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Environmental Characterization and Natural Radioactivity Influential on the Mountains of the Red Sea Coast, Egypt

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Abstract

The Granite Mountains contains a certain amount of natural radioactivity that generally results from the decay of uranium, thorium and ^{40}K isotopes. Calculation of the concentration levels in mountains rock from Knowledge spatial distribution and sources of these isotopes this was very important to avert negative effects the resulting from it. The present work focuses on investigating the distribution, environmental effect and sources of ^{235}U, ^{238}U, ^{232}Th, as well as the activity of gross β and α in rock in some locations in this mountains. Additionally, rocks samples were analyzed by the high-resolution gamma spectrometers techniques. Results detect significant differences in radioactivity in terms of sampling sites variation and clarified a relatively high concentration of ^{238}U in some locations. The activities of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K were measured as well as the radiological hazard parameters were calculated in the samples. The activities concentration average of (^{238}U, ^{226}Ra, ^{232}Th and ^{40}K) were 191.71 ± 23.55, 177.49 ± 25.62, 65.88 ± 6.48 and 192.66 ± 23.19 Bq/kg respectively. The annual effective dose rate (mSv/y), the mean of the absorbed dose rates (D), radium equivalent (Raeq), the external hazard index (Hex) and the internal hazard index (Hin) and the representative level index (Iγ, Iα) were; 0.25 mSv/y, 205.64 nGy/h, 1.01 mSv/y, 286.90 Bq/kg 1.97 and 0.89, respectively. The calculated radiation hazard parameters in some samples were lower than the global average and other were higher than the global average. The (Hex, Hin, Iα and Iγ) exceeded the unity and out of the human health safe limit and it may be harmful to the peoples in the region. Most of the ^{238}U concentrations in the rock are below the World Health Organization permissible limit for rock. The relatively high uranium concentrations in some rock samples suggest a long period of geochemical interactions between rocks, sediments and water. The results indicate higher levels of the activity of ^{222}Rn and ^{226}Ra lower than the permissible limits for rock. The specific activity ratios of ^{226}Ra/^{238}U and ^{232}Th/^{238}U were calculated for evaluation of the behaviors of these radionuclides. These results give the basic values for the distribution of natural radionuclides in the region and will be used as reference information to determine any future changes.

***Keywords:** Mountains red sea; Radiation hazard; Radionuclides; Human health*

Introduction

The radioactive nuclide exists anywhere on the Earth's surface and can usually to be in grouped into four classes to their origin: Cosmogenic radionuclides, primordial radionuclides natural decay series daughters and anthropogenic radionuclides [1]. Primordial radionuclides have existed on earth since its creation during the formation of the earth and are distinguished by their extreme long half-lives compared to the life of the Earth, such as ^{40}K ($T_{1/2}$ = 1.248 × 10^9 years), ^{232}Th ($T_{1/2}$ = 1.405 × 10^{10} years) and ^{238}U ($T_{1/2}$ = 4.468

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$\times 10^9$ years). Cosmogenic radionuclides are produced by the interaction of cosmic radiation with the Earth's atmosphere and surface. Examples of commonly used cosmogenic radionuclides in chronology are ^{14}C and ^{10}Be [2]. Natural decay series radionuclides are generated from the continuous decay of primordial radioactive isotopes (e.g. ^{232}Th, ^{235}U and ^{238}U). The decay processes comprise nuclear transformation associated with emission of different types of subatomic particles [3]. The decay of these daughters' nuclides induce more than 80% of the total effective radiation dose to the environment and are a major source of radiation hazards. Some of short lived radionuclides, such as ^{131}I and ^{137}Cs, are introduced to the environment through human activities including nuclear weapon testing, accidental releases from nuclear power plants, nuclear fuel reprocessing and many other industrial and medical uses, these radionuclides are called anthropogenic radionuclides whereas the other three origins of radionuclides are natural occurring.

