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Potential radiological impact of the phosphate industry on wildlife

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ABSTRACT

The activities of the phosphate industry may lead to enhanced levels of naturally occurring radioactivity in terrestrial and aquatic ecosystems. We performed a preliminary environmental risk assessment (ERA) of environmental contamination resulting from the activities of 5 phosphate fertiliser plants (located in Belgium, Spain, Syria, Egypt, Brazil), a phosphate-mine and a phosphate-export platform in a harbour (both located in Syria). These sites were selected because of the availability of information on concentrations of naturally occurring radionuclides in the surrounding environments. Assessments were generally performed considering highest environmental concentrations reported in the studies. The ERICA Tool, operating in a Tier 2 assessment mode, was used to predict radiation dose rates and associated risk to the selected reference organisms using the ERICA default parameter setting. Reference organisms were those assigned as default by the ERICA Tool. Potential impact is expressed as a best estimate risk quotient (RQ) based on a radiation screening value of 10 μGy h^{-1}. If RQ ≤ 1, the environment is considered unlikely to be at risk and further radiological assessment is not deemed necessary. Except for one of the cases assessed, the best estimate RQ exceeded 1 for at least one of the reference organisms. Internal exposure covered for 90–100 % of the total dose. ^{226}Ra or ^{210}Po were generally the highest contributors to the dose. The aquatic ecosystems in the vicinity of the phosphate fertiliser plants in Tessenderlo (Belgium), Huelva (Spain), Goiás (Brazil) and the terrestrial environment around the phosphate mine in Palmyra (Syria) are the ecosystems predicted to be potentially most at risk.

1. Introduction

The need for investigating potential risks induced by radiological contaminants on non-human biota and ecosystems is now internationally recognised (ICRP, 2003, 2007; IAEA, 1992, 2006, 2011; UNSCEAR, 1996, 2008). Recommendations and guidelines at the international level and a comprehensive system to protect the environment from ionising radiation are under development. A number of approaches/tools to estimate dose rates to wildlife have been developed and some of them are being used in a regulatory context (Copplestone et al., 2001; US-DOE, 2002; Brown et al., 2008). Initially, risk assessment focused exclusively on human health protection, but the demand for risk assessment has now been extended to include wildlife. As a consequence, ecological or environmental risk assessment (ERA) is a discipline that has undergone considerable development in the last decades with guidelines being developed (Environment Canada, 1997; EC, 2003). ERA is an increasingly important component in any decision-making process aiming to provide transparent management decisions on environmental practices and associated problems.

The phosphate industry's activities may potentially lead to increased levels of natural radioactivity in the environment. Phosphate rocks contain relatively high concentrations of naturally occurring radioactive materials from the uranium and thorium decay series (^{238}U and ^{232}Th). The mean uranium content in the ore of Moroccan origin is 125 mg kg^{-1} (1500–1700 Bq kg^{-1} ^{238}U; 1500–1700 Bq kg^{-1} ^{226}Ra; 10–200 Bq kg^{-1} ^{232}Th; Martin et al., 1977). Phosphate ores are particularly insoluble and the primary step in the production process is the leaching of phosphate from the rock with strong acids. In 90% of the cases, ore is treated with sulphuric acid to produce phosphoric acid and gypsum. Uranium and thorium become enriched in the fertiliser to about 150% of their original concentrations and radium reduced to 10% of the original concentration. About 80% of the ^{226}Ra, 30% of the ^{232}Th and 14% of the ^{238}U is left in the phosphogypsum waste (Martin et al., 1977). The production of 1 tonne of phosphate requires the extraction of 3 tonnes of ore, resulting in the generation of 4–5 tonnes of phosphogypsum with a mean ^{226}Ra content of

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800–1250 Bq kg^{-1} (Martin et al., 1977). Historically, the local dump sites, containing several TBq of radium, are often unprotected from rainfall and are hydraulically connected to surface waters and to shallow aquifers.

If hydrochloric acid is used to extract phosphate from the ore, fertilisers and chemicals almost free from radioactivity are produced and most ^{226}Ra present in the phosphate ore is dissolved in the liquid effluent. The ^{238}U released from the ore is precipitated by lime addition and accumulates with CaF_2 sludge on the dump site. Adding a $BaSO_4$ precipitation step can effectively decontaminate the liquid effluents but results in an accumulation of ^{226}Ra in the dump site, potentially creating local environmental problems due to ^{222}Rn emissions (Baetslé, 1991).

Mining, milling, transporting of phosphate ores, manufacturing of phosphate fertilisers and using phosphate fertilisers containing uranium are ways in which workers, public and environment are exposed to enhanced natural radioactivity (IAEA, 2004). Most of these natural radionuclides are found in the solid waste of the phosphate fertiliser industry (such as phosphogypsum), and to a lesser extent in discharged effluents and dust. Many studies in the world have been carried out to assess the risk to man and the environment (e.g. Othman and Al-Masri, 2007; Carvalho, 1997; Martínez-Aguirre et al., 1996).

Radon emanation and particulate air emissions from the mining areas, phosphogypsum piles, from phosphate ore storage and loading activities in harbours, leaching of radionuclides from phosphogypsum into groundwater and effluent discharges to rivers and marine environments have resulted in contamination of the surrounding environment. As a result, enhanced concentrations were recorded in surrounding soil and plants, sediment, water and aquatic organisms, marine biota, groundwater (e.g. Carvalho, 1997; McCartney et al., 2000; IAEA, 2004; Villa et al., 2009).

