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Article in Journal of Physics Conference Series · August 2022

DOI: 10.1088/1742-6596/2305/1/012033

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Radioactivity and Environmental Impacts of Granites from Um Ara, Southeastern Desert, Egypt

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Abstract.Granites from the Southeastern Desert of Egypt were investigated by using in situ γ -ray spectrometer. Consequently, estimating the radiological effects from these rocks were achieved through calculation of some radiological parameters; Annual effective dose equivalent AEDE, Internal hazard index H_{in} , External hazard index H_{ex} , Alpha activity index I_{α} , Excess lifetime cancer risk, ELCR, Annual gonadal dose equivalent AGDE, Gamma activity concentration index I_{γ} and Radium equivalent activity Ra_{eq} . The geological nature of the region under investigation was affected by the radioactivity measurements of the rocks under study. This was observed by the high U, Th, and K values, which exceed the average world value for safety used as building materials. Therefore, the granites of Om Ara may be inappropriate to choose as a building material or as an indoor and outdoor decorative. At the same time, the region under study represents valuable economic minerals sources that can be used in several industrial applications.

Keywords: Natural radioactivity; Southeastern desert; Excess lifetime cancer risk.

1. Introduction

The rugged natural stones are a more significant building material because they are usually suitable for interior and exterior building and decorative materials. Granites shows high concentrations of thorium (Th), potassium (K) and uranium (U).The attention of these radionuclides in granitic rocks is intimately related to their mineral compositions and general petrologic features [1-3].

Since granite under study can be used as a building material, radiation exposure to the population can be increased appreciably by using materials with moderate radioactivity levels. Therefore, it is crucial to calculate the radiation hazards assessment.

This study's objective is to present an assessment for the radiological nature at Um Ara area located at the southeastern desert of Egypt, the other objective of this study is to map the radioactivity background in this area and using the data collected from this map to assess any later radioactivity fluctuation.

For that reason, this research deals with natural radioactivity the mineralogical and chemical study of granite to understand and correlate the relationship of natural radioactivity with the radioactive mineral content and estimate possible implications from radiation exposure.

2. Geological Setting

Um, Ara area is located in the south Eastern Desert of Egypt, and it is bounded between latitudes 22° 38'–22° 41' N and longitude 33° 47' – 33° 53". The geology of Um Ara area comprises Dokhan volcanics and metavolcanic .according to the petrographical background; these granites were classified as alkali-feldspar and monzogranite granite. The monzogranite generally has a homogeneous pinkish equigranular coarse-grained texture. [4-6]

Um, Ara granitic masses cover an area of about 300 km², characterized by moderate topography. The granitic rocks were classified as alkali feldspar and monzogranite granite. It also represented an excellent source of uranium in granitic rocks. [7, 8]

3. Materials and methods

A total number of 128 samples in Um Ara, Southeastern Desert, Egypt, were collected to measure their natural radioactivity levels according to ²³²Th (ppm), ⁴⁰K and ²³⁸U (ppm) (%). in situ γ -ray spectrometer, GS-256 with (NaI(Tl)) 3"×3" crystal detector is used to study rocks in the field radiometrically.

4. Results

4.1 Radioactivity analysis

Measurements of elemental concentrations for eU, eTh, and K were taken expressed in ppm (**Table 1**). From the table, we can notice that the elemental concentrations of eU, eTh, and K have ranged from 11 to 430 with an average value of 48.8 ppm, from 36.5 to 280 with an average value of 69 ppm, and from 2.3 to 7.5 with average value 4.9 %, respectively. The remobilization factor ranged from -7.6 to 350, with an average value of 29.1. The frequency distributions of activity concentrations of the studied radionuclides in the examined granite samples are shown in figure 3.1, suggesting high radionuclides contents of these granites.

From the obtained results, we can deduce that the average values of eTh, eU and K were higher than the reported Earth's crust values, 10.5 ppm, 2.6 ppm and 2.4 % respectively, and also higher than the recommended safety values for using in construction materials, 12.1 ppm, 3.9 ppm and 1.5 %, respectively [9].

