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Gamma-ray measurements of natural radioactivity in sedimentary rocks from Egypt

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Abstract The aim of this study was to measure concentrations and distributions of natural radionuclides occurring in rocks. The activity concentrations ($Bq \cdot kg^{-1}$) of the naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in sedimentary rock samples from Eastern Desert (Um El-Huetat), Nile Valley (Gebel Owina) and from southwest Sinai (Wadi Ghweiba) were measured using a high-purity germanium detector. The samples under investigation (clay, shale and sandstone) were used as raw materials in the construction industry (bricks, ceramics, cement, fillers, etc.). Though the sediments of Egypt have already been investigated in the geological and mineralogical aspects, it is necessary to investigate the natural radioactivity in order to complete their classification. The average concentration values of ^{226}Ra, ^{232}Th, ^{40}K in the surveyed samples were 47 ± 7 , 21± 5, 393±19 $Bq.kg^{-1}$ (clay); 23 ± 5, 30 ± 6, 563 ± 24 $Bq.kg^{-1}$ (shale); and 17 ± 4, 14 ± 4, 299 ± 17 $Bq.kg^{-1}$ (sandstone), respectively. All sediment samples have radium equivalent activities ranging from 55 to 115 $Bq \cdot kg^{-1}$, lower than the limit set in the OECD Report (370 $Bq \cdot kg^{-1}$). The overall mean outdoor terrestrial gamma dose rates fluctuate from 28 to 55 $nGy \cdot h^{-1}$. The external gamma radiation dose due to natural radionuclides present in the samples have been computed and compared with the global averages. In terms of the radiation safety, the natural radioactivity of the sediment in Egypt is below the recommended limits of the gamma dose rate. Therefore, they can be used for all kinds of public buildings.

Key words Natural radionuclides, Shale, Clay , Sandstone, Radiation hazard, Egypt.

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1 Introduction

The natural radioactive elements are distributed everywhere in the biosphere with different concentrations. From the natural "risk" point of view, it is necessary to know the dose limits of public exposures. At the same time, it is necessary to measure the natural environmental radiation level provided by ground, air, water, food, building interiors, etc., for the estimation of the exposures to natural radiation sources. IAEA has published data for the doses accumulated by human beings during their life activities. The exposure to cosmic radiation is about 0.38 $mSv \cdot a^{-1}$, to terrestrial radiation 0.45 $mSv \cdot a^{-1}$ (this increases by nearly 20% for brick and concrete buildings), and to water, food and air 1.5 $mSv \cdot a^{-1}$. The exposure to X-rays diagnostics is about 0.4 $mSv \cdot a^{-1}$and to the other factors like color TV, air flights, and nuclear power plants, the exposure is about 0.1 $mSv \cdot a^{-1}$. Thus, in total human beings accumulate about 2.7 $mSv \cdot a^{-1}$ radiation from natural and man-made sources. The dose limit for exposure to man-made radiation sources is 1 $mSv \cdot a^{-1}$ [1] .

Terrestrial radionuclides are distributed throughout the crust of the earth. Outdoor exposures from sources of terrestrial radiation originate predominantly

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from approximately the top 1-ft layer of the soil [2]. Formations such as limestone and sandstone have the lowest radionuclide concentrations. Some shale formations, particularly those containing a significant fraction of organic matter, can possess higher concentrations of radionuclides. Overall, igneous and metamorphic rocks comprise approximately 90% of the planetary crust. Sedimentary rocks, however, tend to accumulate at the top of the crust. Approximately 75% of the surface of the earth is covered by sedimentary rock.

From the point of view of radioactive concentration, the "normal" clays, which were used as construction materials for the residential and public buildings, etc., contain some U and Th, while the content of K is less than 4%. In cases where the concentration of these radioactive elements in clays is higher than 1×10^{-5} for U, 2×10^{-5} for Th, and higher than 5% for K, it is necessary to review the possibilities of limiting the usage of these clays as construction materials [3]. The reason for this limitation comes from the high degree of the exposures that creates radioactivity for the human beings. In the building industry, the most-used raw materials are clays (production of bricks, tiles, ceramics, fillers, etc.). Therefore, we investigated samples from clay, sandstones and shales (for comparison) from different sites of Egypt.

