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INTERPRETATION OF *NORM* MEASUREMENTS WITH RESPECT TO THE ENFORCEMENT OF DUTCH LEGISLATION

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In the past few years a large variety of objects contaminated with Naturally Occurring Radioactive Materials (NORM) have been found in the Netherlands. NORM is subjected to the Dutch Nuclear Energy Act if the radioactivity exceeds certain limiting values. Limits for possession, handling and disposal of radioactive materials are laid down in the Radiation Protection Decree and the Decree on Nuclear Installations, Fissile Materials and Ores. Limiting values focus on the specific activity and the total activity of the source as a whole. Different limits are set depending on whether or not the radioactivity is in its natural matrix. All limits have to be considered in relation to the definition of the "source". For a selection of cases, results have been interpreted with reference to the enforcement of Dutch legislation.

INTRODUCTION

NORM is subjected to enforcement of the Dutch Nuclear Energy Act [1] when the radioactivity exceeds certain limiting values. This act, in which European Community directives and guidelines are implemented, forms the basis for laying down limits and regulations in decrees. For NORM these are the Radiation Protection Decree (BSK) and the Decree on Nuclear Installations, Fissile materials and Ores (BKSE). Exemption levels and dose limits for radiation protection are defined in the legislation and rules for authorisation and licensing are implemented. In practise this means that for the process industry the emission to air and water and the handling, disposal or reuse of radioactive waste materials are subjected to legislation.

The task of the Dutch Inspectorate for the Environment (IMH) is to enforce the Dutch legislation with respect to NORM. IMH is assisted by the Laboratory of Radiation Research (LSO) of RIVM in screening objects and materials on the spot, performing analyses in the laboratory and advising IMH on interpreting measurements. In the past few years, NORM contaminated objects have become of growing concern for IMH. In general these objects (tubing, valves, etc.) appear when (parts of) plants of the process industry are dismantled. There is also a lot of trading and storage of metal scrap, ore concentrates and smelt in the Netherlands. These materials, usually packed in sea-containers, come in by ship. In the past few years, the radiation levels on the outside of these containers have occasionally found to be high. Implementing legislation for these objects and materials, which is an ongoing process for IMH, is not straightforward. For some cases, e.g. the oil- and gas-producing industry, IMH has already standardised some practical guidelines. This paper presents the results of the analyses for a number of objects and materials along with IMH's interpretation of these results with respect to enforcing Dutch legislation.

DUTCH LEGISLATION

Relevant for objects contaminated with NORM are the exemption levels for radioactive substances as given in the BSK, which makes a distinction between natural radioactivity on the one hand and artificial radioactivity on the other. Natural radioactivity means that the radioactivity is in the natural matrix, in this situation there is a secular equilibrium between mother and daughters in the decay series of the primordial nuclides. A permit is required if natural radioactivity exceeds the limit of 500 $Bq \cdot g^{-1}$, a value at which the specific activity of all the radioactive nuclides in the substance are taken into account. Fourteen nuclides for the ^{238}U-series and 11 and 10 for the ^{235}U-series and ^{232}Th-series, respectively, contribute to the total specific activity. In general, processing NORM (e.g. by the fertiliser or gas- and oil- producing industry) may cause a disturbance of the natural matrix and the secular equilibrium in the decay schemes. Examples of disturbances are the leaching of the mobile radium isotopes away from their geochemically immobile uranium and thorium parents, the escape of the noble gas radon or the evaporation of the lead isotopes. If NORM is not in the natural matrix, in the Netherlands a limit of 100 $Bq \cdot g^{-1}$ is then applied for the specific activity. As previously mentioned, this specific activity is the total of the specific activities of all nuclides present in the sample. The limits for the specific activity set in the BSK are only applicable if, at the same time, the total activity of the source is more than 5 kBq for very high, 50 kBq for high, 500 kBq for moderate and 5 MBq for low radiotoxic nuclides. In the context of BSK, "source" is defined as the total amount of radioactive substance that can be considered as being one unit.

The limits mentioned above apply to materials like ore concentrates as well. Moreover, these materials are sometimes subjected to the BKSE. The BKSE applies to ores that contain a weight percentage of more than 0.1 percent uranium and/or 3 percent of thorium (for monazite the value for thorium is 10 percent). Such ores are considered to contain fissile material. The storage and handling of these ores are subjected to the rules of the BKSE.

