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Studies on materials containing natural radionuclides as part of the European Community strategy on radioactive waste management

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STUDIES ON MATERIALS CONTAINING NATURAL RADIONUCLIDES AS PART OF THE EUROPEAN COMMUNITY STRATEGY ON RADIOACTIVE WASTE MANAGEMENT

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BACKGROUND

The main contributions to human exposure to ionising radiation arise from natural sources - cosmic rays, the radionuclides in the earth's crust and the natural radioactivity of the human body. By comparison, man-made sources are currently responsible for only about 10%, on average, of human exposure. The science of radiological protection, and national and international regulatory regimes, have evolved to ensure safety in the use of these man-made sources of exposure. Less attention has been given to exposures that can occur as a result of the use of naturally radioactive materials. The levels of naturally occurring radioactivity in the formations that make up the earth's crust and in environmental materials vary widely and there are some materials that are extracted for industrial uses that contain radioactivity at concentrations that cannot be disregarded. In some cases, industrial processing can lead to further enhancement of the concentrations either in the product or in waste materials.

Because of possible concerns over the radiation exposure that could result from the handling of such materials and from the use or disposal of the wastes, the European Commission has supported a programme of work in this area over a number of years and has assembled a substantial body of information by means of contracts and contributions from experts.

First studies on waste in the phosphate industry had been commissioned in 1988 in the frame of the research and development program on radioactive waste management and disposal. The Fifth Action Programme on the Environment, approved by the European Council in February 1993 announced a "Community strategy for radioactive waste management". This strategy was published by the Commission in March 1994 [1], and it was specified that "it takes into account all sectors involved: not only the energy sector, which concerns several Member States, but also industrial activities generating waste containing enhanced quantities of natural radionuclides, and the user of radioisotopes in agriculture, medicine, research and industry, which concerns all Member States".

With guidance from national experts acting in a working group, areas of interest for studies in the field of natural radionuclides had been defined and commissioned. The results of individual studies and input from the experts have been collected and published in a report [2], which is drafted to be readable for non-specialists.
This paper summarises the main results for individual material streams and indicates sources for more in-depth information. Materials with natural radionuclides appearing in the nuclear fuel cycle, like uranium mining- and milling residues and depleted uranium are not considered here.

NATURAL RADIONUCLIDES AND SYSTEMS OF RADIOLOGICAL PROTECTION

Of the natural radionuclides in the earth's crust, those which are found to be the main sources of human radiation exposure are ^{40}K (potassium-40), ^{232}Th (thorium-232), ^{235}U (uranium-235) and ^{238}U (uranium-238). Potassium is a common element, the radioactive isotope ^{40}K constituting 0.012 % in its natural form.

The three heavy nuclides (^{232}Th, ^{235}U and ^{238}U) decay to produce other nuclides of other elements, which, in turn decay further through a chain which includes several elements, eventually to end in stable

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isotopes of lead. Significant daughter nuclides in these chains include radium-226 (in the uranium-238 decay series) which is soluble in water. The decay of ^{226}Ra results in the production of ^{222}Rn (radon-222), an inert gas that does not form chemical bonds well and can usually escape via gaseous pathways. ^{210}Pb and ^{210}Po (^{238}U daughters) can occur in compounds which are volatile at high temperatures (several hundred degrees Celsius) and can, therefore, possibly escape by airborne routes. Following release and cooling they can adhere to respirable aerosols, thus depositing and contaminating local surfaces and lungs.

The regulations applying to practices involving exposure to materials containing high concentrations of natural radioactivity show considerable variation across the member states of the European Union and are, in general, less restrictive than those applicable to man-made radionuclides. However, the subject has received more attention in recent years resulting in a more consistent approach to dealing with the avoidable risks posed by radiation from all sources, as stated in Publication 60 of the International Commission on Radiological Protection [3].
The presently valid European Union basic safety standards [4] provide reporting levels above which the material or waste has to be included in a system of registering, licensing and control; the specific activity threshold is 500Bq/g for natural radionuclides.
The Council of the European Union approved a Directive on 13 May 1996 setting out revised basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionising radiation [5]. This could affect industrial practices, although specific action is left to the national safety authorities.