Uranium mineralization is known in association with some carboniferous and cretaceous black shale, in phosphorite deposits and in Oligocene sandstones in many localities in Egypt [4]. The final report of radiometric survey carried out in the Eastern Desert of Egypt (1984) reported isolated small airborne anomaly located in the plain of Wadi Ghweiba. This anomaly attracts the attention because Wadi Ghweiba is located in the zone defined to be one of the most important national developmental projects of the northern part of Gulf of Suez. Therefore, this work aimed to investigate the extension of the radioactive anomalies in Wadi Ghweiba area and to delineate the source rock types responsible for such anomalies.

The objective of the present study was to determine the specific radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in sedimentary rock samples collected from different areas, to assess any radiological hazard and to determine the gamma radiation dose rate and the annual effective dose from samples under investigation, which are used in buildings. The results obtained in the present study are also compared with the corresponding results for sediment of different origins given in the literature.

2 Experimental

The natural radioactivity due to radium, thorium and potassium contents of 43 samples of clay, shale and sandstone was measured. Twenty-three samples were collected from Wadi Ghweiba (southwest Sinai) and 20 samples from Um El-Huetat and Gebel Owina (Eastern Desert) and Nile Valley in Egypt. The collected samples were weighed individually, air-dried for 4 days, pulverized, homogenized, and sieved to pass through a 200-mm mesh. The powdered sediment samples were transferred to an especial disc (50 g) for gamma-activity analysis. Each sample was weighed and carefully sealed for 4 weeks to reach secular equilibrium between ^{226}Ra and ^{232}Th and their respective progeny. The activity of ^{214}Bi and ^{214}Pb in equilibrium with their parents was assumed to represent the ^{238}U activity, while the activity of ^{228}Ac and ^{208}Tl was assumed to represent the ^{232}Th activity. The detector (HPGe) has a photo-peak relative efficiency of about 23% and an energy resolution of 1.95 keV (FWHM) for the 1.332 MeV gamma transition of ^{60}Co. To reduce gamma-ray background, a lead chamber shield with a fixed bottom and a moving cover shielded the detector. The lead shield contained two inner concentric layers of copper and cadmium.

In order to determine the background distribution due to naturally occurring radionuclides in the environment around the detector, an empty polystyrene container was counted in the same manner as the samples. After measurement and subtraction of the background, the activity concentration was calculated. The accumulation time for gamma-ray spectra measurements ranged from 12 to 20 h depending upon the activity levels present in the samples.

3 Results and discussion

The average concentration values ($Bq\cdot kg^{-1}$) of ^{226}Ra, ^{232}Th and ^{40}K in the samples measured collected from different locations are given in Table 1. From the

obtained results from Wadi Ghweiba area, it can be seen that the ^{226}Ra activity concentration for clay is in the range of 17 ± 4 to 50 ±6 Bq·kg^{-1} with an average value of 36 ± 6 Bq·kg^{-1} and the activity concentration of ^{232}Th varied from 19 ± 4 to 28 ± 5 Bq·kg^{-1} with an average of 23 ± 5 Bq·kg^{-1}, while the activity concentration of ^{40}K varied from 372 ±17 to 401 ± 22 Bq·kg^{-1} with an average value of 389 ±19 Bq·kg^{-1}. With respect to shale and sandstone, the activities of ^{226}Ra range from 17± 3 to 27 ± 5 and from 16 ± 3 to 24 ± 5 with the average values of 21 ± 5 and 19 ± 4 Bq·kg^{-1}, respectively, ^{232}Th values were in the range of 21±4 to 34 ±6 and 14 ± 3 to 25 ±4 with the average values of 28 ± 5 and 18 ± 4 Bq·kg^{-1}, while for ^{40}K the average values were 561 ± 24 and 278 ± 17 Bq·kg^{-1}, respectively.