In general, the high contents of radioelements in Um Ara granites indicated that it's advisable to monitor the radioactivity levels of granites before adopting them for use as a building material.

The Egyptian granites are main targets for uranium exploration in the Eastern Desert, which belong to the third epoch of uranium mineralization (0.70-0.50 Ga) during the upper Proterozoic-Cambrian times [10-13]. Um, Ara granites have a ²⁰⁶Pb/²³⁸U age of 603±14 Ma of zircons, which differs from the alkali feldspar micro granites [14]. Uranium deposits related to granitic nature can be divided into secondary and primary uranium deposits [15]. The first is formed in the rocks during magma crystallization. While the second is formed by later geological processes starting from dissolving and transportation of uranium. Then uranium can be divided into three types; (1) interstitial uranium at the grain boundaries, formed by hydrothermal solution migration and granites, (2) uranium in microfractures. and (3) absorbed uranium in chlorite, limonite and montmorillonite. The secondary uranium mineralization in Um Ara alkali feldspar microgranite formed at a two-stage of the metallogenic model as proposed by [16]; the first period was controlled by hydrothermal alteration accompanied by silicification, chlorination, K-feldspathization, and albitization .while the second period occurred near-surface, where water played an important role in moving the early, formed U-bearing minerals. The mineralogical composition of uranium deposits is dominated by uraninite and uranophane [17, 18] in Um Ara granite, attributed to the syngenetic hydrothermal origin. At the same time, accessories are represented by zircon, sphene, topaz and monazite. Uranium can be oxidized as U⁶⁺ in a near-surface area and is insoluble as a U⁴⁺ state.

4.2 Geochemistry of U and Th in the studied granites

The geochemistry of U and Th during magmatic differentiation has been studied in many types of granites from different areas. Generally, during magmatic differentiation, the Th/U ratio remains constant. [19] Suggesting 3.5 to 4 for the Th/U ratios in the granites. These ratios can be increased [20, 21] or decreased depending on the redox conditions, the volatile contents, or alterations by endogen or supergene solutions [22, 23]. Typical eTh/eU ratios in igneous rocks were 3:1 or 4:1. In contrast, the Th/U ratio of the studied granitic rocks was 2.2, indicating uranium enrichment. This was also confirmed by the average value of the remobilization parameter, which has a positive value of 29.1.

The relationships between ²³⁸U, ²³²Th, and ⁴⁰K (Table 1), were presented in figure 2. eU-eTh variation diagram of the studied granitic rocks reflected a strong positive correlation (R= 0.8), which indicated that magmatic processes played an essential role in the concentration of radioelements (Fig. a).

Thorium displayed a significant correlation with potassium ($R = 0.7$), and there was a positive correlation between ^{238}U and ^{40}K ($R = 0.5$), confirming the previous result (Fig. 1b & c).

The variation of eTh and eU against the eTh/eU ratio showed a negative correlation which has confirmed the idea that U and Th enrichment in these anomalies could be controlled by magmatic processes (Fig. 2d & e) [24,25].

Ratios of eTh/eU < 2 suggested relative uranium enrichment and implicates reducing conditions, but ratios >7 indicated preferential removal of uranium, possibly by leaching. The cross plot of eTh/eU and eTh/K ratios presented in figure 2f exhibit that altered granite samples lie in the leached-U sector.

The average value of eTh/K (14.3) is $>2 \times 10^{-4}$, which denoted thorium enrichment in the studied rocks. In general, low eTh/K ratios were an excellent indicator of potassium alterations [26].

Table (1):- Values of ^{238}U (ppm), ^{232}Th (ppm), and K (%), as well as eTh/eU and eTh/K ratios and (eU-eTh/3.5) parameter for Um Ara granite.