The main sources of radiation exposure to human beings are natural and artificial radionuclides. The inevitable feature of life on earth contains radiation exposure due to cosmic from naturally occurring radioactive material (NORM), cosmic and internal source [4]. These expositions originate primarily from gamma radiation grow from the decay of these radioactive nuclides outside the human body. External irradiation from radionuclides naturally present in the around us is an important component of the exposure of human populations. Since this is not the distribution of radioactive isotopes uniformly in nature, and have knowledge of their dispersion in the rocks and rocks enables one to assess any radiation danger potential of occupants that use of these materials is where to build houses. Because of the health risks associated with exposure to internal radiation, many governments and international bodies such as the International Committee adopted for Radiological Protection [5] measures to reduce these risks. There is a need to measure the natural radioactivity due to the gamma radiation dose rate for the implementation of preventive measures whenever the dose has been found that the recommended limits are above. It is very important to estimate the levels of natural radioactivity in the rocks, to evaluate the gamma dose rate emerging from the earth's crust for the outdoor occupation. The main sources of NORM are members of ^{238}U (^{226}Ra) and ^{232}Th decay chains and ^{40}K that are present in various degrees in all media in the environment, including the human body itself. Ago 98.5% of the radiological impacts of the uranium group are generated by radium and its daughter products from ^{238}U and other ^{226}Ra precursors are normally neglected [4]. In terms of the NORM, it is enriched igneous rocks of the rhyolites, granite, carbonatite and alkaline in composition K, Th and U, compared with sedimentary rocks. Nationwide surveys have been done to determine the radium equivalent activity of rock and rock samples in many countries [6-10].

Experimental Technique

Site description

Thirty rocks samples are collected along the Red Sea coast from the city of Suez to Marsa Alam city by long 660 km away. 30 samples were collected from 15 locations along the Red Sea coast of Egypt's as in (FIG.1) and an average of two samples from each site. Samples from S1 to S7 were collected from the interior coast region of the Suez Gulf and the Aqaba Gulf on the Red Sea coast, and as can be seen from (FIG. 1) The samples from S8 to S16 were collected from the mountains of Red Sea chains along the entire area of study, which extended from Suez city (latitude 28030'3900" N) to Marsa Alam city south (Latitude 25040' 2900" N).

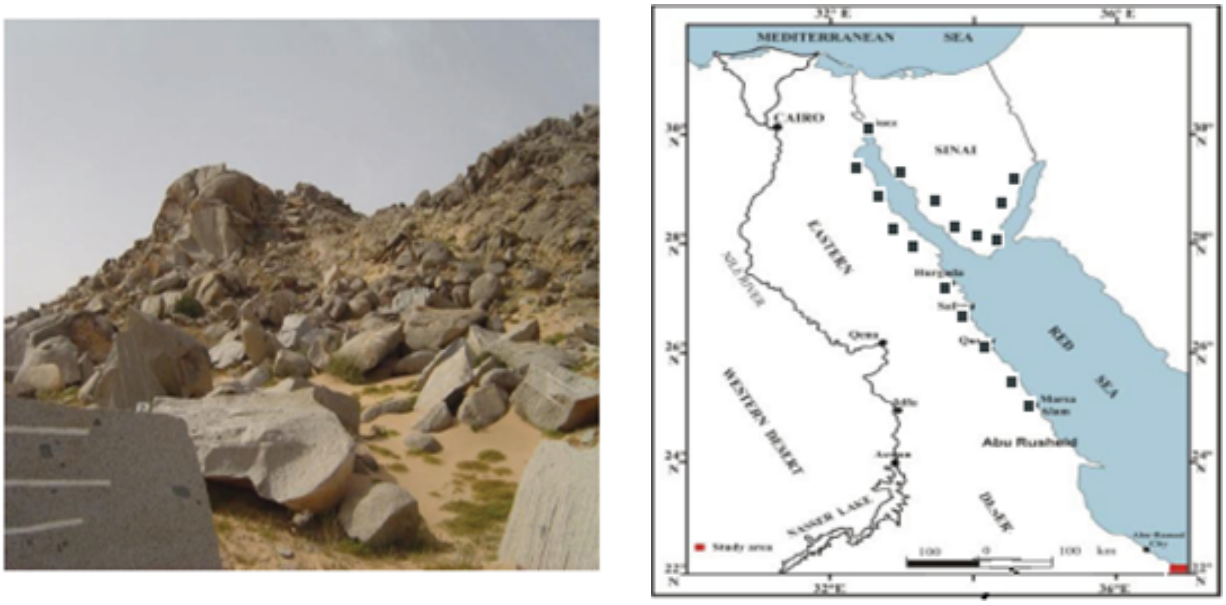

FIG.1. **The mountain red sea location and Location map of the collected samples.**

