention and Control. The Chinese Government is to provide special financial support and approve a certain construction area of about 16 000 m². The research and development base will consist of three major sectors in charge of, respectively, international exchange and training, research and testing, and office management. It is planned to build six major scientific research laboratories and four public support facilities. When the research and development base is completely constructed and able to provide a service, the NSC will become a more comprehensive organization with the combined functions of technical review and assessment, technical research and development, information exchange, international cooperation, as well as human resource training.

As a result of this new, historic development, the NSC will continuously focus on updated development information on nuclear and radiation safety technology from domestic and international society and it will significantly expand technical research on nuclear and radiation safety. It will place more

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emphasis on NORM radiation monitoring, radiation protection, safety regulation, standards making and environmental impact assessment during the process of industrial and agricultural production, as well as environmental monitoring. In the meantime, we are expecting to carry out sustained and extensive technical exchange and cooperation with our international counterparts in the fields of NORM and other areas related to nuclear and radiation safety.

Again, as chair of the National Organizing Committee, I would like to extend my congratulations on the complete success of this conference and also to express my sincere gratitude to our colleagues from the IAEA involved in the organization of the symposium, all the experts' participation, and the warm service from all the conference staff.

Thank you!

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INTERNATIONAL ATOMIC ENERGY AGENCY
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ISBN 978-92-0-104014-5
ISSN 0074-1884