Table 1 Mean activities (^{226}Ra, ^{232}Th and ^{40}K) and radium equivalent Ra_{eq} (Bq·kg^{-1}) in the samples collected from different locations in Egypt

Location	Description	^{226}Ra / Bq·kg^{-1}	^{232}Th / Bq·kg^{-1}	^{40}K / Bq·kg^{-1}	Ra_{eq} / Bq·kg^{-1}
Southwest Sinai (Wadi Ghweiba)	Clay	36 ± 6	23 ± 5	389 ±19	96
	Shale	21 ± 5	28 ± 5	561 ± 24	100
	Sandstone	19 ± 4	18 ± 4	278 ± 17	64
Eastern Desert (Um El-Huetat)	Clay	47 ± 7	21 ± 5	401 ± 21	105
	Shale	25 ± 5	31 ± 6	549 ± 23	108
	Sandstone	15 ± 4	13 ± 4	302 ± 17	55
Nile Valley (Gebal Owina)	Clay	59 ± 8	20 ± 4	390 ± 19	115
	Shale	24 ± 5	30 ± 5	579 ± 24	107
	Sandstone	17 ± 4	11 ± 3	318 ± 18	55

It can also be seen that the highest value of ^{226}Ra is present in clay, while the ^{40}K was found in shale. The minimum values of ^{226}Ra and ^{232}Th are found in sandstone samples and the minimum percentage value of ^{40}K is present in clay samples, which indicate that values of these isotopes depend on the geochemical properties of these samples and their locations.

For clay from Um El-Huetat the values of ^{226}Ra, ^{232}Th and ^{40}K change from 31 ±6 to 59 ±9 Bq·kg^{-1}, from 17.6 ±4 to 29 ± 5 Bq·kg^{-1} and from 391 ±17 to 428 ±22 Bq·kg^{-1}, respectively. The corresponding values of shale samples are from 19 ±4 to 29 ±5 Bq·kg^{-1}, 21.8 ±4 to 38 ±6 Bq·kg^{-1}and 501 ±21 to 598 ±24 Bq·kg^{-1}, respectively. Sandstone samples have values ranging from 11±3 to 19±4 Bq·kg^{-1}, from 9±2 to 16±4 Bq·kg^{-1} and from 289±13 to 321±17 Bq·kg^{-1}, respectively. The mean values of above isotopes are 47±7, 21±5, and 401±21 Bq·kg^{-1}; 25±5, 31±6, and 549±23 Bq·kg^{-1}; and 15±4, 13±4, and 302±17 Bq·kg^{-1} for clay, shale and sandstone, respectively.

From the analytical results of Gebal Owina it can be seen that the ^{226}Ra activity concentration in clay is in the range of 31 ± 5 to 64 ± 8 Bq·kg^{-1} with an average value of 59 ±8 Bq·kg^{-1}. The activity of ^{232}Th varied from 14 ± 3 to 26 ± 4 Bq·kg^{-1} with an average of 20 ± 4 Bq·kg^{-1}, while the activity of ^{40}K varied from 376 ± 18 to 413 ±22 Bq·kg^{-1} with an average value of 390 ±19 Bq·kg^{-1}. In shale**,** the average values of ^{226}Ra, ^{232}Th and ^{40}K are 24 ± 5, 30 ±5 and 579±24 Bq·kg^{-1}, respectively. With respect to sandstone, the activity of ^{226}Ra ranges from 13 ± 3 to 24 ± 4 Bq·kg^{-1} with a mean value of 17 ± 4 Bq·kg^{-1}, while for ^{232} Th and ^{40}K the mean values are 11 ± 3 and 318±18 Bq·kg^{-1}, respectively. The average activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K for all rock samples under investigation (clay, shale, sandstone) are shown in Fig.1.