SCREENING, SELECTION AND ANALYSIS

The general procedure in finding objects contaminated with NORM that have a high activity concentration is to screen the external radiation level or the surface contamination of potential sources and to select those that exceed a pre-set level, e.g. three times the background radiation level. In the Dutch oil- and gas-producing industry, such a screening takes place, in principle, by the industry to decide upon measures that should be taken before the handling or disposal of the objects. Moreover, in the Netherlands many of the (major) scrap metal yards already have the expertise and the equipment to monitor radiation levels in scrap at the entrance of the yard (gate detector). In general the detection level of the gate detector is set as low as possible to avoid missing radioactivity due to shielding. IMH has implemented a general directive according to which scrap yards are obliged to notify IMH about any radioactive scrap found [2]. Where transport and storage of ores and ore concentrates are concerned, some of the incidents were found through controls performed by the customs officials. These incidents are reported to the IMH too.

If IMH decides upon further investigations, RIVM/LSO, under the authority of IMH, subsequently verifies the reported radiation levels on the spot and identifies the objects or substances concerned. On the basis of visual inspection and screening of the external radiation level or surface contamination, the distribution of the contamination is determined. A method for rapid determination of activity concentrations in NORM using gammaspectroscopy was developed by RIVM/LSO to perform analyses on the spot [3]. For these analyses RIVM/LSO uses a van equipped with, among other things, a germanium detector and analysis software. This method gives an indication of the specific activity. Therefore, if possible, subsamples of the objects (e.g. part of a tubing) and/or of the contamination (e.g. sludge, scales, etc.) should be taken for a more accurate analysis in the laboratory.

Mostly, these analyses are performed by gammaspectroscopy. As the radionuclides from the ^{238}U, ^{235}U and ^{232}Th series are not always in secular equilibrium, the specific activities of all nuclides in each series have to be determined. However, on the basis of chemical behaviour and decay it is possible to define a number of subseries in each series. These subseries can be assumed to be in secular equilibrium. Finding one nuclide of a subseries is then sufficient to quantify the subseries as a whole. In this way the analysis is limited to 7 γ-emitting nuclides of the ^{238}U series, 5 for the ^{235}U series and 4 for the ^{232}Th series. The composition and the density of the sample have to be known to correct for the deviation from the calibration source. However, in most cases the composition of the sample is not exactly known, so assumptions have to be made resulting in an increasingly uncertain analysis.

Table 1 gives the results of the analyses for a number of samples. In the table the exposure rate is represented by the range of measurements at the outside of the object (tubing, valve, bag with ore concentrate, etc.), the specific activity is the range of the total specific activity of the different objects or materials and the total activity is the range of the total activity of the defined sources due to the very high radiotoxic nuclides.

CASES

Sludge

At a waste disposal site, a number of drums were found to contain sludge in bags. An exposure rate of more than three times the background was found on the bag surfaces. The waste materials originated from the gas-producing industry. The sludge was very inhomogeneous, containing, for example, pieces of rope, plastics and paper, which made it difficult to get representative samples of the sludge. Only the dry fraction of the sludge, rid of all other substances, was analysed. Since it was found that only ^{226}Ra and ^{228}Ra and their daughters were present in this sludge, the radioactivity of the sludge is considered to be non-natural. The specific activity of the (clean) dry fraction was found to be in the range of 20-50 $Bq \cdot g^{-1}$. The limit of 100 $Bq \cdot g^{-1}$ is therefore not exceeded. In this case the source was defined as the total amount of the (clean) dry fraction of the batch's sludge. This source contained about 50 kBq of very high radiotoxic nuclides. Based on the analysis it was concluded that there was no need for further action by IMH.

Objects originating from the fertiliser industry

A number of times a scrap yard discovered a container with an elevated exposure rate, as indicated by the gate monitor. Objects (tubing, valves, etc.) in the container originated from the fertiliser industry. The objects were found to be contaminated with scale, appearing in the form of $CaSO_4$ (gypsum). Some of the scale was present as a thick layer (a few cm) and was easy to remove. In other cases, the scale formed a thin layer (< 1 mm) and was less easy to remove. Samples of the scale were taken from the different objects and the dry fraction was analysed. In both scales only ^{226}Ra, ^{227}Ac and ^{228}Ra and their daughters were found. The thin scale had a very high specific activity, exceeding the limit of 100 $Bq \cdot g^{-1}$ and the thick scale had a specific activity of less than 100 $Bq \cdot g^{-1}$ (see table 1). Again the limit of specific activity is applied to the dry fraction of the scale without taking the overall weight of the object into account. The source was defined as the total amount of scale per object. Here, the total activity for a number of objects was also above the limits. Therefore IMH reported on the offence for the fertiliser plant originally in possession of the objects. Moreover, IMH demanded that the plant should apply for permit adjustment to prevent the events described above.

