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ASSESSMENT OF RADIATION HAZARDS OF RADIONUCLIDES FOR GRANITE ROCKS FROM GABAL GHAREB, EASTERN DESERT OF EGYPT

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The objective of the present study is to measure the radioactivity concentrations of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K in the Gabal Ghareb region in the eastern desert of Egypt. The activity concentrations were measured by using a high purity germanium (HPGe) detector-based γ-spectrometry. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for the collected rock samples were varied from 6.3±0.32 to 136.73 ± 6.80 Bqkg^{-1} with an average value of 45.75±2.28 Bqkg^{-1}, from 6.05±0.35 to 120.10±6.01 Bqkg^{-1} with an average value of 50.91±2.56 Bqkg^{-1}, and from 377.54±18.87 to 1102.01±55.10 Bqkg^{-1} with an average value of 826.13±39.33 Bqkg^{-1}, respectively. The radiological risk parameters such as radium equivalent (Ra_{eq}), external risk index (H_{ex}), internal risk index (H_{in}), absorbed dose rate (ADR), annual effective dose rate (AEDR), annual gonadal dose equivalent (AGDE) and Excess Lifetime Cancer Risk (ELCR) were calculated to assess the radiation risks associated with the studied rock samples. Most of these parameters are higher than the recommended reference levels. The data obtained provide a valuable future database to estimate the impact of radioactive contamination in the studied area and in the places where the rocks are used.

Keywords: Gabal Ghareb, Eastern Desert, Radiological Risk, Natural Radionuclides, Activity.

INTRODUCTION

Natural radioactive materials (NRM) are widely found in rocks, soil, river soils, sand, sediment, rivers, oceans, building materials, and dwellings. Many studies on NRM were performed and mainly concerned with three issues. The first is to investigate the origins and distributions of

radionuclides on the earth's surface. Second, the natural radionuclides are considered as external sources of radiation to the human body. Most of these radionuclides have external exposure caused by gamma radiation and depends on the geological and geographical conditions (UNSCEAR, 2000). Finally, these studies make it possible to obtain the background baseline of naturally occurring radionuclides for investigating the environmental impact of anthropogenic radioactive sources.

The radioisotopes contained in the soil depend on the nature of the parent rock during soil formation, and biochemical and chemical interactions, which influence the radioisotopes' distribution patterns (Myrick et al. 1983; Tzortzis et al. 2003b). Generally, igneous rocks, such as granitic rocks, have high levels of radioactivity compared to the sedimentary rocks, except for phosphate rocks and some shales, which contain relatively high concentrations of radioisotopes (Tzortzis et al. 2003a; UNSCEAR 1993).

The radioactivity of granitic rocks are highly enhanced with concentrations of uranium and thorium compared to the Earth's crust, upper continental crust, and basaltic or ultramafic rocks, as shown in Table 1 (Örgün et al. 2005; Rudnick and Gao 2003; Tzortzis and Tsertos 2004). Although granitic rocks contain natural radionuclides that contribute to the background radiation levels, they are used most widely as a building material in the construction of homes worldwide. This is one of the most important reasons to measure the natural radioactive isotopes concentration and to assess the related radiological hazards to which the human body might be exposed.

The present study aims to determine the distribution of ^{226}R, ^{232}Th and ^{40}K for some granite rocks collected from the Gabal Gharib region, Eastern Desert of Egypt. In this study, the radium equivalent (Ra_{eq}), absorbed dose rate in air (ADR), annual effective dose rate, external and internal hazard indices (H_{ex}, H_{in}) were calculated and compared with the recommended limit values. In this way the background radiation level and the effect of radiation on the local population were estimated. These

results are of great interest, as granites are widely used as material for construction and decoration, including interior coating.