Rock	eU (ppm)	eTh (ppm)	eK (%)	eTh/eU	eTh/eK	eU-eTh/3.5
granite	29	46	2.9	1.6	15.9	15.9
	140.1	78.9	6.4	0.6	12.3	117.6
	41.5	51.2	2.8	1.2	18.3	26.9
	117.8	82.7	6.7	0.7	12.3	94.2
	21.6	51.3	2.5	2.4	20.5	6.9
	206.6	160.2	6.9	0.8	23.2	160.8
	22.2	46.4	2.8	2.1	16.6	8.9
	153.9	87.6	7.5	0.6	11.7	128.9
	34.3	56.4	4.3	1.6	13.1	18.2
	121.5	88.9	6.6	0.7	13.5	96.1
	40.1	46.6	3.3	1.2	14.1	26.8
	108.9	78.8	6.3	0.7	12.5	86.4
	16.4	46.8	3.2	2.9	14.6	3
	226.1	91.3	7.4	0.4	12.3	200
	38.3	47.8	3.2	1.2	14.9	24.6
	279.9	108.1	6.8	0.4	15.9	249
	23.4	41	3.6	1.8	11.4	11.7
	46.3	90.9	6.7	2	13.6	20.3
	22.5	36.5	3.2	1.6	11.4	12.1
	76.9	82.7	6.8	1.1	12.2	53.3
	23	45.1	2.3	2	19.6	10.1
	141.8	97.5	6.5	0.7	15	113.9
	22.8	42.1	2.3	1.8	18.3	10.8
	76.7	77	7.2	1	10.7	54.7
	28.2	50.8	4.4	1.8	11.6	13.7
	72.8	74.9	6	1	12.5	51.4
	17	50.1	3.8	2.9	13.2	2.7
	56.8	95.6	4.6	1.7	20.8	29.5
	24.9	56.6	4.1	2.3	13.8	8.7
	85.7	98.3	6.3	1.1	15.6	57.6
	17.9	48.5	4	2.7	12.1	4
	80.6	84.7	6.8	1.1	12.5	56.4

	24.2	50.9	4.3	2.1	11.8	9.7
	80.9	84.8	5.6	1	15.1	56.7
	36.4	40.8	3.5	1.1	11.7	24.7
	56.4	70.1	5.1	1.2	13.8	36.4
	33.5	44.2	3.1	1.3	14.3	20.9
	52.3	55.6	5.1	1.1	10.9	36.4
	21	40.5	3.2	1.9	12.7	9.4
	58	80.1	6.3	1.4	12.7	35.1
	11	65	4.8	5.9	13.5	-7.6
	22.5	76.6	6.1	3.4	12.6	0.6
	16.5	54.7	4.4	3.3	12.4	0.9
	32.6	76.5	5.5	2.3	13.9	10.7
	12.3	54.2	4.4	4.4	12.3	-3.2
	32.4	75.6	6	2.3	12.6	10.8

Table continues

granite	14.3	44.1	3.4	3.1	12.9	1.7
	60.5	72.3	6.3	1.2	11.5	39.8
	20.4	59.3	4.2	2.9	14.1	3.5
	46.2	88.4	5.6	1.9	15.8	20.9
	18.9	41.8	3.7	2.2	11.3	7
	31.9	81.9	6.5	2.6	12.6	8.5
	13	54.6	4.1	4.2	13.3	-2.6
	30.1	83.1	5.7	2.8	14.6	6.4
	16	49.1	4.3	3.1	11.4	2
	70.4	77.8	6.8	1.1	11.4	48.2
	20.2	58.2	4	2.9	14.6	3.6
	35.7	76.9	5.7	2.2	13.5	13.7
	17.1	50	3.6	2.9	13.9	2.8
	30.1	65.9	5.3	2.2	12.4	11.3
	21.3	52.1	3.7	2.4	14.1	6.4
	42	83.1	6.3	2	13.2	18.3
	18.9	53.6	3.7	2.8	14.5	3.6
	33	92.4	5.8	2.8	15.9	6.6
	29	61	3.8	2.1	16.1	11.6
	126	77	5.6	0.6	13.8	104
	40	43	2.6	1.1	16.5	27.7
	70	72	4.8	1	15	49.4
	29	49	3.1	1.7	15.8	15
	430	280	6.7	0.7	41.8	350
	38	54	2.4	1.4	22.5	22.6
	140	91	5.5	0.7	16.6	114
	26	65.7	4.8	2.5	13.7	7.2
	31.6	77	6.6	2.4	11.7	9.6

