

APPROACHES FOR REGULATING MANAGEMENT OF LARGE VOLUMES OF WASTE CONTAINING NATURAL RADIONUCLIDES IN ENHANCED CONCENTRATIONS^a

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ABSTRACT

The new Basic Safety Standards (BSS) of the European Union for protection of the public against the dangers of ionising radiation contain nuclide specific exemption levels on reporting. These exemption levels are much lower for the natural radionuclides (10 or even 1 Bq/g, depending on the radionuclide) than under the old directive (500 Bq/g for natural radionuclides). As a consequence, industrial activities with certain minerals or raw materials containing elevated concentrations of radionuclides, which were exempted from the radiation protection control system under the old directive, may have to be declared as radiologically relevant activities and may come under regulatory control. Examples of these activities are the phosphate ore processing, the refining of a number of metals, the use of slag, and the oil and natural gas production. This might give rise, in some cases, to large volumes of waste which should be treated, conditioned and disposed in a controlled way.

Because of the relatively high exemption levels under the old BSS, regulators in most of the Member States of the European Union have hardly been concerned with radiation protection issues in these industries. By order of the European Commission, we have carried out a study on the implications of the new BSS for this type of industrial activities and to recommend pragmatic approaches for regulating the management of large volumes of waste generated by these industries.

The paper gives an overview of the exemption levels for natural radionuclides in the new BSS. Secondly, the industrial activities that give rise to large volumes of waste containing enhanced natural radionuclides are presented. Finally, recommendations and practical approaches are given which can be used to draw up the conditions to be applied to the safe disposal of such wastes. These conditions are, in general, not necessarily the same as used for anthropogenic radioactive waste. In many cases a burial site under some control regime may suffice. Also, a plea is given for recycling of materials, with or without separation or immobilisation of the radioactive contaminants.

INTRODUCTION

All raw materials contain traces of natural radioactivity. Radiological problems connected with the processing of uranium ore for the nuclear fuel cycle are well known. However, the processing of raw materials in various non-nuclear industries also generates waste with concentrations of radionuclides from the natural uranium and thorium decay chains that are not to be neglected. The resulting problems can be considerable as, in general, large volumes of waste are involved.

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International bodies and national authorities have issued directives and regulations for protection of workers and the general public against ionising radiation. The current national laws and regulations in the Member States of the European Union are still based on the former Council Directives [1,2] of the European Commission under the Atomic Energy Treaty of Rome (1957). However, since the exemption levels for natural radionuclides have been relatively high in these Directives, the regulators in most of the Member States haven't paid too much attention to the potential radiation protection problems in the non-nuclear industries, leading to different national interpretations on these issues between the various Member States.

The progress in knowledge of the harmful effects of ionising radiation and the wish to bring harmonisation in the regulations in the Member States has induced the European Commission to issue a revised Council Directive [3], containing the new BSS. In the BSS, specific attention has been paid to natural radiation sources and Member States are obliged to identify work activities that may lead to a significant increase in exposure, both to workers and to members of the public. The national regulations of the Member States have to comply with the BSS within a period of four years, i.e. before 13 May 2000.

The BSS contains exemption levels for reporting of radioactive materials which are no longer of a generic nature, but are specified for each single radionuclide. The exemption levels for natural radionuclides are substantially lower than the former ones. It is therefore to be foreseen that under the new BSS large volumes of waste produced by the non-nuclear industry have to be declared "radioactive".

By order of the European Commission, a study has been performed on the volumes of waste containing natural radioactivity above the exemption levels, generated in the European Union and on practical approaches for the management of such volumes [4]. For this study, information was gathered through literature and by interviews in production plants, both on radiological problems for workers and on waste problems. Only a limited number of branches of the non-nuclear industry were visited. The selection was made in the expectation that those branches will produce the bulk of waste with enhanced natural radioactivity. For each branch one company in the Netherlands and one elsewhere in Europe were visited. From the information gathered an extrapolation to the total European Union was made.

The objective of this study was to inventorize the extent of produced volumes, and to formulate approaches for the management of such volumes.