Study area and Geological setting

Study area of Gabal Gharib is located in the Eastern Desert of Egypt in the northeastern part, lies at 100 km north of Hurghada. It lies between Latitudes 28 05' and 28 10' N and Longitudes 32 50' and 32 58' E (Fig.1). The area has a lot of intermediate to acidic Dokhan volcanic, calc-alkaline granodiorite to adamellite suite of volcanic arc granite and was formed on an active continental margin (Abdel-Rahman and Martin1987). Gabal Gharib (1,751 m) is composed of peralkaline granite and covers an area of about 25 km^2. It is introduced throughout the calcium-alkaline-adamellitegranodiorite. The contacts are very intrusive. A metasomatic zone of 2 km maximum is at the expense of alkaline-limestone host rocks (Abdel- Rahman and Martin1990).

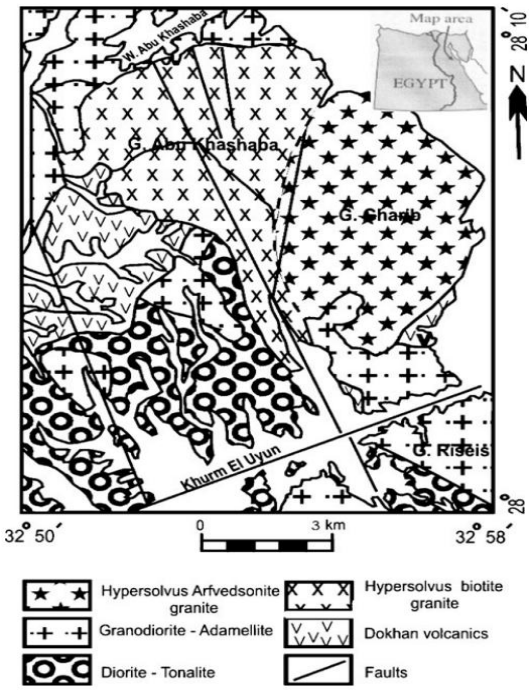

Fig.1: Geological description of Gabal Gharib area modified after Abdel-Rahman and Martin (1990).

Experimental techniques

Sample preparation

A total of 20 rock samples were collected from the Gabal Ghareb mountain region in the eastern desert of Egypt, the samples were cut by porphyry dykes invaded by jasperoid veins and quartz diorite (Arafa, 2004). The samples were crushed by a jaw crusher to get a fine powder, sieved through 200µm mesh and dried for 3 h at a temperature of 105°C to remove moisture. Each sample was weighed and placed in a plastic container with a diameter of 9.52 cm, a height of 8.26 cm and thickness of 0.051 cm. Each container is then stamped tightly with a vinyl tape around the neck to ensure that the radon gas does not run out. Then, the containers were kept in the laboratory for at least 1 month to achieve secular equilibrium between ^{226}Ra and ^{222}Rn and then counted for 8 to 24 h, depending on the concentration of radionuclides.

Radioactivity measurement

The samples were measured in Nuclear Physics Lab, the department of Physics, Faculty of science, Assiut University, using G*amma*-ray spectrometer with a reverse electrode germanium (REGe) detector model GR4020 connected to a Canberra digital spectrum analyzer model DAS-1000 as a data acquisition system. The detector had closed-end coaxial G*amma*-ray crystal made of high purity germanium (HPGe) in a vertical configuration cooled with liquid nitrogen. The energy resolution of the detector reads approximately 2.000 keV and ≤0.925 keV at 1.33 MeV and 122 keV, respectively, while the relative efficiency is 40%. The germanium crystal is located inside a lead shield to reduce the environmental background. The shield consists of four layers, a low carbon steel of (0.38 in.) thick as an outer jacket, a bulk shield of lead of (4 in.) thickness, and graded linings to absorb low energy X-rays of (0.040 in.) tin and (0.062 in.) copper (Canberra Industries, Inc., USA)(Canberra, 2013).

Spectra were analyzed manually or automatically using GENIE-2000 software (Canberra 2013) and peak analysis reports (run by the program itself) using spreadsheets (Microsoft Excel).