Sample collection and preparation

Samples of 30 different types of the rock collected directly from the producers in 15 Egyptian Sites as shown in (FIG.1) we did so in order to avoid eventual miss identifications of the rock. Thus, all of these collected rock samples were classified in accordance to their colors, sites of extraction and mineralogical compositions. The samples were dried in an oven at ^{110}CO till constant dry weight was obtained, mashed and homogenized. After that, the homogenized samples were weight was stored in Marinelli vessel a 250 ml plastic container to its full volume with uniform mass. These containers were tightly protected and externally to ensure that all products daughter of uranium and thorium, and in particular, radon gas formation, not escape. It has been determined net weight of the sample prior to sorting. The samples are placed for 30 to 40 days are stored on before counting to ensure ensure ^{226}Ra and short-lived offspring have to get the radioactive balance [11-14].

Radioactivity measurements

The activity concentration of the natural radioactivity ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in the samples were determined using a high-resolution HPGe γ-spectrometry system with 40% counting efficiency. The resolution of this spectrometer was 1.89 keV at 1332 keV γ -rays of ^{60}Co. The spectrometer efficiency calibration with gamma-ray was performed with the radionuclide-specific efficiency method in order to avoid any uncertainty in gamma-ray intensity as well as the impact of coincidence collection and self-absorption impact of the emitting gamma photons. The set of certified reference materials (IAEA) was used, with densities similar to the rock samples measured after pulverization. This was performed by taking 250 cm^3 counting vials filled up to a height of 7 cm, which corresponds to 170 cm^3, with reference building materials. The measurement duration was up to 90 000 sec and were carried out in the Laboratory, chemical warfare, radioactive materials department, the Egyptian Ministry of Defense. The gained spectra were analyzed with the use of Canberra system with Genie 2000 software version 3.0. Based on the following gamma-ray transitions (in keV), the activity concentrations for the radionuclide were calculated. The ^{226}Ra activities (or ^{238}U activities for samples assumed to be in radioactive equilibrium) were estimated from ^{234}Th (92.38 keV, 5.6%), while γ-energies of ^{214}Pb (351.9 keV, 35.8%) and ^{214}Bi (609.3, 45%), (1764.5 keV, 17%) and ^{226}Ra (185.99 KeV, 3.5%)were used to estimate the concentration of ^{226}Ra. The Gamma- ray energies of ^{212}Pb (238.6 keV, 45%), and ^{228}Ac (338.4 keV, 12.3%), (911.07 keV, 29%), (968.90 keV, 17%) were used to respect the concentration of ^{232}Th. The natural great number of ^{235}U is only 0.72% of the total uranium content and hence was not see in the present work. The C_K of ^{40}K were measured by its own gamma rays (1460.8 keV, 10.7%).

The following relation was used to obtain the lowest limits of detection (LLD) [15]:

$$LLD = \left(\frac{4.46 S_b}{\varepsilon I_\gamma} \right) Bq / Kg \quad (1)$$

Where Sb is the estimated standard the error of the net background count rate in the spectrum of the radionuclide, ε represents the counting efficiency and Iγ is the abundance of gamma emissions per radioactive decay. The LLD value for ^{238}U was obtained to be 3.21 Bq/kg while that of ^{232}Th and ^{40}K were 2.44 and 10.81 Bq/kg respectively.

Result and Discussion

Activity concentrations and ratios

Activity concentrations were calculated subtraction of the background. The activities were determined by measuring their respective decay daughters [16]. It has been counting the empty polystyrene containers in the same way as samples to determine the distribution of the background due to naturally occurring radionuclides in the environment surrounding the detector. The intensity of the activity concentrations was calculated for each line taking into account the mass of the sample, the branching ratios of the γ -decay, the time of counting and the efficiencies of the detector [17,18]. The activity concentrations of the samples were calculated from equation (2):