	32.1	72.5	4.1	2.3	17.7	11.4
	55.8	85.6	5.8	1.5	14.8	31.3
	24.4	58.7	4.1	2.4	14.3	7.6
	56.9	85.1	5.5	1.5	15.5	32.6
	30.1	51.9	4.1	1.7	12.7	15.3
	50.9	76.5	5.9	1.5	12.9	29
	12	59.1	3.4	4.9	17.4	-4.9
	81.9	91.2	6.2	1.1	14.7	55.8
	18	50.1	4.2	2.8	11.9	3.7
	24	70.2	5.5	2.9	12.8	3.9
	11.1	50.1	4.1	4.5	12.2	-3.2
	29.6	72.4	5.6	2.4	12.9	8.9
	18.5	51.3	4.1	2.8	12.5	3.8
	27.8	83.7	6.1	3	13.7	3.9
	14.8	59.3	4	4	14.8	-2.1
	47.9	97.6	6.3	2	15.5	20
	19.4	60.7	4	3.1	15.2	2.1
	33.1	89.9	5.8	2.7	15.5	7.4
	16.9	65.6	4.3	3.9	15.3	-1.8
	38.1	97.3	6.5	2.6	14.9	10.3
	16.3	58.9	4.5	3.6	13.1	-0.5

Table continues

granite	41.6	82.4	5.8	2	14.2	18.1
	18.9	50.4	3.5	2.7	14.4	4.5
	27.3	59.6	5.1	2.2	11.7	10.3
	17.2	51.2	3.5	3	14.6	2.6
	30.2	67.3	5.6	2.2	12	11
	20.6	42.3	3.6	2.1	11.8	8.5
	99.1	90.2	6	0.9	15	73.3
	17.7	44.8	3.5	2.5	12.8	4.9
	89.6	84.2	7.1	0.9	11.9	65.5
	35.2	42.3	4.2	1.2	10.1	23.1
	53.6	61.5	5.8	1.1	10.6	36
	53	51.8	2.9	1	17.9	38.2
	70.2	66	4.8	0.9	13.8	51.3
	30.1	50.1	3.2	1.7	15.7	15.8
	78.1	67.9	5.6	0.9	12.1	58.7
	25.7	51.6	3.5	2	14.7	11
	79.9	88.7	6.2	1.1	14.3	54.6
	18.4	53.9	4.4	2.9	12.3	3
	40.1	71.2	5.7	1.8	12.5	19.8

	16.1	66.4	3.8	4.1	17.5	-2.9
	33.2	76.6	6.3	2.3	12.2	11.3
	15	52.7	3.5	3.5	15.1	-0.1
	74.4	77	6.3	1	12.2	52.4
	13.9	53.6	3.4	3.9	15.8	-1.4
	32.5	80.1	6	2.5	13.4	9.6
	13.5	73.8	4.4	5.5	16.8	-7.6
	24.7	87.2	6.4	3.5	13.6	-0.2
	12.5	55	4.2	4.4	13.1	-3.2
	24.7	80.2	5.6	3.2	14.3	1.8
	13.6	57.5	3.2	4.2	17.9	-2.8
	26.7	90	6.1	3.4	14.8	1
	12.5	65.4	4.1	5.2	15.9	-6.2
	22.5	79.8	5.8	3.5	13.8	-0.3
Aver.	48.8	69	4.9	2.2	14.3	29.1
Crust	2.9	10.8	2.7			
Safety for building	4	12.3	1.6			