Objects originating from the gas-producing industry

A scrap yard reported that for a load of tubing, where each tube length measured about 8 m, elevated levels of surface contamination were detected on the inside of some of the tubing. The exposure rate on the outside was not elevated. The tubing originated from the gas-producing industry. RIVM/LSO selected two lengths of tubing with a contamination monitor. From these two lengths, small pieces (about 20 cm) were cut and taken to the laboratory for analysis. These pieces contained scale, which could partly be removed by scraping and decontamination techniques. Another part of the scale was in the form of plating on the tubing and could not be removed in this way. On analysis of samples of the removable part of the scale it was found to contain only ^{210}Pb and daughters in secular equilibrium. The specific activity ranged from 250 to 500 $Bq \cdot g^{-1}$ (see table 1). For the different samples the limit of 100 $Bq \cdot g^{-1}$ was applied again only to the dry fraction of the scale. The source is defined as the total amount of scale in a tubing. Based on the samples analysed, the distribution of the scale was concluded to be very inhomogeneous. It was not possible to determine the total activity of the source without additional investigations, e.g. measuring the surface contamination over the total length of the inside of the tubing. On the basis of the specific activity, IMH concluded the doubt around the limit of total activity exceeded to be reasonable. However, from measurements performed by the facility that decontaminated the tubing before disposal, it was concluded that at several places in the tubing, the surface contamination for α-radiation was higher than 0.4 $Bq \cdot cm^{-2}$. This is higher than the limiting value in the practical guidelines set by the oil- and gas-producing industry [4]. Therefore the deco-facility and the original owner of the tubing were given a warning and were urged to improve their control measures.

Slagwool

Back in the years 1960-1970 a type of slagwool containing enhanced levels of radioactivity was used in e.g. fire-doors. A number of times in the last years such fire doors were found at a scrap yard. The specific activity of the slagwool was in the range of 150 to 200 $Bq \cdot g^{-1}$ (see table 1). As this material contained non-natural radioactivity the limit of 100 $Bq \cdot g^{-1}$ was applied. The source was defined as the total amount of slagwool in the door. Estimating this to be a few kg indicates that also the limit of 5 kBq for the very high radiotoxic nuclides was exceeded. As both limits were exceeded, IMH documented the offence and obliged the owner of the doors to apply for a permit on handling and disposing of radioactive materials.

Smelt

A container with a load of large pieces (20 - 50 kg) of metal melts was found for which the radiation level, by accident, triggered the gate detector of a nearby scrap yard. The smelt originating from (Fe Si Sn) ore which had already undergone a process of smelting and for which part of the Sn was removed. The pieces were (partly) covered by a thin, hard to remove, layer of slag of a different composition. It was estimated that less than 5% of the smelt consisted of slag. From the screening of the radiation level most of the radioactivity was concluded to be in the slag layer. One piece was found that consisted mostly of slag. After grinding, this piece and also a piece with an average thickness of the slag-layer were analysed on the spot by gammaspectroscopy [3]. The specific activity of the slag was about 100 $Bq \cdot g^{-1}$, the smelt showed no activity above the detection limit (see table 1). Considering the uncertainty in the value of the specific activity IMH, decided that the limit for the specific activity was not exceeded. As a prevention measure, IMH gave the company a warning and advised it to implement detection techniques like e.g. a gate detector.

Ore concentrates

In most of the cases the ores had already undergone some treatment, e.g. grinding and removing unwanted by-products by using separation techniques (e.g., flotation or gravimetric techniques). In general, a secular equilibrium had been found, so the radioactivity in the ore concentrates was considered to be in the natural matrix. Therefore the limit of 500 $Bq \cdot g^{-1}$ could be applied. In one specific case, Baddeleyite, it was found that the ^{232}Th subseries was out of secular equilibrium. Therefore the 100 $Bq \cdot g^{-1}$ limit has to be applied. As it is not clear which process is responsible for the observed disturbance, IMH decided to perform additional investigations before implementing the limits. For the other cases, where the limit of 500 $Bq \cdot g^{-1}$ was exceeded (see table 1), IMH made a report of the offence. As warehouse companies have little influence on the product they store, at least one company concerned, decided to apply a permit for the handling and storing of radioactive materials to prevent future problems.