The background radiation level can be determined by used an empty cylindrical beaker which counted at the same time as the samples under the same geometry. The net peak areas of gamma rays of measured isotopes were corrected by using the background spectra. The ^{226}Ra radionuclide have been estimated from the 351.9 keV and 295.22 keV gamma peaks of ^{214}Pb and 609.3 keV, 1120.3 keV and 1764 keV gamma peaks of ^{214}Bi. On the other hand the ^{232}Th radionuclide was estimated from the 911.2 keV, 209.25 keV, 338.32 keV, and 968.97 keV gamma peaks of ^{228}Ac, the 238.6 keV gamma peak of ^{212}Pb , 2614 keV and 583.19 keV gamma peaks of ^{208}Tl. While the ^{40}K radionuclide was estimated using the 1461 keV gamma peak from ^{40}K itself.

The activity concentrations of radionuclides in the samples were calculated using the following relation:

$$A_{E\gamma,i} = \frac{NP}{t_c.I_\gamma(E_\gamma).\varepsilon(E_\gamma).M} \quad (1)$$

Where: *NP* is counts number of a given peak area corrected for background peaks of a peak at energy E_γ, t_c is counting lifetime, $I_\gamma(E_\gamma)$ is the number of gammas per disintegration of this nuclide for a transition at energy E_γ, $\varepsilon(E_\gamma)$ is the detection efficiency at energy $E\gamma$ and M is the mass in kg of the measured sample.

Radiological hazard parameters

Radium equivalent (Ra_{eq})

The radium equivalent (Ra_{eq}) was calculated on the basis of an estimate: 370 Bq Kg^{-1} of ^{226}Ra, 259 Bq Kg^{-1} of ^{232}Th and 4810 Bq Kg^{-1} of ^{40}K produced the same dose rate of gamma radiation (Krieger, 1981; Beretka and Mathew, 1985). Therefore, the Ra_{eq} of a sample is given by:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

Where A_{Th} is activity concentration of ^{232}Th in Bq kg^{-1}, A_{Ra} is activity concentration of ^{226}Ra in Bq kg^{-1}, A_K is activity concentration of ^{40}K in Bq kg^{-1}.

Absorbed dose rate (ADR)

The total dose rate absorbed into the air (nGy h^{-1}) due to the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}) can be calculated utilizing the formula described by Beck et al. (1972) and UNSCEAR (1993).

$$ADR(nGyh^{-1}) = 0.43A_{Ra} + 0.666A_{Th} + 0.042A_K \quad (3)$$

Annual effective dose rate (AEDR)

According to the ADR, the annual effective dose rate (AEDR), outdoor and indoor was calculated using the formula:

$$AEDR\left(\frac{msv}{y}\right) = ADR\left(\frac{nGy}{h}\right) \times 8760\left(\frac{h}{y}\right) \times 0.2 \times 0.7\left(\frac{sv}{Gy}\right) \times 10^{-6} \quad (4)$$

Measuring the occupancy factor indoor of construction materials is estimated to be 0.8, so the equation is:

$$AEDR\left(\frac{msv}{y}\right) = ADR\left(\frac{nGy}{h}\right) \times 8760\left(\frac{h}{y}\right) \times 0.8 \times 0.7\left(\frac{sv}{Gy}\right) \times 10^{-6} \quad (5)$$

Where the conversion coefficient from the absorbed dose in air to the effective dose is (0.7 Sv/Gy), and the outdoor occupancy factor is (0.2) as suggested by (UNSCEAR 2000).

External Hazard Index (H_{ex})

Exposure to natural gamma radiation is assessed by evaluating the external hazard index (H_{ex}). H_{ex} only considers the risk of external exposure and can be calculated for each sample tested according to the following equation (Beretka and Mathew 1985):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (6)$$

Where:A_{Ra}, A_{Th} and A_K are the activities concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq.kg^{-1}, respectively.

Internal hazard index (H_{in})

Besides the external danger, radon and its short-lived products are also dangerous to the respiratory system. Internal exposure to radon and its secondary products is quantified by the H_{in} internal risk index, which define as follows (Beretka and Mathew 1985):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (7)$$

For the safe use of granite stones as a building material, H_{ex} and H_{in} should be smaller as a unit (Al-Zahrani 2017; Xinwei 2005).

