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Influence of grain size on radionuclide activity concentrations and radiological hazard of building material samples

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HIGHLIGHTS

- Measurement of naturally occurring radionuclides in different grain sizes for some building material samples.
- We determined radium equivalent activity, the representative level index, and annual absorbed dose rate for all samples.
- The radionuclide activity concentrations and radiological hazard depends on the grain size.

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ABSTRACT

The knowledge of radioactivity content in various radionuclides in building materials plays an important role in health physics; therefore, we measured the amount of naturally occurring radionuclides in building material (sand, granite, marble, and limestone) samples of different grain sizes by using NaI (Tl) and MCA1024 gamma-ray spectrometers. Data analyses were performed to determine ^{226}Ra, ^{232}Th, and $^{4\circ}$K activity concentrations. The results revealed an inverse relationship between activity concentration and grain size of the samples. The radium equivalent activity (Ra_{eq}), representative level index I, and annual absorbed dose rate were calculated.

1. Introduction

Natural radioactivity is ubiquitous in the earth's environment, including several geological formations such as soils and rocks, plants, water, air, and building materials (Ramassay et al., 2004; Rati et al., 2010; Kayakökü et al., 2016). Building materials are obtained from rocks and soils. The natural radioactivity in building materials is mainly because of uranium (^{238}U) and thorium (^{232}Th) series and the radioactive isotope of potassium ($^{4\circ}$K). All these can be sources of both internal and external radiation exposure. Internal exposure occurs through inhalation of radon gas, and external exposure occurs through the emission of penetrating gamma-rays Shahbazi-Gahrouei (2003). Radon and Thoron and their progeny are the major sources of radiation exposures (Charles, 2001).

Buildings are very important in human life because people spend most of their time (about 80%) indoors (home or office) (Beretka and Mathew, 1985). In addition to terrestrial and cosmic radiations in the atmosphere, indoor gamma radiation mainly originates from building materials. Therefore, measuring the radioactivity of building materials will allow us to assess any possible radiological hazards to human health and improve the standards and guidelines for use and management (Stoulos et al., 2003; Mavi and Akkurt, 2010).

Granite contains relatively high concentrations of naturally occurring radioactive materials from the uranium and thorium decay series (^{238}U and ^{232}Th). Sand, granite, marble, and limestone samples are very common in Egypt. Because of the use of building materials that contain uranium, workers and the environment are exposed to enhanced doses of radiations. During handling, packing, and transportation of these materials, some workers could be exposed to additional external gamma-rays. Therefore, construction workers may be exposed to high intensity radiation from building materials of different grain sizes, and this could negatively affect their respiratory tract. When they are exposed for a long time, it could cause an inflammation. Therefore, the abovementioned building material samples were selected for the present study.

The radioactivity of the samples depends on the grain size. Several works have been performed to explain the reason why the radionuclide activity concentrations increase when the particle size decreases. Some differences can be observed depending on which type of samples are investigated (Megumi and Mamuro, 1977; Kalkwarf et al., 1985; Raj Kumari et al., 2015); therefore, by knowing ^{226}Ra, ^{232}Th, and $^{4\circ}$K activity concentrations for a particular grain size, a choice can be made to select the building material of a grain size that has minimum radiation risks. Therefore, sand, granite, marble, and limestone samples were

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analyzed as these are used as building materials world. This work aims to study the effect of grain size on radionuclide concentrations in some Egyptian building materials used in the construction of walls and floors using gamma-ray spectrometry and to estimate the radium equivalent activities (Ra_{eq}), which are related to the external gamma dose rates.

2. Experimental techniques

2.1. Sampling and sample preparation

For radioactivity measurement, samples of commonly used building materials, namely sand, granite, marble, and limestone were collected randomly from Qena and Aswan governorate, which are located in the southern part of Egypt, and tested for their natural radioactivity content and studying the effect of grain size on the radioactivity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K. Sand was collected from Qena, and other samples were collected from Aswan.

Some of the samples were air-dried at room temperature in open air to ensure that any residual moisture present in the samples was removed. The samples were placed in an oven for 48 h at 105 °C. Granite, marble, and limestone samples were initially broken into granules using a manual hammer. These granules with large grain size were then crushed into small pieces using a ball-milling (FRITSCH-Germany). The samples were sieved by using an electrical sieve (FRITSCH-Germany) of different sizes, 2000, 1800, 1000, 560, 425, 200, and 80 µm to separate the samples into seven categories.

The samples were hermetically sealed in a cylindrical plastic bottle (250 ml), and then each sample was placed in a container that provides a reproducible geometry. The container was closed to avoid the leakage of Rn-222 and Rn-220 from the samples and left for four weeks to reach equilibrium among Ra-226, Th-232 and their daughter products before radiometric analysis (Olszewska-Wasiolek, 1995).

2.2. X-ray fluorescence

X-Ray Fluorescence is an analytical technique that uses the interaction of X-rays with a material to determine the elemental composition of the material. In this study, we analyzed the samples by using JEOL JSX 3222 Element Analyzer equipped with an energy- dispersive X- ray fluorescence system (JEOL, Japan).