EXEMPTION LEVELS

The exemption levels are based on calculations using several scenarios for exposure of workers and members of the public to radioactive materials. The criterion for calculating the levels was a radiation dose of $10 \mu\text{Sv}\cdot\text{a}^{-1}$, which was regarded as being a trivial dose [5]. Although this is not specified in the BSS, the calculations were carried out for moderate quantities of radioactive substances only, being defined [5] as sources smaller than 1 m^3 . It is evident that, in general, industrial waste with natural radionuclides in enhanced concentrations exceeds this volume.

According to the former Directives of the European Commission materials were exempt from of interference by the relevant authorities when the specific radioactivity remains below 100 Bq.g^{-1} , or 500 Bq.g^{-1} for natural radioactivity in an undisturbed matrix. A short listing of the reporting levels as stated in the new BSS for only natural occurring radionuclides of the uranium and thorium decay chains is given in Table I. Both levels, the absolute value of radioactivity, as well as the specific activity, have to be exceeded before reporting of a material is required. The reporting level for the quantity of activity in Becquerel is in general only a factor 1,000 higher than the numerical value for the reporting level of specific activity in Becquerel per gram. It must be emphasised that in the non-nuclear industry the specific activity will determine the exemption. The exempted amount of activity will always be exceeded, as industrial stocks on hand are normally in the order of tons of material.

The BSS is vague about large volumes of waste with natural radioactivity. However, it is to be foreseen that several wastes and residues will be subject to reporting. The disposal could then involve substantial financial, practical and logistic problems.

Table I Quantities and concentrations of natural radionuclides, specified in the BSS.

The suffix 'sec' means with the decay chain in secular equilibrium,
the suffix '+' means with only short living daughters in equilibrium.

Nuclide	Quantity (Bq)	Concentration (Bq.g ⁻¹)
<u>U-238 series</u>		
U-238 sec	10 ³	1
U-238+	10 ⁴	10
Th-234+	10 ⁵	10 ³
U-234	10 ⁴	10
Th-230	10 ⁴	1
Ra-226+	10 ⁴	10
Rn-222+	10 ⁸	10
Pb-210+	10 ⁴	10
Bi-210	10 ⁶	10 ³
Po-210	10 ⁴	10
<u>Th-232 series</u>		
Th-232 sec	10 ³	1
Ra-228+	10 ⁵	10
Ac-228	10 ⁶	10
Th-228+	10 ⁴	1
Ra-224+	10 ⁵	10
Rn-220+	10 ⁷	10 ⁴
Pb-212+	10 ⁵	10
Bi-212+	10 ⁵	10
<u>U-235 series</u>		
U-235+	10 ⁴	10
Th-231	10 ⁷	10 ³
Pa-231	10 ³	1
Ra-223+	10 ⁵	10 ²

PROBLEMS IN THE INDUSTRY

The inventorization in the separate branches of the non-nuclear industry of possible problems with waste yielded the following results.

Coal

The specific activity in coal is, in general, not high and on average of the same order as the mean concentration in the earth crust. However, there is a great spread in the specific activity depending on the origin and even on the specific pit [6]. Measured specific activities are from 1 to over 500 Bq.kg⁻¹. As a rule of thumb, the specific activity in the remaining ash after burning of the coal is a factor of 10 higher. The greatest consumption of coal is for electricity production. Total consumption in the EU of coal, brown coal and peat is about 300 Mton.a⁻¹, resulting in about 30 Mton of ash [7]. Most fly-ashes will be exempted from regulatory control as the specific activity remains under the reporting level of 1 Bq.g⁻¹. However, the few batches that exceed the reporting level may make it necessary to set up a control and certification system for the radioactivity of fly-ashes. The amount of 30 Mton.a⁻¹ of fly-ash makes reuse preferable to dumping. However, the radioactive components may make reuse more difficult.

Steel

Problems with radioactive waste in large quantities are to be expected only at blast furnaces. The total annual crude steel production in the EU is about 130 million tons [8]. The content of natural radioactivity in ore is, in general, low, <50 Bq.kg⁻¹. However, the volatile ²¹⁰Po is largely released via stacks and the large carry-overs give rise to environmental contamination. Sludge produced at blast furnaces contains large amounts of ²¹⁰Pb. By radioactive ingrowth ($T_{1/2}=22.6$ y) ²¹⁰Bi and ²¹⁰Po are formed. The amount of contaminated blast furnace dust for the EU is estimated at 260 ktons.a⁻¹.