$$C = (CSP)\ net / I \times Eff \times M \quad (2)$$

The results of analysis of activity concentration of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K radionuclides in all samples for different locations of the study area are presented in TABLE 1. The activities concentration of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K of most the rocks samples exceed the average level of these radionuclides in regular 288.61 ± 38.71, 303.16 ± 46.24, 98.46 ± 13.47 and 286.90 ± 31.29 Bq/kg, respectively, (TABLE 1). The range of measured activity of ^{238}U in the rock samples was 97.92 to 289.71Bq/kg with an average of 191.71 Bq/kg. The minimum value obtained in sample No. 28 (Location 14) and a maximum for the sample No. 3 (Location 2). The differences are result of the geochemical structure and source of a rock types in a certain area. The range of measured activity concentration of 232Th for the rock samples was 13.34 to 98.46Bq/kg with an average of 65.88 Bq/kg. The minimum value obtained in sample No. 30 (Location 15) and a maximum for the sample No. 10 (Location 5). The differences are significant in all samples. The activity concentration rang of ^{40}K was 111.19 to 286.90 Bq/kg, with an average value of 192.66 Bq/kg. These differences also attributable to

the rock type differences in the region under investigation. The measured mean of the radiological hazards in the place were higher than the global average of rocks [4] except for ^{40}K is lower. The mean activity concentrations are 5.4, 5.1, 2.2 and 0.48 times of the worldwide mean concentrations of these elements as: (35 Bq/Kg for ^{238}U and ^{226}Ra) and (30 Bq/kg for ^{232}Th) and (400 Bq/kg for 40K). This indicates that the study area is composed of rocks having low potassic values. The large different of the activity concentration values are due to their present in the mountain environment and their chemical, physical and geochemical properties, in addition to that the expansion of the study area on the Red Sea borders. The differences are attributable to the geochemical composition and origin of rock types in a particular area. For a detailed study, $^{238}U/^{226}Ra$, $^{238}U/^{40}K$ and $^{232}Th/^{40}K$ ratios are calculated and tabulated in TABLE 1. It is generally expected that ^{238}U and ^{226}Ra being in the same series, are in equilibrium, however, diversity of their ratio from unity was found in the present measurements. ^{238}U/226Ra varied in the range of 0.74 and 2.03 with an average value of 1.14. $^{238}U/^{226}Ra$ ratios for most of the 30 rock samples are higher than unity, reflecting a state of radioactive disequilibrium between U and its daughter, ^{226}Ra (TABLE 1). The disequilibrium state her is concerning to U-enrichment. The activity concentrations ratios of $238U/^{40}K$ were found to have a wide range from 0.46 to 2.34 Bq/kg, with an average value of 1.08 Bq/kg, which is almost equal to unity, the global ratio (UNSCEAR, 2000) [4]. Ratio of $^{232}Th/^{40}K$ ranged from 0.08 to 0.69 with an average value of 0.33. This ratio can be used as an indicator of the relative occurrence of these radionuclides.

The elemental ratio of eth/eu (in ppm) was varied from 0.26 (Sample No.29) to 2.23 (Sample No.1) with an average of 1.13. eth/eu ratio is an indicative for the relative depletion or enrichment of radioisotopes, (TABLE 1) eth/eu ratio for continental crust, varies from 3.84 to 4.2 [19,20]. The concentration ratios are higher than unity for most of the sampling sites, which shows low geochemical mobility of thorium [21]. The arithmetic mean of all studied rock samples (1.13) of eth/eu ratio is much lower than the Clark's value (3.5), which indicates U-enrichment in the rock samples in the studied area. The uranium and thorium concentration in the present study are lower than uranium and thorium concentration in the down crust value. The high concentrations results of 238U in these areas of study are due to the presence of phosphate and granite rocks with highly enriched with this radioactive nuclide and the weathering effects [22].

Radium equivalent (Raeq) and exposure rate

Radium equivalent (Raeq) index is a radiation hazard index used on a large scale. It is appropriate indicator to compare the specific activity of samples containing different concentrations of ^{226}Ra, ^{232}Th (228Ra) and ^{40}K. It is defined on the assumption that 10 Bq/kg of ^{226}Ra, 7 Bq/kg of 232Th and 130 Bq/kg of ^{40}K produce the same gamma dose rate. It was calculated as follows [23]:

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_{K} \quad (3)$$

Where CRa, Ck and CTh are the activity concentrations of ^{232}Th, ^{40}K and ^{226}Ra in Bq/kg. In this study the Raeq ranged between 116.80 and 449.40 Bq/kg, with a mean of 286.13 Bq/kg. It is concluded that for all the samples analyzed, the radium equivalent activity value is lower than the permissible limits of 370 Bq/kg, except for samples (3, 6, 8, 9, and 10). No regular trend in the variation of the terrestrial radioactivity has been observed from the study area. The exposure to ionizing radiations from natural sources happens as a result of the naturally happening radioactive components in the soil and rocks. A human body assimilates the majority of the radiation energy conveyed to it. External gamma dose rating due to the terrestrial sources is essential not only because it contributes considerably to the mass dose but also because of differences in the person dose related to this pathway. Their effects in the air can be expressed in terms of exposure rate or absorbed dose rate by using the conversion factors from radioelement concentrations in the samples to exposure rate or absorbed dose rate. The ground level exposure rate can be calculated from the apparent concentrations of K (%), AU (ppm), and ATh (ppm) using the expression [24,25]:

$$E\ (\mu Rh\text{-}1) = 1.505\ K\ (\%) + 0.653\ eU\ (ppm) + 0.287\ eTh\ (ppm) \quad (4)$$

The estimated exposure (μR/h) for the studied samples is listed in TABLE 2. In this study E (μR/h) of the samples were calculated by using Eq. (4). E (μR/h) of the rock samples for the different types and the locations from where they were collected are investigated in this study given in TABLE 2. The values of E (μR/h) ranged from (5.68 to 22.52) (μR/h), with the mean value of 16.15 (μR/h).

Absorbed and effective dose rate (D, $D_{4\pi}$, D_{eff}, D_{ex})

Absorbed dose rates due to γ-radiations in air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by [4]. Diversion factors used to count absorbed gamma dose rate (D) in air per unit activity concentration in Bq/kg (dry weight). Conversion factors used to compute absorbed gamma dose rate (D) in air per unit activity concentration in Bq/kg (dry weight) corresponds to 0.462n Gy/h for ^{226}Ra, 0.604 nGy/h for ^{232}Th and 0.042 nGy/h for ^{40}K. Therefore D can be calculated as follows [4]:

$$D = 0.462\ CRa + 0.604\ CTh + 0.0417\ CK \quad (5)$$

TABLE 1. **Activity concentration of 238U, ^{226}Ra, ^{232}Th, ^{40}K and radium equivalent doses, Ratio between ^{226}Ra, ^{232}Th and ^{40}K in activity concentration (Bq/kg) and (ppm). Ratio between ^{226}Ra, ^{232}Th and ^{40}K in activity concentration and (ppm).**