2.3. Gamma spectrometric measurements

Radionuclide activity concentrations (^{226}Ra, ^{232}Th, and ^{40}K) in the building material samples were determined using the NaI (Tl) detector. The detector mainly consists of 3 × 3 in. NaI (Tl), S-1212-I model, and a 1024 microcomputer multichannel analyzer, 5510 Ortec-Norland. The detector has a peak gamma-ray efficiency of 2.3×10^{-2} at 1332 keV, energy resolution of 7.5% at 662 keV, and operation bias voltage of 805 V dc. The detector was placed inside a massive cylindrical lead shield has quarter 25 cm to decrease the background radiation. The detector was connected to preamplifier, main amplifier, analog to digital converter, and multichannel analyzer. The system was calibrated for energy using standard point sources (^{60}Co, ^{137}Cs) and for efficiency using standard QCYB41. To prepare a standard sand sample, first, a plastic container (250 ml) was filled with sand. A well-defined amount of the standard solution QCYB41 (2 ml) was added to sand sample. This was then heated to 90 °C until the mixture had evaporated to near dryness. The drying was completed in an air oven at 80 °C (Harb, 2008). The standard solution used was obtained from Physikalisch Technische Bundesanstalt, Germany.

Every sample was positioned parallel to the detector for 10–24 h for ^{226}Ra, ^{232}Th, and ^{40}K concentrations measurement. The resultant spectrum of each sample was acquired using the Genie 2000 software package. In general background was counted normally every week prior to sampling under the same condition of sample measurement. Activity concentration of ^{40}K was directly measured from its own gamma-ray at 1460.8 keV, while activity concentration of ^{226}Ra was measured using gamma-lines at 351.92 keV (35.1%) of ^{214}Pb and at 609.32 keV (44.6%) of ^{214}Bi. ^{232}Th activity concentration was determined using gamma-energies at 911.16 keV (26.6%) of ^{228}Ac and at 2614 keV (35.8%) of ^{208}Tl.

Table 1
Weighted mean values of the activity concentration of the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in sand, granite, marble, and limestone samples of different grain sizes.

Grain sizes (µm)	Activity concentration (Bq/kg)		
	^{226}Ra	^{232}Th	^{40}K
Sand sample			
2000–1800	42.27 ± 2	14.88 ± 1	615.41 ± 53
1800–1000	47.44 ± 2	21.64 ± 1	648.28 ± 56
1000–560	52.15 ± 2	21.99 ± 1	685.79 ± 59
560–425	53.75 ± 2	26.99 ± 2	733.1 ± 63
425–200	59.86 ± 3	29.25 ± 2	741.78 ± 64
200–80	65.28 ± 3	47.16 ± 3	805.81 ± 69
< 80	66.8 ± 3	50.98 ± 3	849.8 ± 73
Granite sample			
2000–1800	156.67 ± 7	333.85 ± 20	5769.22 ± 496
1800–1000	235.85 ± 10	482.34 ± 29	5932.01 ± 510
1000–560	246.77 ± 11	509.59 ± 51	5996.64 ± 516
560–425	259.79 ± 11	515.46 ± 45	6348.8 ± 546
425–200	267.36 ± 12	655.91 ± 40	6779.79 ± 583
200–80	341.39 ± 15	724.73 ± 44	6869.34 ± 591
< 80	372.45 ± 16	746.84 ± 45	6880.03 ± 592
Marble sample			
2000–1800	52.82 ± 2	16.84 ± 1	152.91 ± 13
1800–1000	64.45 ± 3	20.83 ± 1	216.16 ± 19
1000–560	67.02 ± 3	20.85 ± 1	222.26 ± 19
560–425	68.07 ± 3	25.99 ± 2	250.05 ± 22
425–200	73.03 ± 3	36.13 ± 2	268.92 ± 23
200–80	81.49 ± 4	39.82 ± 2	274.2 ± 24
< 80	83.7 ± 4	42.05 ± 3	315.88 ± 27
Limestone sample			
2000–1800	33.98 ± 1	2.72 ± 0.18	186.82 ± 16
1800–1000	36.4 ± 2	4.44 ± 0.27	218.93 ± 19
1000–560	44.35 ± 2	5.17 ± 0.31	258.59 ± 22
560–425	46.53 ± 2	6.52 ± 0.4	313.56 ± 27
425–200	47.97 ± 2	10.13 ± 0.62	371.05 ± 32
200–80	49.72 ± 2	11.82 ± 0.73	385.22 ± 33
< 80	52.05 ± 2	13.15 ± 0.83	443.93 ± 38

2.4. Calculation of radium equivalent activity and hazard index

Radium equivalent dose rate (Ra_{eq}), external hazard index, internal hazard index, and representative level index were calculated using the following formulas (Beretka and Mathew, 1985; Krieger, 1981; Beretka and Mathew, 1985; NEA-OECD, 1979).

$$Ra_{eq} = A_R + (A_{Th} \times 1.43) + (A_K \times 0.077), \tag{1}$$

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_K/4810), \tag{2}$$

$$H_{in} = (A_{Ra}/185) + (A_{Th}/259) + (A_K/4810), \tag{3}$$

$$I = (A_{Ra}/150) + (A_{Th}/100) + (A_K/1500), \tag{4}$$

where A_{Ra}, A_{Th}, and A_K are the activity concentrations of Ra, Th, and K in Bq/kg, respectively. Ra_{eq} equation is based on that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th, or 4810 Bq/kg of ^{40}K produce the same gamma dose rate. For safe building materials inside the house and factories, the acceptable value for H_{ex} is less than unity (Beretka and Mathew, 1985).

2.5. Calculation of absorbed dose rate and annual effective dose equivalent

The gamma dose rate (D) in the outdoor air at 1 m above the ground