Excess lifetime cancer risk (ELCR)

Possible carcinogenic effects are determined by estimating the prospect of cancer in a population of individuals for a specific lifetime as a result of expected consumption (and exposure) and specific dose response data. The Excess Lifetime Cancer Risk (ELCR) can be determined using the following statement (Ravisankar et al.,2014):

$$ELCR = AEDR \times DL \times RF \quad (8)$$

Where RF represents the risk factor of 0.05 and DL represents duration of life of 70 years (Taskin et al, 2009).

Annual gonadal dose equivalent (AGDE)

The annual equivalent dose in gonad (AGDE), due to the specific activity of ^{226}Ra, ^{232}Th and ^{40}K, was calculated by the following formula (Krishnamoorthyet al., 2014):

$$\text{AGDE} = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_K \quad (9)$$

RESULTS AND DISCUSSIONS

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in rock samples collected from Gabal Ghareb together with their uncertainties and average values are listed in Table 1 and shown in Fig.2. The highest values of ^{226}Ra, ^{232}Th and ^{40}K concentrations are 136.73±6.80, 120.10±6.01 and 1102.01±55.10 Bq Kg^{-1} respectively, where the minimum values are 6.3±.32, 6.05±.35 and 377.54±18.87 Bq Kg^{-1} respectively.

The variations in natural radioactivity levels in different samples are due to changes in the concentration of these elements in the geological formations. Table 1 shows that the average values of ^{226}Ra, ^{232}Th and ^{40}K concentrations are higher than the recommended reference level of 35, 30, and 400 Bq kg^{-1} respectively (UNSCEAR 2000).

Radiological hazards

Table 2 and Fig 2 show that the average value of radium equivalent (Ra_{eq}) is 182.16 $Bqkg^{-1}$, which is lower than the recommended limit of 370 $Bqkg^{-1}$ (UNSCEAR,2000). Only the sample G20 has a value higher than the recommended limit.

The obtained results of absorbed dose rates together with the annual effective dose rates for the examined samples were inserted in.The absorbed dose rate, ADR, varies from 22.42 to 172.10 nGy h^{-1} with an average value of 86.67$nGyh^{-1}$ that is slightly higher than the worldwide average of absorbed gamma dose rate of 59 nGy h^{-1} (UNSCEAR 2000). Fig 4 shows that only two samples G3 and G12 are lower than the reference value of ADR.

The annual effective dose rate outdoors and indoor were also listed in Table 2.The average values of AEDR (out) and AEDR (in) are 0.11 and 0.42 $mSvy^{-1}$ respectively. It is quite clear from Table 2 that the average value of AEDR (out) is higher than the world average of 0.07 $mSvy^{-1}$ (UNSCEAR 2000).

Table 2 shows the variation in the values of the external and internal hazard index in the different studied samples. It is quite clear that the average values of these parameters (H_{ex} and H_{in}) of 0.49 and 0.62 respectively are lower than unity (the recommended safety limit). This

reflects that gamma radiation has no harmful effects to workers or people who live nearby this area. Fig 5 shows that the values of H_{in} for samples G17, G19 and G20 are higher than unity.

Table1: Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the granitic rock samples of the studied area.

Sample ID	**Activity concentration in (Bq kg^{-1})**		
	^{226}Ra	**^{232}Th**	**^{40}K**
G1	36.10±1.82	55.79±2.83	873.41±43.67
G2	31.05±1.54	48.99±2.44	832.71±41.60
G3	6.30±.32	6.05±.35	377.54±18.87
G4	44.65±2.23	62.35±3.11	932.87±46.64
G5	28.69±1.42	31.96±1.59	688.97±34.44
G6	24.41±1.22	34.98±1.74	722.17±36.12
G7	26.06±1.34	31.67±1.58	930.39±46.51
G8	31.70±1.57	29.69±1.48	990.03±49.50
G9	27.38±1.36	18.97±.95	860.43±43.00
G10	32.90±1.64	40.87±2.04	818.45±40.92
G11	25.42±1.27	36.64±1.83	1102.01±55.10
G12	20.76±1.02	18.57±.93	558.88±27.92
G13	35.43±1.77	59.42±3.22	975.72±48.81
G14	28.78±1.42	35.65±1.78	867.33±43.40
G15	32.44±1.62	53.31±2.66	854.85±42.78
G16	44.55±2.23	58.43±2.92	917.22±45.86
G17	130.68±6.52	107.79±5.40	825.41±41.27
G18	63.62±3.18	67.83±3.39	739.65±37.00
G19	107.40±5.37	99.12±5.00	788±39.41
G20	136.73±6.80	120.10±6.01	866.63±43.32
Min	6.3±.32	6.05±.35	377.54±18.87
Max	136.73±6.80	120.10±6.01	1102.01±55.10
Average	45.75±2.28	50.91±2.56	826.13±39.33