Sample	Location	C_U (Bq/Kg)	C_{Ra} (Bq/Kg)	C_{th} (Bq/Kg)	C_k (Bq/Kg)	Ra_{eq} (Bq/Kg)	e_{Th}/e_U	C_U/C_{Ra}	C_U/C_K	C_{Th}/C_K
1	L1	128.13 ± 17.45	126.07 ± 16.37	91.62 ± 10.95	269.04 ± 26.11	277.81	2.23	1.02	0.48	0.34
2		158.47 ± 17.34	175.62 ± 33.95	95.04 ± 12.21	195.80 ±24.87	326.61	1.87	0.90	0.81	0.49
3	L2	288.61 ± 38.71	249.75 ± 34.34	80.58 ± 6.90	223.52 ± 32.91	382.19	0.87	1.16	1.29	0.36
4		218.98 ± 24.34	217.08 ± 20.94	95.00 ± 12.20	210.78 ±19.22	369.17	1.35	1.01	1.04	0.45
5	L3	179.52 ± 19.53	131.57 ± 14.12	76.73 ± 5.49	208.46 ± 18.54	257.35	1.33	1.36	0.86	0.37
6		259.44 ± 28.50	303.16 ± 46.24	58.46 ± 6.21	174.65 ± 21.26	400.21	0.70	0.86	1.49	0.33
7	L4	281.51 ± 39.03	240.56 ± 30.57	80.39 ± 6.83	126.34 ± 15.27	365.24	0.89	1.17	2.23	0.64
8		229.67 ± 22.08	296.70 ± 53.59	91.35 ± 10.86	286.90 ± 31.29	449.42	1.24	0.77	0.80	0.32
9	L5	213.68 ± 24.59	287.71 ± 33.90	62.12 ± 3.13	132.26 ± 13.56	386.72	0.90	0.74	1.62	0.47
10		235.57 ± 28.05	286.61 ± 32.45	98.46 ± 13.47	278.89 ± 28.97	448.89	1.30	0.82	0.84	0.35
11	L6	189.95 ± 18.18	134.41 ± 12.95	91.35 ± 10.86	132.41 ± 13.51	275.23	1.50	1.41	1.43	0.69
12		177.82 ± 17.64	161.84 ± 21.71	68.27 ± 2.39	189.66 ± 23.09	274.07	1.19	1.10	0.94	0.36
13	L7	149.45 ± 9.99	125.51 ± 16.60	48.27 ± 4.95	192.96 ± 34.05	209.39	1.01	1.19	0.77	0.25
14		116.88 ± 20.34	107.89 ± 23.82	54.81 ± 3.55	188.74 ±32.82	200.80	1.46	1.08	0.62	0.29
15	L8	133.86 ± 13.35	125.87 ± 16.45	62.31 ± 4.20	202.82 ± 26.91	230.60	1.45	1.06	0.66	0.31
16		239.94 ± 26.68	143.95 ±19.04	38.27 ± 8.62	241.97 ± 28.26	217.30	0.50	1.67	0.99	0.16
17	L9	199.9 ± 17.67	98.51 ± 27.67	54.62 ± 2.63	150.80 ± 18.18	188.22	0.85	2.03	1.33	0.36
18		230.76 ± 31.62	215.78 ± 20.41	65.77 ± 5.47	147.90 ± 19.02	321.23	0.89	1.07	1.56	0.44
19	L10	285.71 ± 39.10	260.74 ± 38.84	54.81 ± 3.55	277.49 ± 28.56	360.48	0.60	1.10	1.03	0.20
20		259.74 ± 36.61	233.77 ± 27.78	51.16 ± 3.90	111.19 ± 19.67	315.48	0.61	1.11	2.34	0.46
21	L11	227.99 ± 18.20	206.99 ± 16.81	87.96 ± 9.61	253.24 ± 21.53	352.28	1.20	1.10	0.90	0.35
22		129.87 ± 16.85	116.88 ± 20.14	58.46 ± 4.21	112.23 ± 19.36	209.13	1.40	1.11	1.16	0.52
23	L12	108.89 ± 23.84	98.90 ± 27.51	51.16 ± 3.90	178.88 ± 26.04	185.83	1.46	1.10	0.61	0.29
24		199.6 ± 17.56	159.64 ± 22.61	91.35 ± 10.86	209.44 ± 18.83	306.40	1.42	1.25	0.95	0.44
25	L13	243.75 ± 25.11	224.78 ± 24.10	76.93 ± 5.56	174.65 ± 31.26	348.23	0.98	1.08	1.40	0.44
26		123.87 ± 18.95	116.88 ± 20.14	69.43 ± 2.81	266.90 ± 25.49	236.71	1.74	1.06	0.46	0.26

27	L14	129.99 ± 16.80	125.99 ± 16.40	57.31 ± 5.64	181.69 ± 30.78	221.93	1.37	1.03	0.72	0.32
28		97.92 ± 18.34	89.91 ± 21.20	36.54 ± 4.26	140.85 ± 21.06	153.01	1.16	1.09	0.70	0.26
29	L15	175.92 ± 25.67	161.93 ± 31.67	14.62 ± 5.31	188.01 ± 27.61	197.31	0.26	1.09	0.94	0.08
30		135.92 ± 34.33	99.55 ± 26.20	13.34 ± 3.77	131.45 ± 22.49	116.80	0.31	1.51	1.34	0.13
Min		97.92 ± 18.34	89.91 ± 21.20	13.34 ± 3.77	111.19 ± 19.67	116.8	0.26	0.74	0.46	0.08
Max		288.61 ± 38.71	303.16 ± 46.24	98.46 ± 13.47	286.90 ± 31.29	449.4	2.23	2.03	2.34	0.69
Mean + SD		191.71± 23.55	177.49 ± 25.62	65.88 ± 6.48	192.66 ± 23.19	286.13	1.13	1.14	1.08	0.33

TABLE 2. **Absorbed dose rate, indoor annual effective dose (mSv\y), outdoor annual effective dose (mSv\y), external annual effective dose, and ELCR (outdoor and indoor).**