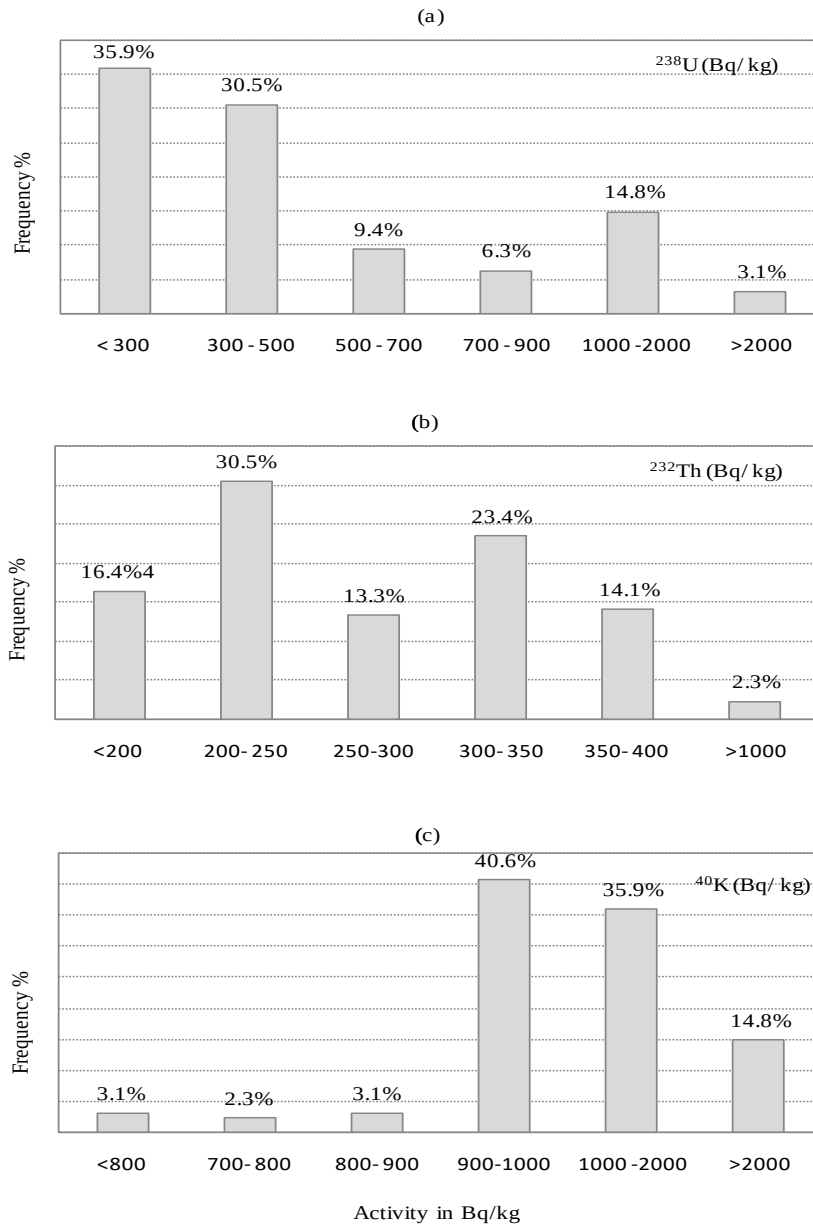


Figure (1): Frequency distributions of activity concentrations (Bq/ kg) for Um- Ara granite rocks (a) ^{238}U (b) ^{232}Th and (c) ^{40}K