***Table 1*: Results of analysis of NORM-samples (see text)**

Object/ origin	Characterisation	Exp.rate $\mu Sv \cdot h^{-1}$	Spec. act. $Bq \cdot g^{-1}$	Total activity kBq
Drums with bags of sludge/ gas-producing industry	Sludge: ^{226}Ra-, ^{228}Ra and daughters, ^{210}Pb/^{226}Ra ratio as low as 0.2.	0.1-0.5	20-50	50
Tubing/ gas-producing industry	Scale, thin layer, partly removable. Only ^{210}Pb and daughters.	Background (0.03)	250-500	about 3-10
Tubing with a rubber lining/ fertiliser industry	Scale, thin layer, partly removable; ^{226}Ra-, ^{227}Ac- and ^{228}Ra- and daughters. Almost no activity due to ^{232}Th-series.	0.2-1.5	7000-17000	1000-10000
Tubing, valves, etc. with thick scales/ fertiliser industry	Scale, thick layer, easy removable. Relatively high values of ^{210}Pb. Almost no activity due to ^{232}Th-series.	0.1-0.5	1-60	5-8000
Slagwool/ Fire doors	Most activity is due to ^{232}Th series.	0.1-0.2	150-200	
Bags with ore concentrates/ Africa	Natural matrix. Exception found is baddeleyite: ^{232}Th is not is secular equilibrium.	1-45	150-2500	1 bag: 1300 - 22500
Smelt originating from (Fe Si Sn)/ Brasil	Smelt with (about 5%) slag. Almost no activity due to ^{238}U series	0.1-1.5	< 100 (slag: 100-200)	about 35000

DISCUSSION AND CONCLUSION

From the above selection of cases discussed it is clear that for the enforcement of the general rules in the legislation a set of practical guidelines is needed. IMH has already agreed upon such a set of guidelines with the oil- and gas-producing companies [4]. In the set of guidelines three different steps are defined. First, these companies have implemented screening limits for their installations: for closed installations the exposure rate has to stay below a level of 3 times the background, and for open installations the level of the α- or β-activity should be less than 3 times the background. Second, if the screening limits are exceeded, (parts of) the installation should be investigated to ascertain whether or not the specific activity of the NORM contamination exceeds 100 $Bq \cdot g^{-1}$. Third, if this is the case, (parts of) the installation should be decontaminated before handling or disposal. Moreover, IMH is in agreement with the oil- and gas- producing industry that disposal of decontaminated materials may take place if the surface contamination remains under the limit of 4 $Bq \cdot cm^{-2}$ for β- and γ-radiation and under 0.4 $Bq \cdot cm^{-2}$ for α-radiation. These surface contamination limits were also applied by IMH to materials originating from the dismantling of a fertiliser plant. At the moment consideration is being given to also applying the limits for surface contamination to scrap metal presented for decontamination. A further suggestion for the metal and scrap-metal branch is that if a load of scrap contains radioactivity, the screening method already used by the oil- and gas-producing branch of industry will also be applied to the objects present in the load.

At the moment the implementation of the EU's new Basic Safety Standards Directive does not seem to be altering the practice of enforcement. However, as discussed above, it would be very useful if limits with respect to surface contamination were to be part of the new legislation. It should be noted that the above mentioned limits for surface contamination are not based on dose or risk evaluations but on practical arguments only (e.g. technical possibilities and detection limits). Such an evaluation is needed for the implementing the limits in the legislation. In addition, applying limits also have to be considered in relation to the definition for the "source". It is not always a straightforward matter to define the "source": is it only its scale within the object (e.g. tubing) or is it the container as a whole (e.g. the total smelt load)? In practice, IMH applies the concept that each object (tubing, valve, etc.) is defined as the source, for the ore concentrate, the containment (e.g. the bag) with the ore is considered as the source and in the case of large pieces of smelt, each separate piece may be considered as the source.

On the basis of the cases described, it is concluded that a thorough investigation is needed to prove that a law has been broken. There should not only be a detailed determination of the (radiological) characteristics of the objects or materials found but also of the implementation of the (general) rules laid down in the legislation. For this it is the intention of IMH to stipulate exactly how the rules have been implemented up to now and to continue to improve this information on the basis of ongoing accumulation of experience: "learning by doing".

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