The system of radiological protection for workers and the public recognises that practices that give rise to acceptably low levels of exposure may be exempted from regulatory control. Recommended radiological criteria for exemption from reporting have been incorporated in the basic safety standards Directive. Default values, in terms of total activity and activity concentration, have been derived on the basis of the above mentioned criteria. These are listed in Annex 1 of the Directive. Thus a practice may be exempted from the requirement for reporting if it involves only materials that do not exceed in total one or other of the exemption values. Member States may, in exceptional circumstances, define different values from those listed in the Directive, but on the basis of the same general criteria.

In a contract with KEMA [6], regulations and trend in legislation in the Member States have been reviewed, and problems arising in some branches of industry have been identified.

PROCESSES LEADING TO ENHANCED CONCENTRATIONS

The following sections give a very brief summary of the processes leading to production of such waste, an indication of the radioactivity content of the waste, and some approximate quantities where available.

Power production from fossil fuels

The combined concentrations of ^{238}U and ^{232}Th in coal are generally in the range 30 to 50 Bq/kg but higher levels have been recorded in coal from particular sources. In the ash from combustion of coal, radioactive concentrations are enhanced by a factor of about 10 for most radionuclides but by about two orders of magnitude for ^{210}Po and ^{210}Pb. The total concentrations in the fly-ash are of a similar order to the reporting levels in the revised basic safety standards Directive of 1996. Probably the highest concentrations (and exposures) are related to the coal-mining and combustion at Freital in Eastern Germany; for some time, coal was even mined in order to extract uranium [7].
Various assessments of the radiological impact of the management of coal ash have been undertaken and these show that the impact on workers and also on the public is low. Similarly, the radiological impact of emissions of radioactivity from power station stacks has been shown to be very low.

On the basis of limited data, the impacts of other fossil fuels such as oil, gas and peat are likely to be less than for coal.

Phosphate ore processing and use

The concentrations of natural radioactivity in phosphate ore vary from about 100 to about 5000 Bq/kg and are dominated by the contribution of the ^{238}U series. Chemical processing results in the uranium being enriched in fertiliser to about 150% of the initial ore concentration, whilst most of the radium is left in the phosphogypsum waste. The overall position is that, in typical cases, concentrations in the ore, in the phosphate product and in the waste materials are generally around the reporting levels of the 1996 Directive.

One area of concern is the re-use of phosphogypsum waste in building materials which could lead to problems of effective regulatory control.

Two studies ([8] and [9]) have been devoted to assessing all aspects of phosphate ore processing and the management of associated materials.

Oil and gas production

Oil and gas production give rise to scales and sludges containing naturally occurring radioactive materials at levels of the order of 10^5 Bq/kg which are well above the reporting levels of the 1996 Directive. Total arisings of these waste materials in the European Union amount to a few thousand cubic metres per year.

Assessments of the radiological impact of the scales and sludges generally indicate doses to individual workers of around 1 mSv/a. These exposures do not exceed dose limits and for off-shore workers are more than offset by lower ambient levels away from land. The radiological impact on the public of disposal of scale material is not well documented. The eventual disposal of off-shore structures is an issue that requires further attention.

The current practice of dealing with material from oil and gas production in the European Union has been assessed [10].

Rare earths and zirconium products

Concentrations of naturally occurring radioactivity in rare earths and zirconium ores and in products and wastes are generally around 10 000 Bq/kg. Occupational exposures during the processing of these materials have been estimated, using conservative estimates, to be in the range of a few mSv/a, mainly from internal exposure.

The dose to the public due to liquid and airborne effluents from the processes have been shown to be very low. More significant are the estimates of dose resulting from the disposal to landfill of waste materials. Individual doses resulting from subsequent developments of a landfill site have been estimated to be up to 100 μSv/a. Assessments of the collective dose over the very long term from landfill disposal of the wastes range up to 10^5 man-Sv [11].

Metal smelters

Radionuclide concentrations in iron ore are generally low and even in slags and other wastes are below the reporting levels. Higher levels occur in aluminium, tin and titanium ores and concentrations in both raw materials and wastes can be around the reporting levels. Very high radionuclide concentrations occur in pyrochlore, the source of niobium and this is reflected in the products and the wastes.

Assessments of the radiological impact of operations associated with metal smelting generally indicate that worker doses are low with the exception of those involved with pyrochlore, where doses of up to a few mSv/a could be received.