A preliminary assessment of the potential impact on wildlife by the P-industry was carried out for following case studies: (1) Belgium: P-fertiliser plant at Tessenderlo-Chemie; (2) Syria: P-mining (Palmyra), P-fertiliser plant (Homs) and P-export platforms (Tartous port); (3) Spain: P-fertiliser plant (Huelva estuary); (4) Egypt: P-fertiliser industry (Nile River); (5) Brazil: P-fertiliser plant (State of Gioás, Bugre and Mogi Rivers). The hypothesis of this study is that predicted dose rates to wildlife at these sites require further detailed investigation (beyond a simple screening assessment) to assess protection status, because they have enhanced environmental concentrations associated with the environmental release of natural radionuclides in these areas likely to lead to significant doses exceeding the screening dose rates for some species.

2. Approach

ERA is a multistage process, starting with hazard identification, followed by exposure, effects and risk assessment. The first stage of any ERA is the problem formulation, which deals amongst others with the characterisation of the contaminant source term and the identification of potential ecological targets and the associated exposure pathways. The ERICA non-human biota assessment tool (Brown et al., 2008) was used to calculate dose rates to the reference organisms available in the default configuration of the tool, based on this information. Default parameter values available in the ERICA tool incorporate a steady-state representation of transfer which results in a conservative assessment. This is within frame of the current regulatory framework, which requires analysing consequences of chronic, long-term releases, which tend to be essentially at equilibrium in biota. Transfer parameters and occupancy factors are also selected such that they lead to conservative dose assessments (Brown et al., 2008).

2.1. Derivation of environmental concentrations

Information on environmental contamination levels (soil, sediment and water concentrations) was collected from literature data. In cases where, for the selected case studies, environmental concentrations at different locations were presented, we generally considered the higher concentration areas (data selection is well referenced below). For soils and sediments in the vicinity of P-mines or P-export platforms in harbours, secular equilibrium for the ^{238}U chain was assumed and if no information on all daughter radionuclides (or mother radionuclide) was available equilibrium with the closest ascending or descending available member of the decay chain was assumed. Therefore, it is considered that ^{234}Th, ^{234}U and ^{230}Th are in equilibrium with ^{238}U and that ^{210}Po is in equilibrium with ^{210}Pb and ^{226}Ra. Assuming a representative value of 20% for the loss due to ^{222}Rn emanation from rock and soil (UNSCEAR, 2000), the ^{210}Po and ^{210}Pb concentrations in soil, sediment and water constitute 80% of the concentration of ^{226}Ra.

Limited or no information was available for ^{232}Th chain radionuclides. For the Brazil scenario, reported concentrations for ^{238}U and ^{232}Th were comparable and for the Nile River scenario the whole ^{232}Th decay chain was considered to be in equilibrium. For the Syrian study, based on available information on the ^{232}Th chain daughters (^{224}/^{228}Ra concentration being only 1% of the ^{226}Ra concentration), the dose contribution from the ^{232}Th chain was considered negligible compared to the ^{238}U chain.

Since the Dose Conversion Coefficients (DCC) of a parent nuclide in the ERICA tool include all daughters with half-life up to 10 d, only daughter nuclides with half-life >10 d were considered separately.

For releases from phosphogypsum piles (H_2SO_4 wet process), no equilibrium with the parent nuclide was assumed for ^{226}Ra, given that U is mainly retained in the fertilisers. For releases from the phosphate-fertiliser plant of Tessenderlo-Chemie in Belgium (HCl wet process) to the aquatic environment, 30–40% of the radium in the ore is retained in the CaF_2 sludge together with virtually all U and Th (Martin et al., 1997). The surface water contamination was mainly due to the radium released with the soluble $CaCl_2$ waste streams, in which no U or Th was present (Vanmarcke and Paridaens, 1999). Therefore, no equilibrium with the parent was assumed for ^{226}Ra released to the rivers.

Where concentrations in water were not provided, they were calculated using the default solid–liquid equilibrium distribution coefficients (K_d) provided by the ERICA tool. The concentration ratios (CR) for the selected radionuclides and the default reference organisms used in the assessment were the default values provided in the ERICA tool (Brown et al., 2008; Beresford et al., 2008a; Hosseini et al., 2008). The assessment results are very dependent upon the choice of CR and, where relevant, K_d values. A number of papers have now considered the importance of these parameters and their variability on overall assessment uncertainty (Beresford et al., 2005, 2008b, 2008c).

Only the ^{238}U series and ^{232}Th series (if data available) were considered in the dose evaluation. The ^{235}U-series was not considered because ^{235}U in naturally occurring radioactive materials is present in a much smaller quantity than ^{238}U (natural abundance of 0.72%) and therefore it does not contribute significantly to the overall dose. Indeed, simple imput of U-radionuclides at its natural abundance concentrations in the ERICA tool shows that ^{235}U only contributes 1% to the dose rate of aquatic organisms, compared to ^{238}U.

2.2. Assessment of dose rate and associated risk

The ERICA assessment tool, developed under EU-sponsorship (Beresford et al., 2007; Brown et al., 2008) was used to assess