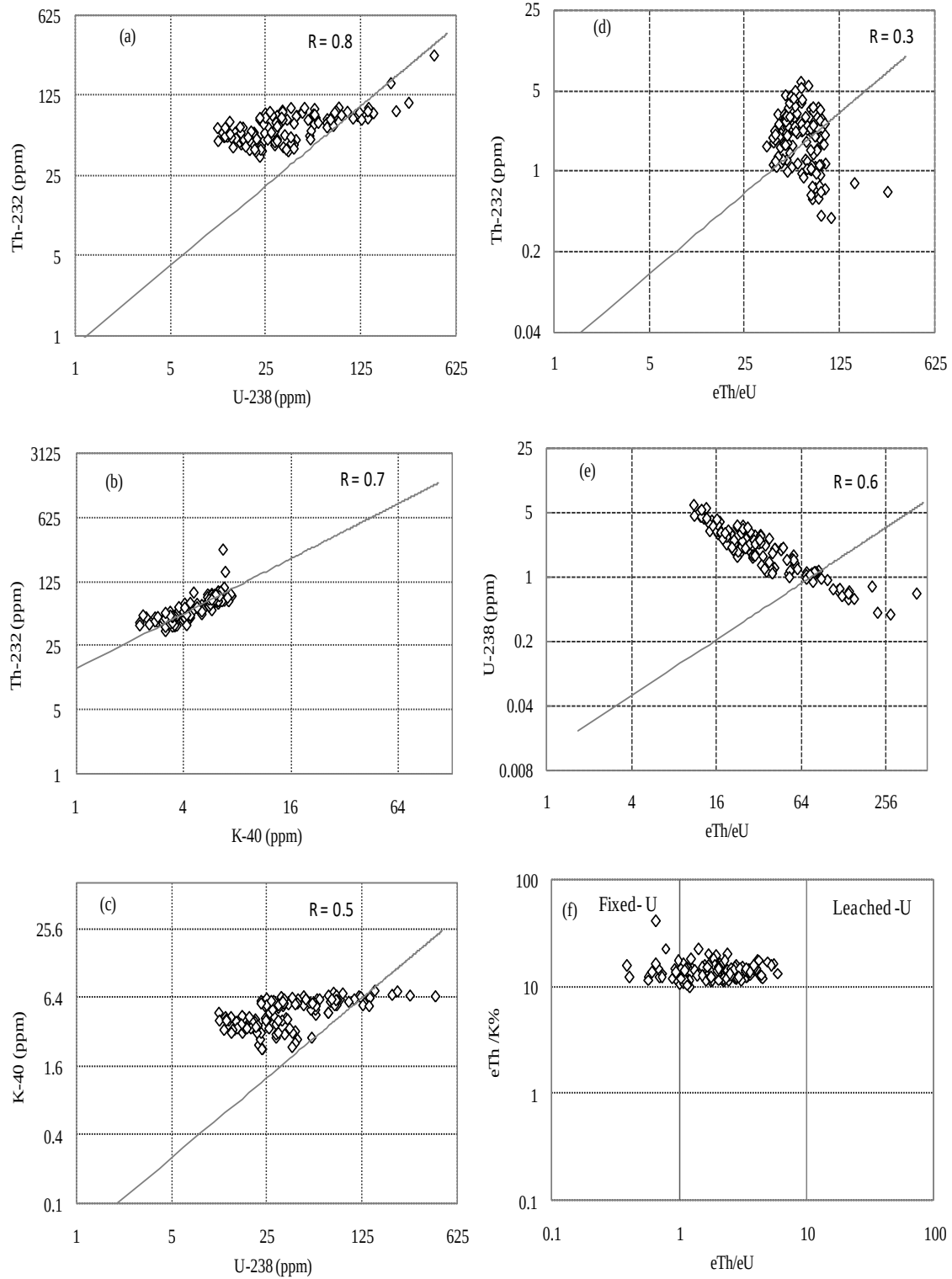


Figure (2):- (a) U vs. Th, (b) K vs. U and (c) K vsTh (d) eTh/eU vs eTh, (e) eTh/eU vs eU and (f) eTh/eU vs eTh/K% for granite.

4.3 Evaluation of radiological hazard effects

ETh and eU in ppm and K, in %, were converted to activity concentration, Bq/kg, using the conversion equations given by IAEA [27-29] and Polish Central Laboratory for Radiological Protection [28]. The specific parent activity of a sample containing one ppm, 1.1% of ^{40}K is 310 Bq/kg by weight, one ppm of Ra is 11.3 Bq/kg, of U, is 12.43 Bq/kg, and one ppm of Th 4.26 Bq/kg. Therefore, it is possible to predicate the radiation effect by measuring the following values.

