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The fate and behaviour of enhanced natural radioactivity with respect to environmental protection

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ABSTRACT

In contrast to the monitoring and prevention of occupational radiation risk caused by enhanced natural radioactivity, relatively little attention has been paid to the environmental impact associated with residues containing enhanced activity concentration of naturally occurring radionuclides. Such materials are often deposited directly into the environment, a practice which is strictly forbidden in the management of other types of radioactive waste. In view of the new trends in radiation protection, the need to consider the occurrence of anthropogenically enhanced natural radioactivity as a particular unique case of environmental hazard is quite apparent. Residues containing high activity concentrations of some natural radionuclides differ from radioactive materials arising from the nuclear industry. In addition, the radiation risk is usually combined with the risk caused by other pollutants. As such and to date, there are no precise regulations regarding this matter and moreover, the non-nuclear industry is often not aware of potential environmental problems caused by natural radioactivity. This article discusses aspects of environmental radiation risks caused by anthropogenically enhanced natural radioactivity stored at unauthorised sites. Difficulties and inconclusiveness in the application of recommendations and models for radiation risk assessment are explored. General terms such as "environmental effects" and the basic parameters necessary to carry out consistent and comparable Environmental Risk Assessment (ERA) have been developed and defined.

1. Introduction

Radioactivity is a primordial property of matter and the environment. Since life evolved on Earth, every living organism has been exposed to ionising radiation. Although national attitudes to the management of naturally occurring radioactivity are variable, there is often a tendency to remove exposures arising from such sources from radiological control in the process of exclusion and exemption. This normally occurs where naturally occurring radionuclides are not amenable to control. Potassium-40 in the human body is a good example of this category, or where radiological controls, in cases where excessive resources would be required in implementation, might be considered unwarranted. Nonetheless, there are many areas on Earth which have elevated "natural background" radiation caused either by the geological or geochemical structure of underlying rocks and whether or not this can cause a negative or positive effect on humans is a subject of mixed opinion within the wider scientific community. In contrast, if concentrations of natural radionuclides have been changed deliberately or accidentally by humans, radiation protection principles dictate that the situation is managed in an active manner. There are two abbreviations documented in scientific literature: NORM (Naturally Occurring Radioactive Material) and TENORM (Technologically Enhanced Naturally Occurring Radioactive Material) used for the description of such situations (Michalik, 2009). Alterations in the natural state resulting in TENORM, or enhanced levels of NORM, can result in an incremental increase in the radiation risk to people and non-human biota. Hence, the monitoring and prevention of occupational radiation risk caused by enhanced natural radioactivity has become obligatory in many cases for industries processing NORM/TENORM. Usually, the applied radiation risk constraints have been derived based on the basic principles of radiological protection in the same way as risks related to artificial radioactivity are considered. Case-specific risk scenarios have also been developed with the requirement of developing criteria that are applicable for industry operators (Radiation Protection 107, 1999).

Compared to the occupational risk and the risk to members of the public, which are often comprehensively assessed and managed in many cases of NORM/TENORM industries, relatively little attention has been paid to the potential, direct environmental impact associated with residues originating from these industries. Recently, however, the problem has been elucidated to a certain degree. The new

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recommendations of the ICRP have pointed out the necessity to consider radiation risk to non-human biota independently from the risk to humans (ICRP Publication 103, 2007). Simultaneously the draft of new IAEA Basic Safety Standards (2010) as well as the European counterpart has underlined the importance of radiation risk caused by NORM/TENORM. The combination of both new requirements generates a new subject of radiation protection: environmental risk caused by residues containing enhanced natural radioactivity. As a natural consequence of public concern, uranium has often been at the centre of interest for scientists working on natural radioactivity. However, due to its chemical toxicity, uranium is arguably a poor representative example of interactions between natural radionuclides and the environment (Sheppard et al., 2005). Although few studies have considered the impact of NORM on the environment, there are several recent publications that illustrate a burgeoning interest in this subject. The environmental impact arising from an area contaminated by uranium mill tailings and radium production wastes was considered by Evseeva et al. (2009). The study suggested that decreases in chromosome aberration frequency were occurring more slowly for herbaceous plants from this NORM contaminated area compared to plants contaminated by anthropogenic radionuclides arising from the Chernobyl accident. The genotoxic effects of sediment and water contaminated by radium isotopes were investigated by Geraskin et al. (2011). Hosseini et al. (2011) applied a recently developed environmental impact assessment methodology to selected NORM contaminated sites allowing the dose-rates for a suite of organisms to be contextualised in terms of various screening and protective benchmarks.

