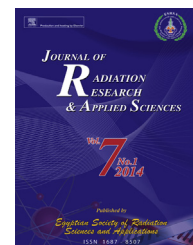


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Natural radioactivity assessment of a phosphate fertilizer plant area

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ABSTRACT

Rock phosphate ore processing and disposal of phosphogypsum contribute to enhanced levels of natural radionuclides in the environment. The concentration of naturally occurring radionuclides in soil, rock phosphate and phosphogypsum samples collected around a phosphate fertilizer plant were determined. Also the external background gamma levels were surveyed. ^{238}U , ^{232}Th , ^{226}Ra and ^{40}K activities in soil samples were 21–674 Bq/kg, 11–44 Bq/kg, 22–683 Bq/kg and 51–295 Bq/kg respectively. The external background gamma radiation levels in the plant premises were ranging from 48 to 133 nGy/h.

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1. Introduction

Phosphate rocks of sedimentary origin contain Uranium (^{238}U), Thorium (^{232}Th) and its decay products in addition to phosphate minerals (Roessler, 1990). Considerable variations are found in the chemical composition of rock phosphate from different mining areas. In general, sedimentary phosphate rocks, or phosphorites, originated in a marine environment, are characterized by activity concentrations of uranium much higher than those of volcanic and biological rocks. Reported values of ^{238}U in rock phosphate range from 1.0 to 5.7 Bq/g (Barisic, Lulic, & Miletic, 1992; Guimond &

Hardin, 1989; Heijde, Klijn, & Passchier, 1988). These phosphates are largely used for the production of phosphoric acid, fertilizers and hence phosphate fertilizer industries are considered to be a potential source of natural radionuclide contamination. Their radioactivity leading to health problems from radiation at the level of the industrial processes which involves mining and transportation of phosphate ores and production of fertilizers. At the usage level, when fertilizers dispersed into the geo and biospheres, have a potential to transfer to living beings. Leaching of the minerals and wastes is another potential source of radioactivity dissemination which may contribute to enhanced exposure of workers, public and the environment to these radionuclides.

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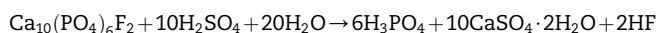
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Phosphogypsum is a waste by-product from the processing of phosphate rock by the “wet acid method” of fertilizer production, which currently accounts for over 90% of phosphoric acid production. The wet process is economic but generates a large amount of phosphogypsum (5 tons of phosphogypsum per ton of phosphoric acid produced) (USEPA, 2002). World phosphogypsum production is variously estimated to be around 100–280 mega tonnes per year (Parreira, Kobayashi, & Silvestre, 2003; Yang, Liu, Zhang, & Xiao, 2009). The nature and characteristics of the resulting phosphogypsum are strongly influenced by the phosphate ore composition and quality. Wet processing causes the selective separation and concentration of naturally occurring radium (Ra), uranium (U) and thorium (Th): about 80–90% of ^{226}Ra is concentrated in phosphogypsum while nearly 86% of U and 70% of Th end up in the phosphoric acid. Determining the types of impurities present can be very important when defining waste management processes and environmental policies. The discharge of phosphogypsum (PG) on earth surface deposits is a potential source of enhanced natural radiation and heavy metals, and the resulting environmental impact should be considered carefully to ensure safety and compliance with environmental regulations (Bolivar, Garcia-Tenorio, & Vaca, 2000; Shakhshiro et al., 2011).