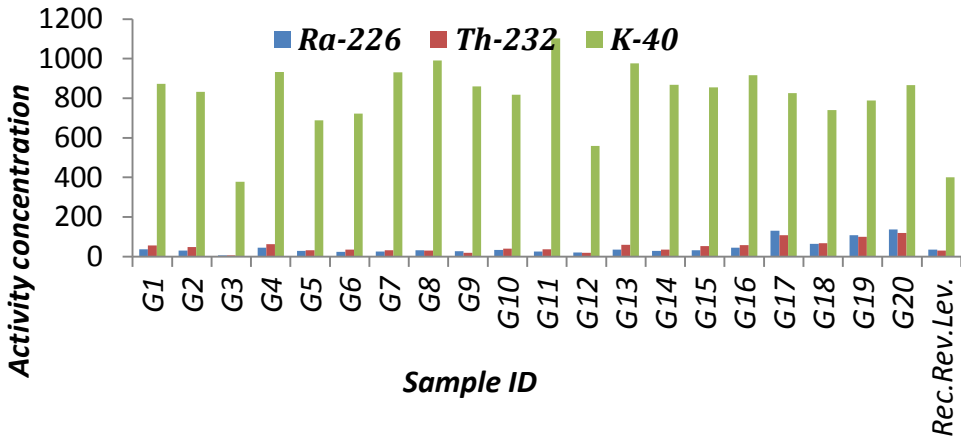

Fig .2: The distribution of the activity concentration of ^{226}Ra, ^{232}Th and^{40}K in study samples.

Table 2: Radiological parameters in granitic rock samples of studied area.

sample	Ra (eq) [Bq kg^{-1}]	ADR [nGy/h]	AEDR(out) [mSv/year] out	AEDR(in) [mSv/year] in	H $_{ex}$	H $_{in}$	AGDE [μSv y^{-1}]	ELCR × 10^{-3}
G1	183.13	87.06	0.11	0.43	0.49	0.59	618.98	0.39
G2	165.22	78.91	0.10	0.39	0.45	0.53	562.19	0.35
G3	44.02	22.42	0.03	0.11	0.12	0.14	163.30	0.10
G4	205.64	97.47	0.12	0.48	0.56	0.68	691.51	0.43
G5	127.45	61.50	0.08	0.30	0.34	0.42	438.60	0.27
G6	130.04	62.74	0.08	0.31	0.35	0.42	448.41	0.28
G7	142.99	70.25	0.09	0.34	0.39	0.46	505.06	0.31
G8	150.39	75.76	0.09	0.36	0.41	0.49	532.92	0.33
G9	120.76	60.24	0.08	0.29	0.33	0.40	434.06	0.26
G10	154.37	74.26	0.09	0.37	0.42	0.51	529.50	0.33
G11	162.67	80.16	0.10	0.39	0.44	0.51	577.74	0.35
G12	90.34	44.28	0.06	0.22	0.24	0.30	317.24	0.19
G13	195.53	93.24	0.12	0.46	0.53	0.62	664.24	0.41
G14	146.54	71.25	0.09	0.35	0.40	0.47	510.28	0.31
G15	174.50	83.12	0.11	0.41	0.47	0.56	591.50	0.37
G16	198.73	94.40	0.12	0.46	0.54	0.66	669.89	0.42
G17	348.38	160.15	0.20	0.78	0.94	1.29	1113.55	0.70
G18	217.57	101.42	0.13	0.50	0.59	0.76	712.35	0.45
G19	309.82	142.59	0.18	0.70	0.84	1.13	993.64	0.62
G20	375.19	172.10	0.21	0.84	1.01	1.38	1196.61	0.75
Mean	182.16	86.67	0.11	0.42	0.49	0.62	613.58	0.38