Exposure of the public due to releases from the processes are generally assessed to be low. However, the potential doses from landfill disposal of waste could be more significant at up to 10 mSv/a as a result of possible intrusion and site development scenarios[/11/].

Copper mine tailings

The problem of management of copper mine tailings is restricted, within the European Union, to the Mansfeld region of the former German Democratic Republic where copper mining and smelting was undertaken from the middle-ages up to the mid-twentieth century. This has left a legacy of about 110 million cubic metres of waste in over 1000 waste piles. The concentrations of natural radioactivity in the waste range from hundreds to thousands of Bq/kg. The situation is under investigation by the German authorities with a view to developing a basis for decisions on remedial actions[/7/].

Building materials

A number of waste materials containing enhanced levels of radioactivity are used in the manufacture of building materials. Generally the most significant waste material is phosphogypsum where it is used in place of natural gypsum in various building products. Less widespread, but of radiological significance is the use of mining wastes and slags, particularly in Germany.

Assessments of occupational exposure during the manufacture of building materials using recycled waste suggest that, even on conservative assumptions, individual doses are at most a few mSv/a.

Studies have shown that the contribution to external doses to members of the public for time spent indoors or in the vicinity of buildings is typically about 0.6 mSv/a without use of materials containing enhanced levels of radioactivity. This indicates that there is significant potential for increased external exposure where recycled materials are used for building purposes. Probably of greater importance in most cases is the increased exposure to radon though data on this aspect are inconsistent.

The Chemical industry

The chemical industry is very diverse and involves a great number of different types of material of terrestrial origin. A review for the European Commission reported that a number of attempts had been made to get information from the chemical industry about the radioactivity in their waste streams. It was concluded that there is a lack of first hand knowledge about the radioactivity of chemicals and raw materials used in the chemical industry. However, it is likely that in the industry there are raw materials which contain varying levels of radioactivity.

Disused sealed radium-containing sources

Prior to 1950 radium was the only source in common use. It was used for medical purposes and research within hospitals, clinics, universities and other research laboratories. Typically medical radium needles and tubes range from a few tens of mg (tens of Mbq) to a few hundreds of mg (hundreds of Mbq) of radium. After that time, with cobalt-, cesium-, and strontium- sources becoming available, many users replaced radium sources, donated them to other hospitals or even countries, disposed of the sources in a controlled or uncontrolled manner and in some cases simply forgot about their existence.

Industry started to use radiation sources in gauges, irradiation facilities and industrial radiography. Radium continued to be used by researchers for various purposes such as instrument calibration. ^{226}Ra/Be sources were also used. These sources exceed 500mg (20GBq) and in some cases can be larger than 1000mg (40GBq).

A large variety of sources became available for medical and industrial use after 1960/70. It was during this period that some form of regulatory control or documentation of radiation sources became more common.

Non-registered sources disused sources continue to be a source of concern. In order to dispose of these sources they need to be located and retrieved by experienced personnel. In some cases the existence of the source is unknown whereas in others the location of an identified source may not be known. In many cases it will be a case of raising awareness amongst people who are likely to come into contact with spent sources whilst in others it will be necessary to encourage people to admit to sources in their possession. In the latter case, some incentives may be required to ensure that the source is safely disposed of rather than continuing to be held, possibly in an inappropriate manner.

In a study [12], risks and ways to improve the situation have been identified. An on-going follow-up study will quantify this waste-stream and propose concrete measures for management.

FURTHER ACTION BY THE EUROPEAN COMMISSION

The previously mentioned revised Directive 96/29/EURATOM on basic safety standards on radiation protection, which must be implemented in national law by the year 2000, contains reporting levels for natural radionuclides. As a result, Safety Authorities will have to prepare regulatory provisions for wastes containing enhanced levels of natural radionuclides.

The Commission services have conducted several studies to assess the origin, quantities and whereabouts of waste with enhanced concentrations of natural radionuclides, which may present a danger to health by ionising radiation to workers and public. A communication on this subject is planned for early 1998.

The Commission will:

- summarise the origin, industrial processes and arisings of wastes,
- describe the radiological consequences of the presence of such waste in different scenarios as far as available information allows,
- discuss possible regulatory means, and
- recommend an approach to regulation, licensing and control.

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