The activity concentration of natural radionuclides in NORM/TENORM residues can be high enough to classify them as radioactive waste but, in particular, they differ significantly in quantity and quality from radioactive materials arising from the nuclear fuel cycle or spent radioactive sources (Baxter, 1996). Furthermore, such residues are often deposited directly into the environment, a practice which is strictly forbidden in the management of other types of radioactive waste. In addition, the derived exposure to radiation is usually combined with their chemical toxicity and the risk caused by the occurrence of other pollutants. It results in that in many cases of NORM/TENORM waste, control measures are applied just to avoid the risk related to the presence of non-radioactive pollution. However these controls are often not adequate or sufficient to avoid the radiological impact on either people or the environment. Pathways of exposure originating from NORM/TENORM will differ depending on the type of waste industry. Residues, containing enhanced concentrations of natural radionuclides, cover a great variety of chemical compounds enclosed in different matrices. For this reason, each case involving the presence of NORM/TENORM, and even sometimes in the same branch of industry, needs to be assessed in a separate way depending on the source of raw materials or (industrial) processes applied (O'Brien and Cooper, 1998). Moreover, behaviour of individual radionuclides during the technological process and following release into the surrounding environment can be influenced by the properties of the original or derived minerals (Paul and Pillai, 1986). For example, radium contained in phosphate rock or phosphogypsum (Hull and Burnett, 1995) can behave differently to radium in zircon (Morawska and Jeffries, 1994) and radium in scales from the oil and gas industry (El Afifi et al., 2009; Godoy and Petinatti da Cruz, 2003) or tailings from coal mining (Leopold et al., 2007). The various behaviours associated with the above forms are also important when the NORM/TENORM residues are stored in conditioned repositories. This largely explains why NORM/TENORM cannot be managed directly by applying the rules designed and developed for typical categories of radioactive waste. For the same reasons, the possible impact on the surrounding environment caused by NORM/TENORM cannot be directly compared with effects related to the common presence of terrestrial radionuclides in a non-altered state where an assumption that all natural radionuclides in the soil substratum are in secular equilibrium is well justified. In spite of this, the characterisation of natural background provides a useful means of contextualising dose rates when an environmental impact assessment is carried out (Beresford et al., 2008).

As such and to date, there are no precise regulations regarding NORM/TENORM in the context of the environment and moreover, the non-nuclear industries of concern are often not aware of potential environmental problems caused by natural radioactivity.

2. Features of environmental impact of NORM/TENORM residues

Deposition of NORM/TENORM residues in the environment firstly results in enhanced exposure to external gamma radiation, this being largely determined by the physical attributes of the waste. Furthermore, NORM/TENORM wastes differ significantly from radioactive wastes from the nuclear industry in terms of:

- source geometry, location and the types of dispersion-transfer models that need to be applied: typical NORM repositories have the appearance of common waste dumps and tend to have more in common with industrial waste than with spent nuclear fuel cycle or dispensable radioactive sources,
- total amount: NORM/TENORM residues are usually bulk materials, e.g. phosphogypsum, slag, sediments, and sometimes water,
- ambient conditions: residues, when deposited at a site unauthorised from a radiological point of view, are usually in direct contact with the environment, which means that they are exposed to meteorological conditions (water and wind erosion) and unlimited access by biota.

The location, extent and severity of radiological hazards can be altered by the migration of mobile fractions of radionuclides to the areas surrounding a particular waste dump. Hence, accurate measurements of total radionuclide concentrations in waste materials and compartments within the surrounding environment are crucial for the assessment of potential radiological risk. However, even when it is possible to gather data about the physical occurrence and distribution of each particular radionuclide, this information would give only a part of the knowledge necessary to evaluate the potential for harmful exposures. Radionuclides released into a neighbouring ecosystem can become involved with the processes which occur at the interface between the abiotic and biotic environments e.g. in processes occurring in the rhizosphere, and finally can be transferred through all trophic levels. Furthermore, radiation risk may be amplified by direct interaction high LET radiation and living matter. Therefore, in order to assess environmental impact, regardless of how this is defined, information on radionuclide chemical composition and speciation, interaction and transfer within affected ecosystems and spatial and temporal distributions of radionuclide species, influencing their mobility and biological uptake, is essential. It is thus necessary to estimate relevant parameters such as:

- radionuclides speciation (e.g. particle size, charge, valency and geochemical association),
- TF (substrate-biota transfer factor),
- K_d (solid–liquid distribution coefficients) are usually defined for four typical types of soil although published collated data does not contain information for waste,
- mineral composition.

Natural radionuclides are subject to sequential decay. Thus, it has been usually inferred that most of the radionuclides in the ^{238}U and ^{232}Th decay series occur in a state of secular equilibrium, but in NORM/TENORM residues arising from industrial processes, the state of secular equilibrium is usually strongly altered. In acquiring the data listed above, it is necessary to characterise the material. The initial elemental composition of the residue, influencing its physical and chemical properties, changes with time due to the processes of decay and ingrowth. Besides the new elements that appear in the process of