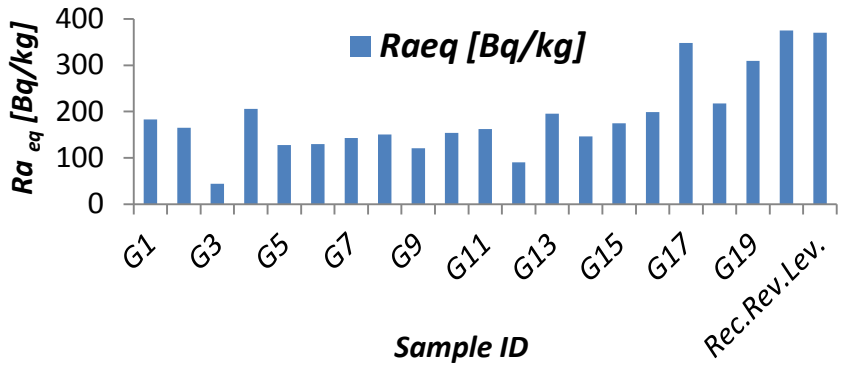

Fig. 3: The distribution of Ra_{eq} for the study samples.

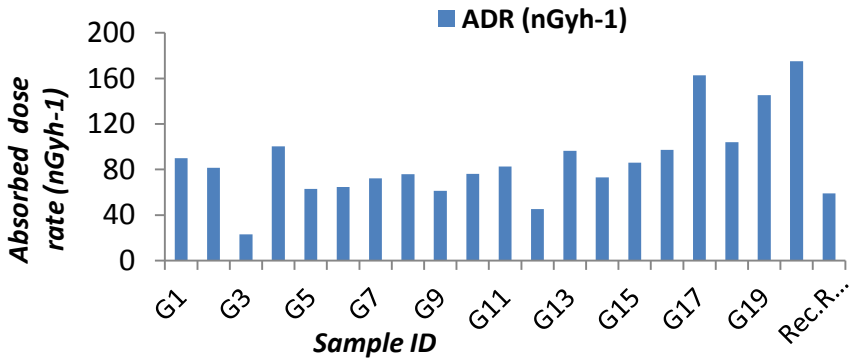

Fig. 4: The distribution of ADR for the study samples.

The AGDE values in μSvy^{-1} are listed in Table 2 and shown in Fig.6. The values for all studied samples are above the global average for soil 300 μSvy^{-1} (Xinwei et al. 2006; Zaidi et al. 1999), referring to a significant radiation risks associated with these types of granitic rocks.

The ELCR values of all the studied samples are listed in Table 2 and their distribution is illustrated in Fig.7.All ELCR values are above the world average of 0.29×10^{-3} (UNSCEAR 2000), except for samples G3, G5, G6 and G12. The average value of ELCR was 0.38×10^{-3}, which is well above the global average. This indicates to that these samples pose a risk and are harmful to humans. .

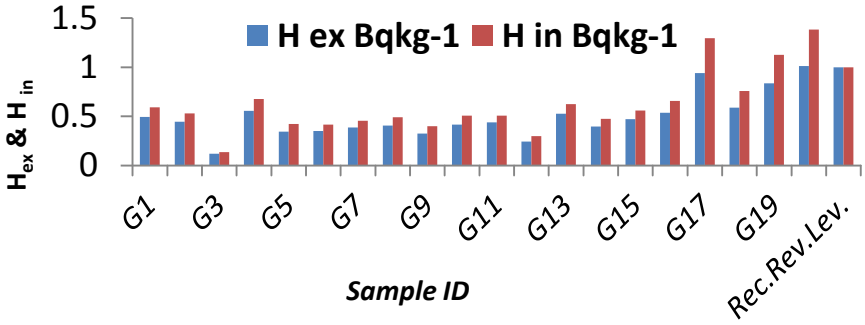

Fig .5: The distribution ofH_{ex} and $H_{in}$for the study samples.

