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Assessment of natural radioactivity in fertilizers and phosphate ores in Egypt

Nabil M. Hassan [a,*], Nassif A. Mansour [a], Mohamed Fayez-Hassan [b], Ebrahim Sedqy [a,c]

[a] *Department of Physics, Faculty of Science, Zagazig University, Zagazig 44519, Egypt*
[b] *Department of Experimental Nuclear Physics, Nuclear Research Centre, EAEA, Cairo, Egypt*
[c] *Faculty of Engineering (Basic Science), Sinai University, El Arish, Egypt*

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Abstract

Radionuclide concentrations of uranium (^{238}U), thorium (^{232}Th) and potassium (^{40}K) maintained in 10 phosphate ore and 5 fertilizer samples were measured using a high-purity germanium detector. The concentrations of these radionuclides in the phosphate ore samples were relatively high for ^{226}Ra at 871.37 ± 91.90 Bq kg^{-1} and relatively low for ^{232}Th and ^{40}K at 19.21 ± 2.42 Bq kg^{-1} and 176.06 ± 17.66 Bq kg^{-1}, respectively. The concentrations of these radionuclides were low in the fertilizer samples. The radiological hazards of the radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal (H_{in}) indices and annual effective dose due to the presence of these radionuclides in the investigated samples were calculated. The released radon from the selected samples was measured using the Alpha Guard radon monitor. Subsequently, the radon emanation coefficient and its exhalation rate were calculated.

Keywords: Radionuclide; Radiological hazard; Radon; Alpha Guard; Phosphate ores; Fertilizers; Egypt

1. Introduction

The phosphate industry is considered as an important contributor to national economies for several countries. Phosphate rocks are extensively used as a source of phosphorus mainly for fertilizers, as well as for phosphoric acid, gypsum used in building materials and other specialty chemicals [1]. The mineralogical composition of a phosphate ore is dominated by fluorapatite [$Ca_{10}F_2(PO_4)_6{\cdot}CaCO_3$], goethite and quartz, with minor amounts of Al-phosphates, anatase, magnetite, monazite and barite [2,3]. Heavy metals and trace elements, such as cadmium (Cd) and nickel (Ni), are also detected in phosphate ores [4]. Moreover, several studies have shown that phosphate rocks maintain various amounts of naturally occurring radioactive materials (NORM), e.g., uranium, thorium, their decay products and potassium [5–7]. Therefore, mining and processing of phosphate ores redistribute these radionuclides among the various products, by-products and wastes of the phosphate industry. Effluent discharges into the environment, as

* Corresponding author. Tel.: +20 1069926033.
E-mail address: nmmh1976@Zu.edu.eg (N.M. Hassan).
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well as the use of phosphate fertilizers in agriculture and by-products, such as gypsum (phosphogypsum) in the building industry, are possible sources of radiation exposure to the public [6,7].

The most important sources of external and internal exposure are the gamma radiation and alpha particles emitted from the radionuclides of the uranium (^{238}U) series, thorium (^{232}Th) series and ^{40}K present within phosphate rocks. External exposure occurs directly by γ-rays, whereas internal exposure to α-particles arises from the inhalation of radon and its progenies. Consequently, the α-particle dose is delivered directly to the bronchial tissue, creating a potential for radiogenic lung cancer [8–10]. Therefore, radiation released from NORMs (phosphate ores and fertilizers) theoretically has a potential to cause cancers in individuals who are exposed at significant levels; thus, the monitoring of natural radioactivity in human surroundings is of significant importance from the viewpoint of radiation protection [11].

Radon (^{222}Rn with $T_{1/2}$ ~3.82 d) and its progenies (main source of internal exposure) are present in the environment due to the presence of radium that has decayed to radon, i.e., an radioactive gas. From the viewpoint of protection against indoor radon, it is important to understand the generation and the possible migration processes of radon and the factors that influence these processes [12–14]. When radon generated from radium decay in the solid grains of the material, a fraction of the radon escapes into the pore spaces among the solid grains. The ratio of the amount of radon that enters the pore spaces over the amount of radon generated is called the emanation coefficient [11,15]. The emanation coefficient is expected to increase with water content because an increase in the amount of water in the pore spaces increases the probability of capturing radon atoms in the pore spaces [9,11]. Before undergoing radioactive decay, some of the radon in the pore spaces migrates from the point of generation in the materials into the atmosphere; that is, the radon is exhaled from the surface of the materials. The exhalation rate is defined as the amount of activity of released radon per unit surface area per unit time [16–19]. A common classification of materials based on the potential risk of radon exposure can be established by evaluating the emanation coefficients and exhalation rates.

Recently, international awareness of NORM as a potential source of ionized radiation has increased significantly. Egypt as a country is now paying more attention to the radiation released from phosphate ores and fertilizers used in its markets and agriculture. Therefore, in the present work, concentrations of ^{238}U, ^{232}Th, and ^{40}K in commonly used phosphate ores and fertilizers in Egypt were measured using a high-purity germanium detector (HPGe). The concentration of radon released from these materials was measured using an Alpha Guard radon monitor. Consequently, the radon emanation coefficient and its exhalation rate were calculated. In addition, hazard indices of radium equivalent activity (Ra_{eq}), external (H_{ex}), internal (H_{in}) and annual effective dose associated with these radionuclides were calculated and compared with worldwide safety limits according to UNSCEAR equations.

2. Materials and methods

2.1. Sample preparation

Ten samples of phosphate ores were collected from different phosphate mines in Egypt (El Sebaea, Abu Tartor and Safaga). The common fertilizers used in Egypt were collected from different factories. The selected samples were crushed to fine powder forms and sieved through a 1 mm mesh size to remove the larger grain sizes and making them more homogenous. Then, the samples were dried in a temperature controlled furnace (oven) at 110 °C for 24 h to ensure that moisture was completely removed. After moisture removal, the samples were cooled in a desiccator prior to radionuclide and radon measurements.

2.2. Measurement of radionuclide concentrations with γ-ray spectroscopy

After the samples were prepared, they were packed in plastic containers, i.e., normal cylindrical plastic containers (6-cm diameter and 8 cm height) made from polyethylene. The containers were sealed using an adhesive to avoid any possibility of radon leakage. Each sample was stored in a sealed container for 30 days to achieve radioactive secular equilibrium between ^{238}U and its daughters. An empty container with the same geometry was sealed and left for 30 days to measure the background. Because radium (^{226}Ra) and its progeny produce 98.5% of the radiological effects of the uranium series, the contributions of ^{238}U and the precursors of ^{226}Ra are normally ignored. Therefore, the reference of ^{238}U series radionuclides is often written as ^{226}Ra instead of ^{238}U [20].

Radionuclide concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were measured using a vertical closed-end coaxial HPGe detector manufactured by ORTEC (model: GMX-70230 EG&G) with an active area of 190 cm^2, a measured efficiency of 70% and an energy resolution of 2.3 keV at 1332.5 keV (located at the Egyptian