Natural Gas and Oil

Radon enters natural gas in geological formations by diffusion from radium deposits. Oil is often contaminated with radium salts. The radon concentration in natural gas produced in the Netherlands and the Dutch section of the North Sea continental shelf is on average 337 Bq.m⁻³ [4]. In addition to the gaseous radon, water with dissolved salts and solid materials, like sand, are brought to the earth's surface. These latter substances are separated in processing units, forming sludges and fixed scales. The deposits contain radium and the decay products of radon with a specific activity of up to several hundreds of Bq/g ²²⁶Ra and/or ²¹⁰Pb. The volume of waste produced per unit of product is rather low. However, the huge amounts of primary fossil fuels, produced in the EU, leave a not insignificant volume of waste at the production sites, totalling 10,000 m³.a⁻¹. This volume has to be stored in a final repository as α -active material.

Phosphate

Table II gives a summary of the most important sources from which phosphate rock is imported into the EU. About 90% of the phosphate is used for fertilisers. Second best is the use in detergents.

Table II Some typical values of activity concentrations in phosphate rock [6].

Origin	Specific Activity (Bq.kg ⁻¹)		
	U-238	Ra-226	Th-232
Florida	1500	1600	16
South Carolina	4800	4800	78
Morocco	1700	1700	30
Kola	90	40	90
China	150	150	25

Elemental phosphorus

A phosphorus plant near Flushing in the Netherlands has a capacity of 80,000 ton.a⁻¹ of phosphorus, which is nearly the total phosphorus production capacity in the EU. The turnover of ore is 600,000 ton.a⁻¹. The plant produces about 675,000 ton.a⁻¹ of slag with an activity of about 1 Bq.g⁻¹ of ²³⁸U and its progeny, in equilibrium to ²¹⁰Pb. The slag is mainly used for road construction. Due to the high process temperatures, the volatile nuclides ²¹⁰Pb and ²¹⁰Po escape. The dust from the furnaces is filtered and largely recycled to the sintering process. This recycling enriches the dust up to 800 Bq.g⁻¹ of ²¹⁰Pb. Dust is partly extracted and processed to calcinate. The volume of the calcinate amounts to 1000 ton.a⁻¹ with an activity of up to 800 Bq.g⁻¹ of ²¹⁰Pb and 60 Bq.g⁻¹ of ²¹⁰Po. By ingrowth the activity of ²¹⁰Po will reach the activity of ²¹⁰Pb within a year. This is clearly radioactive waste.

Phosphate fertilisers

The common process used for extracting phosphorus from the rock for the production of fertiliser is dissolution in sulphuric acid. The radioactive impurities in the rock are split into different streams. Radium and its progeny are largely co-precipitated with calcium into gypsum and released as slurry into surface water or stored on land. Uranium and thorium remain mostly in the phosphoric acid and get thereupon into the fertiliser. A typical value for the specific ²²⁶Ra activity of gypsum is 0.7 Bq.g⁻¹. The specific activity of the fertiliser is proportional to the amount of P in the product. Values range from 0.3 - 3 Bq.g⁻¹ for ²³⁸U [9].

In the EU, yearly 6-7 MT of phosphogypsum is released directly into the sea and 8-11 MT is stored on dumpsites [10]. Radioactive components dissolved in seawater give negligible problems for the food chain. However, a significant part of the radium is adsorbed on sediments, contaminating estuaries and giving problems when sediments are used for landfill.