Industrial processing of rock phosphate to manufacture phosphatic fertilizers involves the production of phosphoric acid according to the following chemical reaction (Haridasan, Paul, & Desai, 2001):



Only 15% of world phosphogypsum production is recycled as building materials, agricultural fertilizers or soil stabilization amendments and as a set controller in the manufacture of Portland cement. The remaining 85% is disposed of without any treatment. This byproduct is usually dumped in large stockpiles exposed to weathering processes, occupying considerable land areas and causing serious environmental damage (chemical and radioactive contamination), particularly in coastal regions. The radionuclide ^{226}Ra produces radon gas (^{222}Rn), which has a short half-life of 3.8 days, an intense radiation capacity, and causes significant damage to internal organs (USEPA, 2002). For this reason the USEPA has classified phosphogypsum and rock phosphate as “Technologically Enhanced Naturally Occurring Radioactive Materials” (TENORM) (USEPA, 2002) and phosphogypsum exceeding 370 Bq/kg of radioactivity has been banned from all uses by the EPA since 1992. Depending on the quality of the rock source, phosphogypsum can contain as much as 60 times the levels normally found prior to processing. The most important source of phosphogypsum radioactivity is reported to be ^{226}Ra (Rutherford, Dudas, & Samek, 1994). Due to the fact that this by-product accumulates a fraction of the natural radionuclides originally present in the minerals treated, it can be concluded that these industries through the phosphogypsum may produce a radioactive impact on their nearby environment (Bolivar, Garcia-Tenorio, & Garcia-Leon, 1995).

The storage of phosphogypsum in large land areas without any prior treatment can cause serious environmental contamination of soils, water and the atmosphere. Negative

atmospheric impacts can be caused by the erosion of phosphogypsum piles and the release of highly polluting substances, due to the presence of hazardous vapors containing heavy metals, sulphates, fluorosilicates, hydrogen fluorides, phosphorus, cadmium and ^{226}Ra (Marovic & Sencar, 1995). Atmospheric agents can transport the contamination to neighboring areas. Another matter of concern is the leachability of hazardous elements from phosphogypsum and thus the contamination of groundwater underlying phosphogypsum stacks (May & Sweeney, 1982, pp. 1–19, 1983, pp. 1–19). Since phosphogypsum waste is generally transported and disposed as aqueous slurry, phosphogypsum piles can be affected by tidal variations and dissolution/leaching of the elements naturally present in the phosphogypsum can occur. Dissolved elements may be deposited in nearby soils or transferred to waters and finally to living beings (Reijnders, 2007).

Hence, phosphogypsum disposal by piling the waste into open environment subjecting it to natural weathering processes without any treatment may lead to radioactive contamination. In this study, the distribution of ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K in soil was investigated in and around a phosphate fertilizer industry which stores and disposes its solid waste in the surrounding environment. The main goal is to evaluate the impact of such waste on soil and to identify the most influencing factors on contaminants distribution from a fertilizer plant producing about 25,000 MT phosphoric acid which results in an annual generation of more than 120,000 MT of phosphogypsum.

2. Materials and methods

2.1. Sample preparation

Soil samples were collected twice a month for a period of 3 months from 20 locations in and around a phosphate fertilizer plant (Fig. 1). Rock phosphate samples and gypsum were also collected from the location from their respective storage or disposal areas. Soil samples were crushed and cleaned to remove the clinker and other organic debris. The samples were dried for 24 h in an air-circulation oven at 110 °C. Samples were further ground, homogenized and about 100 g of each sample were filled in plastic containers of 6.5 cm diameter \times 7.5 cm height and sealed to make them airtight. The containers kept undisturbed for 6 months so that to provide the longest-lived intermediary radionuclide ^{234}Th (24.1 days) the opportunity to come to within 1% of secular equilibration with its parent ^{238}U . After attainment of secular equilibrium the samples were subjected to gamma-ray spectrometric analysis.

2.2. Gamma spectrometric analysis

The gamma-spectrometry system having an n-type coaxial HPGe detector (DSG, Germany) shielded with 7.5 cm thick lead having 100% relative efficiency and a resolution of 2.1 keV at 1.33 MeV gamma energy of ^{60}Co was used. The gamma spectra acquired for 60,000 s were analyzed using a 64K PC based multichannel analyzer and gamma spectrometry software.

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