4.3.1 Radium equivalent activity, Ra_{eq}

It is the activity sum of the, ^{232}Th , ^{238}U and ^{40}K radionuclides based on the assumption that 4710 Bq/kg of ^{40}K , 261 Bq/kg of ^{232}Th , and 372 Bq/kg of ^{238}U produce the same gamma-ray dose rate [30-32]

$$Ra_{eq} = A_U + 1.43 A_{Th} + 0.077 A_K$$

where A_K , A_{Th} , and A_U are the specific activities of ^{40}K , ^{232}Th , and ^{238}U in Bq/kg. Ra_{eq} were estimated from the collected rocks and is presented in Table 2. From the recorded results, we can observe that the calculated values of Ra_{eq} ranged between 514.6 and 7097.6, with an average value of 1120.4 Bq/kg. The obtained values were more than the recorded world value (369 Bq/kg).

4.3.2 Gamma activity index, I_γ

The restriction on building materials for gamma radiation is based on a dose range of 0.3 –1 mSv/y. To examine whether a building material meets these limits of dose criteria, the gamma activity concentration index I_γ is calculated from the following equation [33,34]:

$$I_\gamma = \frac{A_U}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000}$$

The presented equation depend on the radionuclides contribution to the external irradiation, it sums the three specific activities for gamma radiation index to form unit sum. $I_\gamma \leq 2$ related to a dose criterion of 0.3 mSv y^{-1} , while $I_\gamma \leq 6$ related to 1 mSv y^{-1} [35].

Thus, the activity concentration index should be used only as a screening tool for identifying materials that might be of concern to be used as construction or covering materials. According to this dose criterion, materials with $I_\gamma \geq 6$ should be avoided. In contrast, these values correspond to dose rates higher than one mSv/y, the highest dose rate value in air recommended for the population [9, 33].

Values of gamma activity index (I_γ) for Um Ara granite ranged between 1.8 and 24.1 with an average value of 3.9, higher than the acceptable value 2.

The value showed $1.9 \leq I_\gamma \leq 5.9$, where the gamma dose for these samples is higher than 0.29 mSv y^{-1} , but this value is less than the recommended dose rate in air value 0.9 mSv/y.

4.3.3 Alpha activity index, I_α

Alpha particles indices are related to radon inhalation produced from building materials. The alpha index (I_α) is defined as follows:

$$I_\alpha = \frac{A_U}{200} \leq 1$$

The ^{238}U activity concentrations in contracting materials are 199 and 99 Bq/kg, respectively, as suggested by ICRP, 1994. Therefore, $I_\alpha = 0.9$ when the maximum concentration value of ^{238}U is 199 Bq/kg, the indoor radon concentration could exceed 199 Bq/kg.

I_α ranged between 0.69 and 26.4, with an average value of 3. It indicates that the average value of the alpha activity index is higher than the acceptable value.

4.3.4 Hazard indices (H_{ex} and H_{in})

External hazard index, H_{ex}

The external hazard index (H_{ex}) measures the external hazard due to the emitted gamma radiation. The equation calculated it.

$$H_{ex} = A_K/4810 + A_{Th}/259 + A_U/370 \leq 1$$

To reduce the radiation risk from constructing materials, this index value must be less than one. For that, the Ra_{eq} value must be less than 369 Bqkg⁻¹.

Values of H_{ex} ranged between 1.4 and 19.2, with an average weight of 3. This result indicates that this value is higher than the acceptable value. This suggests that Um Ara granites' radiation risk can't be negligible.