Zircon

Zircon is the common name for zirconium silicate (ZrSiO₄). It is a clear white material, mined at a variety of places around the world. It is always contaminated with radioactive materials. About 350,000 ton.a⁻¹ is imported into the EU. Typical values of the specific activity are 5 Bq.kg⁻¹ of ²³⁸U and 0.5 Bq.kg⁻¹ of ²³²Th. Zircon is used for about 1/3 in ceramics, 1/3 in foundries, 1/3 in

refractories and minor volumes in other applications. Before being used in other industries, it is milled at several sites into various grades, down to a median grain size of 1 μm . At zircon mills, the unrecoverable part of zircon is less than 1 % of the turnover. Under the current regulations the waste can be discharged to normal dumping sites. The volume of contaminated waste can be kept small by good housekeeping. Due to the radioactivity of zircon, other industries using the milled product, especially industries with refractory materials, are likely to have a waste problem as well. The volumes might be considerable; a rough estimate for the EU is 100,000 ton per year.

Table III gives an overview of the volumes of waste with enhanced natural radioactivity produced in the non-nuclear industry.

Table III Overview of annual volumes of waste with enhanced natural radioactivity in the EU from the non-nuclear industry, which produce the bulk of waste.

Type of industry	High active*) (kton)	Medium active*) (kton)	Low active*) (kton)	Minor active*) (kton)
Coal	1 (dust)	260	< 300	30,000
Steel				
Gas and oil				
Phosphate				
Zircon				
		1	750 (slag) 14,000-18,000 (gypsum)	
			1	100

- *) High active waste; has to be stored as radioactive waste already under the old exemption levels.
 Medium active waste; has a specific activity just under or above the old exemption levels.
 Low active waste; will exceed current reporting levels.
 Minor active waste; around current reporting levels.

RECOMMENDATIONS

To cope with the problem of disposal of these large volumes, pragmatic approaches for their management have to be sought. A number of recommendations can be given. Some can be implemented without many implications, others require further research and calculations. For some recommendations, adaptation of the national legislation on the storage of radioactive waste may be necessary in certain Member States.

Burial sites

Creation of a number of burial sites in the EU, in a radius of 100 - 200 km around the main industrial agglomerations. Arrangements and provisions of these burial sites can be comparable

with those pertaining to sites for other industrial waste. Burial sites should be prohibited for the public and fenced-off. They should be put under the control of institutions, which are competent on radiation control and the handling of radioactive waste. After closure and covering of the burial site, the land can be given in long lease for certain uses, e.g. sports fields or parks, but housing development should be avoided. This is a space-saving solution and it can be assured that the risks to the public are acceptable.

Interim storage

In industrial processes the equilibrium in the decay chains of uranium and thorium can be disturbed. In some wastes only the nuclides $^{210}\text{Pb}/^{210}\text{Po}$ occur. This is for example the case with dust filtered from stack emissions from high temperature processes by which lead and polonium are volatilised, such as the E-filter dust in the phosphorus processing industry as described. The half-life time of ^{210}Pb is 22.3 year. Each century the radioactivity decays with a factor of 20. So, the waste can be handled or reused as non-radioactive waste after a surveyable period. Interim storage for at most a few centuries, dependent of the original activity, will suffice.

Diluting

The ALARA-principle implies that all exposures shall be kept low. In the Basic Safety Standards of the IAEA [11] the trivial individual annual radiation dose is set at $10\text{ }\mu\text{Sv}$ and the collective annual radiation dose at 1 manSv . It is not unlikely that both criteria can be met by diluting waste containing natural radioactivity into the environment. From the viewpoint of radiation exposure to man, dilution may be beneficial compared to dumping or even controlled storage. The option has to be proven by a risk assessment. A typical example is the dispersion of phosphogypsum into the sea. Individual doses become trivial and the collective dose remains low. However, discharge into estuarine and coastal waters should be avoided as marshlands, harbour sludge or seafood may be contaminated with the potential result of enhanced exposure of man.

Dispersion of ashes by high stacks may also be beneficial, provided the dust concentration near ground level does not exceed certain limits.

Spreading

Terrestrial radiation varies from one region to another. Spreading of waste with relatively low concentrations of natural radioactivity over the land may not, or only with a low percentage, increase the original radiation field. If such condition is fulfilled spreading can be allowed. The permissiveness of this solution can be demonstrated by the use of phosphate fertiliser on farmland. Some PK-fertiliser will exceed the new exemption levels slightly. Calculations show that extended use of phosphate fertiliser will increase the dose rate in the air above ground level with only 0.8 nGy/h , compared to a background value of $30\text{--}70\text{ nGy/h}$. Even if, at a later stage, the land is used for permanent residence purposes, the annual dose will increase only $7\text{ }\mu\text{Sv}$. The consumption of crops cultivated on such farmland is negligible [6].