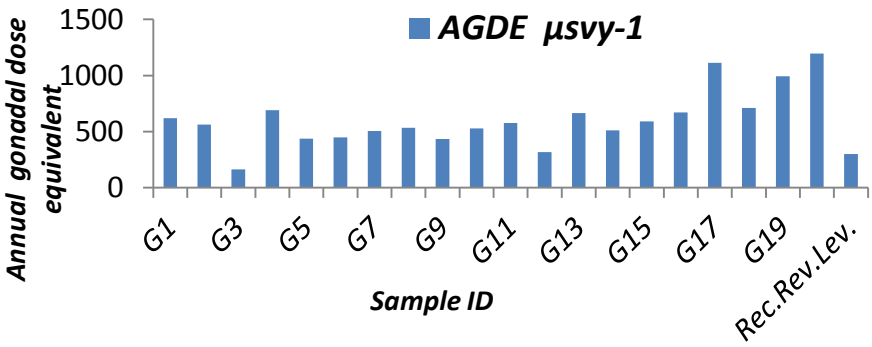

Fig .6: The distribution of AGDE for the study samples.

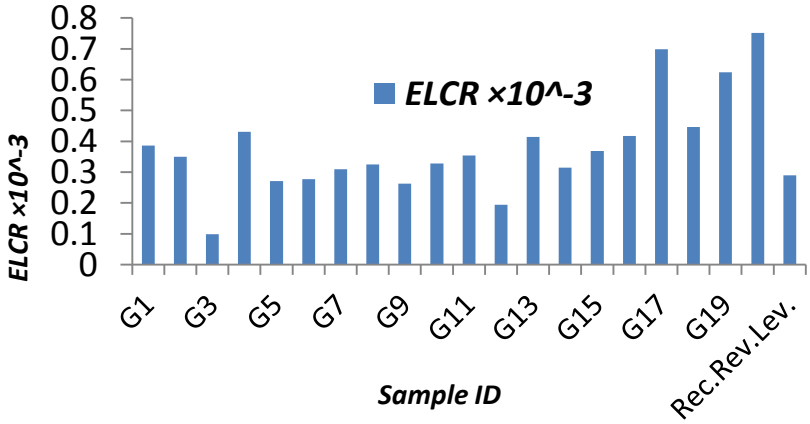

Fig .7: The distribution of ELCR for the study samples.

The correlation coefficient of natural radionuclides for the study samples

The relationships between ^{226}Ra, ^{232}Th and ^{40}K were computed and diagrammatically shown in Fig.8, which illustrates a good correlation exists between ^{226}Ra and ^{232}Th where the correlation coefficient (R^2 = 0.89) is nearly unity. There is a poor correlation between ^{226}Ra and ^{40}K and between ^{232}Th and ^{40}K, where the correlation coefficients are R^2=0.0184 and 0.0768 respectively.