Internal hazard index, H_{in}

The internal hazard index (H_{in}) controls the internal exposure to ²²²Rn and its radioactive levels. The following equation gives it.

$$H_{in} = A_U/185 + A_{Th}/259 + A_K/4810 \leq 1$$

If the uranium concentration values in samples under investigation are half that of the acceptable limit, then the value of H_{in} should be less than 1. Values of H_{in} ranged from 1.8 to 33.5, with an average value of 4.7. The results are more than the required values, this indicates that the radiation risks from these samples increases, so it can't be used as a building material.

4.3.5 External dose from naturally occurring radionuclides

The external absorbed dose rate in outdoor air, D_{out}

The external absorbed dose rates D can be calculated according to the following formula;

$$D = A_{Ei} \times C_F$$

A_{Ei} is the activity concentration (Bq kg⁻¹), and C_F is the dose conversion factor (absorbed dose rate in the air per unit activity per unit mass in units of nGy h⁻¹ per Bq kg⁻¹). the considered dose rate conversion factors for ⁴⁰K, ²³²Th, and ²³⁸U were determined where $C_K = 0.0429$, $C_{Th} = 0.5835$ and $C_U = 0.4551$.

The external absorbed dose rate in indoor air, D_{in}

D_{in} is calculated as follows[36]:

$$D_{in} = D_{out} \times 1.4$$

Values of D_{in} ranged from 320.6 to 4321.6, with an average value of 704.3 nGy/h. It is observed from these values that the average values for granite samples under study were much higher than the acceptable world values and higher than fair Egypt values (Table 1).

4.3.6 Annual effective dose, AEDE

The AEDE is determined using the following equations:

$$AEDE \text{ (outdoor)} = 0.7 \text{ Sv /Gy} \times 0.2 \times D_{out} \text{ nGy/ h} \times 8760 \text{ h}$$

$$AEDE \text{ (indoor)} = 0.7 \text{ Sv /Gy} \times 0.8 \times D_{in} \text{ nGy/ h} \times 8760 \text{ h}$$

Using these equations, the annual effective doses for children and infants are calculated with the numerical values of 0.8 and 0.9 of conversion factor for infants and children.

The annual outdoor effective dose of Um Ara granitic rocks varied from 0.3 to 3.9 with an average value of 0.6 mSv, from 0.3 to 4.4 with an average value of 0.7 mSv, and from 0.4 to 5 with an average value of 0.8 mSv for adults, children, and infants, respectively. At the same time, the annual indoor effective doses were ranged from 1.6 to 21.8 with an average value of 3.5 mSv, from 1.8 to 24.9 with an average value of 3.9 mSv, and from 2 to 28 with an average value of 4.4 mSv for adults, children, and infants, respectively. The total annual effective dose equivalent of Um Ara rocks is 4.1 mSv for adults, 4.7 mSv for children, and 5.2 mSv for infants. These values were greater than the world's value (0.49 mSv).

4.3.7 Annual gonadal dose equivalent, AGDE

(AGDE) was calculated using the following formula.

$$AGDE = (0.314 \times A_K + 4.18 \times A_{Th} + 3.09 \times A_U) \times 10^{-3}$$

Values of AGDE ranged between 1.6 and 21.8, with an average value of 3.5 mSv. The results are higher than the world value.

4.3.8 Excess lifetime cancer risk (ELCR)

Excess lifetime cancer risk (ELCR) is shown as

$$ELCR = AEDE \times DL \times RF$$

AEDR is the Annual Equivalent Dose Equivalent, DL is the Average Duration of Life, and R.F. is the Risk Factor (Sv^{-1}), i.e., fatal cancer risk per Sievert. Values ranged from 6.5×10^{-3} to 89.8×10^{-3} . This value is higher than the world value (0.29×10^{-3}).

5. Conclusion

The geological structure of the region under investigation was affected by the radioactivity levels of the rocks under study. This was observed by the high U, Th, and K values, which exceed the average world value for safety used as building materials.

Therefore, the granites of Om Ara may be inappropriate to choose as a building material or as an indoor and outdoor decorative. At the same time, the region under investigation shows high economic values of minerals concentration.

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