Phosphogypsum is widely used as a beneficial soil amendment. Doses due to consumption of crops grown on such amended soils are still acceptable. However, radon exhalation may pose a problem when the amended agricultural land is used for housing [12].

Product dependent clearance levels

The scenarios used for the calculation of the exemption levels [5] are of a generic art. For many wastes the incorporated solid natural radioactivity does not spread as easily as assumed in generic models and so it reaches man to a lesser extent. The risk of the disposal of waste depends also on the product. For example, the risk of spreading of the radioactivity from a refractory stone made with zircon sand is lower than the risk of dispersion of uncovered fly-ash. The clearance level for the disposal of waste from certain products can therefore be higher than the exemption level. However, before specifying special product dependent clearance levels a risk analysis should be performed. In the calculations the exhalation of radon has to be taken into account.

Recycling

There is a tendency in Europe to reuse materials in order to decrease the volumes of waste. Some superfluous materials with enhanced concentrations of natural radioactivity from one production process may be used as raw materials or semi-manufactured products in another process without unacceptable radiation risks. A risk analysis is however needed in such cases. Large volumes are reused already in a variety of applications. Fly-ash from coal burning is used widely as an additive to cement; the bottom-ash is used for road construction. It is essential that the reuse of this large volume of low active waste remains possible. Slags from the phosphorus production are extensively used for road construction; another option under study is the use in dikes. The reuse of phosphogypsum is studied worldwide.

Underground

Natural radioactive waste can be stored in cavities in the ground. This may be an alternative for burial sites, perhaps even with a better public acceptance. It is not necessary to specify the same requirements for storage in those cavities as are made for the underground disposal of waste from the nuclear fuel cycle. Cavities used can be old mine cobs, still intact old mines, cavities in salt domes; liquid wastes can be pumped into exhausted oil/gas fields.

Separation

Volumes of waste with enhanced natural radioactivity may be reduced considerably through separation of radionuclides from the bulk of the waste. The small volumes of concentrated waste may then be handled at radioactive waste repositories.

Decontamination

The dismantling of installations in the near future will yield large volumes of waste contaminated with natural radioactivity, e.g. from fertiliser plants, glass furnaces or oil/gas platforms. After decontamination the base materials may be recycled. The volume of the remaining contaminant is only small compared with the original volume.

CONCLUSIONS

A variety of valuable minerals contain minor quantities of uranium, thorium and their descendants and are, therefore, more or less radioactive. Minerals with enhanced natural radioactivity can cause radiation problems during processing. Residues to be removed as waste can give a radiation dose to the public, especially if large volumes are involved.

The exemption levels recommended in the new BSS for waste containing enhanced levels of naturally occurring radionuclides are substantially lower than the former levels. Consequently, solutions have to be found for the disposal of large volumes of radioactive waste from the non-nuclear industry.

Recommendations are given for the practical management. The recommended measures can be introduced separately, but combinations are also possible, depending on the location where the waste is produced, the volumes, the possibilities of recycling and other factors. Some recommendations may be implemented without serious implications. Others will require investigations and studies. In some Member States legislation should be adapted before the given recommendations can be implemented.

As, in general, the specific activities and leachability of the radioactive elements are low, the natural radioactive wastes may be stored in repositories other than those for nuclear waste. Special burial sites may be selected, especially for wastes containing only $^{210}\text{Pb}/^{210}\text{Po}$.

Diluting and spreading may be good options, provided the risks have been calculated by means of risk assessment models. Both individual doses and collective doses can be even lower than in case of storage.

Before recycling the base materials, decontamination or separation of the radioactive elements may be necessary, for which techniques are yet to be developed.

The disposal of phosphogypsum creates the biggest problems, due to its volume. Possibilities of reusing the gypsum have to be studied.

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