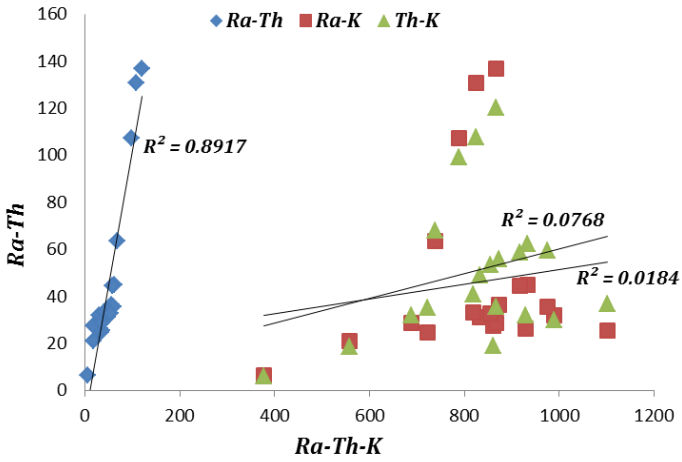

Fig.8:The correlation coefficient of natural radionuclides for the study samples

CONCLUSION

Twenty rock samples were collected from Gabal Ghareb area (Eastern desert of Egypt) to determine the specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K using HPGe detector. The average values of the activity concentrations for ^{226}Ra, ^{232}Th and ^{40}K are higher than the recommended reference levels of 35, 30, and 400 Bqkg^{-1} respectively. For all samples, the estimated values of Ra_{eq} are lower than the recommended limit of 370 Bqkg^{-1}, while the average value of ADR are higher than the global average absorbed dose rate of 59 nGyh^{-1}. The average value of AEDR Outdoors is higher than the world average annual effective dose rate of0.07 mSvy^{-1}.The average values of H_{ex} and H_{in} are lower than unity. The average values of AGDE and ELCR are higher than the world average of 300μSvy^{-1} and 0.38×10^{-3} respectively. Therefore, the above

results indicate to a high radiological hazards coming from the use of the examined rock samples.

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تقدير المخاطر الأشعاعية للنويدات المشعة للصخور الجرانيتية بجبل غارب ، الصحراء الشرقية ،مصر.

تهدف الدراسة الحالية الى حساب نسب العناصر المشعة طبيعياً وهى (البوتاسيوم-٤٠ ، الراديوم- ٢٢٦،الثوريوم- ٢٣٢) فى منطقة جبل غارب فى صحراء مصر الشرقية وحساب الجرعة الإشعاعية ومخاطر التعرض الناتجة من إشعاعات جاما الناتجة من تلك العناصرالمشعة.

تم استخدم مطياف أشعة جاما الذى يحتوى على كاشف الجرمانيوم عالى النقاء (Canberra, GR4020-model) (HPGe)

تم تقديرقيمة النشاط الأشعاعى للعناصر (البوتاسيوم-٤٠ ، الراديوم-٢٢٦،الثوريوم- ٢٣٢) فى العينات الصخرية المجمعة ووجد أن قيمتها لعنصر البوتاسيوم-٤٠ تتراوح بين ٦.٣±٠.٣٢ إلى ١٣٦.٧٣±٦.٨٠ بيكريل/ كجم بقيمة متوسطة ٤٥.٧٥±٢.٢٨ بيكريل/ كجم ،وللراديوم- ٢٢٦ من ٦.٠٥±٠.٣٥ إلى ١٢٠.١٠±٦.٠١ بيكريل/ كجم بقيمة متوسطة ٥٠.٩١±٢.٥٦ بيكريل/ كجم ،ولعنصر الثوريوم- ٢٣٢من ٣٧٧.٥٤ ±١٨.٨٧ إلى ١١٠٢.٠١ ±٥٥.١٠ بيكريل/ كجم بقيمة متوسطة ٨٢٦.١٣ ±٣٩.٣٣ بيكريل/ كجم .

تم أيضا حساب بعض المعاملات مثل المكافئ الراديومى (Ra_{eq}) ، و خطر التعرض الخارجى (H_{ex})، و خطر التعرض الداخلى (H_{in})، ومعدل الجرعة الممتصة (ADR) ، ومعدل الجرعة الفعالة السنوية (AEDR) ، ومكافئ الجرعة المناعية السنوية (AGDE) و مخاطر السرطان (ELCR) تم حسابهم لتقييم مخاطر الإشعاع المرتبطة بعينات الصخور المدروسة .معظم هذه المعاملات أعلى من المستويات المرجعية الموصى بها .توفر البيانات التي تم الحصول عليها قاعدة بيانات مستقبلية قيّمة لتقدير تأثير التلوث الإشعاعي في منطقة الدراسة وفي الأماكن التي يتم فيها استخدام الصخور.