It is evident from the results that the minimum values of ^{226}Ra, ^{232}Th and ^{40}K are found in sandstone, while the maximum ^{226}Ra is present in clay but for ^{232}Th and ^{40}K are present in shale. Also, both radium and thorium have low values compared to that of potassium. Th/U ratio ranges from 0.8 to 1.3. Shale generally has higher thorium contents than sandstone and ferruginous sandstone, which may be related to the effects of adsorption capacity of clay minerals in shale with concentrated thorium. The high Th/U ratio indicates that part of uranium content is mobilized from these rocks and incorporated to the stream sediments. Analyses of five fine-grained (clay fractions) stream sediment samples at Eastern Desert show high ura-

nium content (59 Bq·kg^{-1}) and low Th content (20 Bq·kg^{-1}). The Th/U ratio in the stream sediment sample is 0.34, indicating that U is incorporated to the stream sediments and gets adsorbed on the clay fractions due to its high mobility.

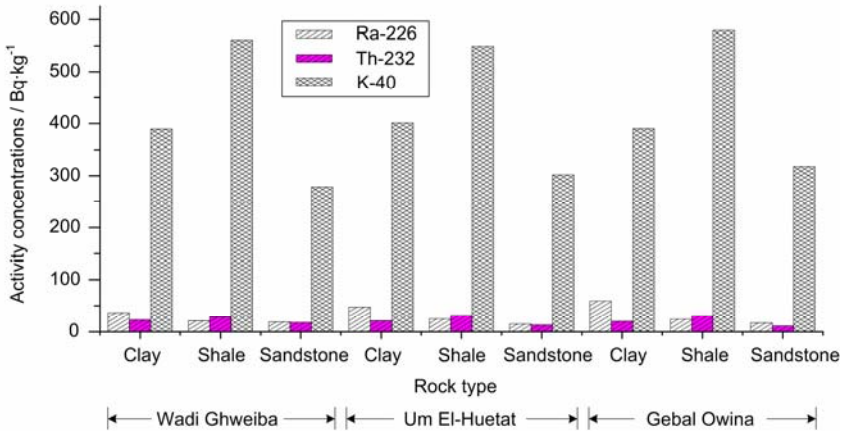

Fig.1 Activity concentration of ^{226}Ra, ^{232}Th and ^{40}K (Bq·kg^{-1}) for sedimentary rock samples.

Clay minerals are mainly composed of plate-like secondary aluminum silicate with small particle size and have a negatively charged surface. Therefore, clay particles have the ability to absorb cations on their surface. That is why the highest radionuclide activity concentrations occur in clay soils and the lowest occur in sandy soil [5,6].

These variations and the large spread in the data are a reflection of different geological regions of the samples. The concentrations of natural radionuclides in all measured samples were below the world averages for building materials, i.e. 50, 50 and 500 Bq·kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively [7,8] . The results of the present work indicate that the area under investigation has a normal level of natural background, so these materials do not pose a significant radiological hazard when used for construction of buildings.

From the obtained data, it is evident that the mean value of ^{40}K in all samples that were measured was found to be higher when compared with ^{226}Ra and ^{232}Th. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in all sediment samples under test are lower than the most published data.

Table 2 compares the reported values of natural radionuclides in the sediment samples, obtained in other studies [9], with those determined in the present study. On comparison, it is found that ^{226}Ra is in the same range, but both ^{232}Th and ^{40}K are always less than those of other countries' samples.

Table 2 Comparison of natural radioactivity ^{226}Ra, ^{232}Th and ^{40}K (Bq·kg^{-1}) in sediment with those in other countries.

Rock type	^{226}Ra	^{232}Th	^{40}K	Ref.
	17	14	299	Present work
Sandstone	26.3	24	325	[9]
	—	40	700	[10]
	23	30	563	Present work
Shale	39.9	41	814	[9]
	—	70	1100	[10]
	47	21	393	Present work
	41	27	410	[11]
Clay	38.3	44.7	635	[12]
	51.7	65.3	747	[13]
	43.2	53.7	631	[14]

It is important to point out that these values were not the representative values for the countries mentioned but for the regions from where the samples were collected.

Distribution of ^{226}Ra ,^{232}Th and ^{40}K in sediment is not uniform, so that with respect to exposure to radiation, the radioactivity has been defined in terms of radium equivalent activity (Ra_{eq}) in Bq.kg^{-1} to compare the specific activity of materials containing different amounts of ^{226}Ra ,^{232}Th and ^{40}K (Table 3). It is calculated through the following relation [15]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.07A_K \quad (1)$$

where A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K (in Bq·kg^{-1}), respectively. While defining Ra_{eq} activity according to Eq. (1), it has been assumed that 370 Bq·kg^{-1} of ^{226}Ra or 259 Bq·kg^{-1} of ^{232}Th or 4810 Bq·kg^{-1} of ^{40}K produce the same gamma dose rate.

Estimation of radioactive equilibrium between uranium and thorium and their daughters in the estimation of their concentration may be in error. If the estimation is not valid in some situation, the external gamma dose estimation is not likely to be in error since in the case of uranium series nearly 90% of the total gamma dose is given by ^{226}Ra and its daughters, and in the case of thorium series the percentage contribution of ^{228}Ac and its daughters are in the same range.

For calculating the absorbed dose rate in air at a height of 1.0 m above the ground from measured radionuclide concentrations in environmental materials the following formula was applied [16]:

$$D = 0.427A_{Ra} + 0.662A_{Th} + 0.043A_K \quad (2)$$

The external hazard index H_{ex} can be calculated by the following equation [8,16]:

$$H_{ex} = 0.0027A_{Ra} + 0.0038\,A_{Th} + 0.0002A_K \leq 1 \quad (3)$$

The absorbed dose rate is found to vary from 28 to 55 nGy·h^{-1}. It is observed that the value is lower than the global average value (55 nGy·h^{-1})[16]. The calculated external hazard is less than unity (0.15 to 0.31 (mSv)) as listed in Table 3.

Table 3 Air absorbed dose and external gamma radiation dose in sediment samples

Location	Rock type	Total air absorbed dose rate / nGy·h^{-1}	External γ-radiation dose / mSv
Southwest Sinai (Wadi Ghweiba)	Clay	47.3	0.26
	Shale	51.6	0.28
	Sandstone	32.0	0.18
Eastern Desert (Um El-Huetat)	Clay	51.2	0.29
	Shale	54.8	0.30
	Sandstone	28.0	0.15
Nile Valley (Gebal Owina)	Clay	55.2	0.31
	Shale	55.0	0.29
	Sandstone	28.2	0.15

4 Conclusion

The equivalent uranium and thorium obtained from sediment samples could be considered possessing low radioactive level. The result of ^{226}Ra, ^{232}Th and ^{40}K show that the concentration values of specific activities of samples from the areas under study are all at normal level with average values of 47 ± 7, 21 ± 5, 393 ±19 Bq·kg^{-1} (clay); 23 ± 5, 30 ± 6, 563 ± 24 Bq·kg^{-1} (shale) and 17 ± 4, 14 ± 4, 299 ± 17 Bq·kg^{-1} (sandstone) Bq·kg^{-1}, respectively. The gamma absorbed dose rates in air of rocks in areas under study are comparable to the average global terrestrial radiation of 55 nGy·h^{-1}. For sediment samples the values of absorbed dose rates fluctuate from 28 to 55 nGy·h^{-1}, with a mean value of 40.5 nGy·h^{-1}. The average radiation hazard parameters for all samples under investigation are lower than the acceptable value and also less than all the published data. The calculations of dose rate and external hazard index indicate that there is no high exposure for either inhabitants or workers dealing with transportation of sediment and there is a good safety index for all building materials. For all sediment samples under investigation, the radium equivalent values are lower than the acceptable value 370 Bq.kg^{-1} and the level index in all samples is less than unity, which is in a good agreement with other published results.

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