Sample	**Location**	**E (μ R/h)**	**D (nGy/h)**	**$D_{4\pi}$ (10^{-8} Gy/h)**	**D_{eff} outdoor (mSv\y)**	**D_{eff} indoor (mSv/y)**	**$D_{eff\ (EX)}$ (mSv/y)**	**ELCR outdoor X10^{-3}**	**ELCR indoor X10^{-3}**	**ELCR Ex X10^{-3}**
1	L1	16.99	126.36	49.24	0.15	0.62	0.77	0.54	2.55	2.71
2		16.90	148.32	48.24	0.18	0.73	0.91	0.64	3.00	3.18
3	L2	22.48	174.75	56.57	0.21	0.86	1.07	0.75	2.89	3.75
4		19.32	168.08	53.90	0.21	0.82	1.03	0.72	2.01	3.61
5	L3	15.92	117.13	42.42	0.14	0.57	0.72	0.50	3.15	2.51
6		18.64	183.65	54.85	0.23	0.90	1.13	0.79	2.86	3.94
7	L4	22.52	166.33	46.84	0.20	0.82	1.02	0.71	3.53	3.57
8		19.98	205.77	68.55	0.25	1.01	1.26	0.88	3.04	4.42
9	L5	16.62	177.01	49.90	0.22	0.87	1.09	0.76	3.52	3.80
10		21.41	205.19	67.71	0.25	1.01	1.26	0.88	2.13	4.40
11	L6	18.12	124.35	37.77	0.15	0.61	0.76	0.53	2.15	2.67
12		16.20	125.07	42.78	0.15	0.61	0.77	0.54	1.65	2.68
13	L7	13.19	96.01	36.69	0.12	0.47	0.59	0.41	1.58	2.06
14		11.13	91.75	35.33	0.11	0.45	0.56	0.39	1.81	1.97
15	L8	12.78	105.31	39.45	0.13	0.52	0.65	0.45	1.72	2.26
16		16.50	100.36	41.72	0.12	0.49	0.62	0.43	1.47	2.15
17	L9	15.13	85.71	30.92	0.11	0.42	0.53	0.37	2.52	1.84
18		19.45	146.70	44.30	0.18	0.72	0.90	0.63	2.85	3.15
19	L10	21.91	166.07	59.22	0.20	0.81	1.02	0.71	2.48	3.56
20		17.83	144.40	40.97	0.18	0.71	0.89	0.62	2.76	3.10
21	L11	19.59	160.81	55.75	0.20	0.79	0.99	0.69	1.63	3.45
22		11.55	94.99	29.86	0.12	0.47	0.58	0.41	1.46	2.04
23	L12	10.29	84.92	33.03	0.10	0.42	0.52	0.36	2.39	1.82
24		18.51	139.22	47.33	0.17	0.68	0.85	0.60	2.73	2.99
25	L13	19.46	158.90	49.10	0.19	0.78	0.97	0.68	1.86	3.41
26		12.76	108.24	45.20	0.13	0.53	0.66	0.46	1.74	2.32
27	L14	11.80	101.37	36.91	0.12	0.50	0.62	0.44	1.20	2.18
28		8.54	70.10	26.78	0.09	0.34	0.43	0.30	1.57	1.50
29	L15	13.31	91.73	35.66	0.11	0.45	0.56	0.39	0.93	1.97
30		5.68	54.05	20.22	0.07	0.27	0.33	0.23	0.93	1.16
Min		5.68	54.05	20.22	0.07	0.27	0.33	0.23	0.93	1.16
Max		22.52	205.77	68.55	0.25	1.01	1.26	0.88	3.53	4.42
Mean + SD		16.15	130.76	44.24	0.16	0.64	0.80	0.56	2.20	2.81

Where: CTh, CRa and Ck are the activity concentrations of 232Th, 226Ra and ^{40}K in Bq/kg.

The absorbed dose rate in air enclitic by absolute thicknesses of soils can be counted according to the next formula (D4π) [26, 27]:

$$D4\pi = 0.104\ CRa + 0.130\ CTh + 0.09\ CK \quad (6)$$

Where D4π (10−8 Gy/h) refers to the total absorbed dose rate.

To rating annual effective doses, calculations must be taken off the transformation coefficient from the absorbed dose in air to an effective dose. The mean numerical values of those parameters modify with the age of the population and the environmental at the location considered. The annual effective dose rate Deff in (mSv/y) outdoor and indoor occupancy was calculated by the following formula [4]:

$$\text{Deff (mSv/y)}_{\text{out}} = D \times 24\ \text{hour} \times 365.25\ \text{days} \times 0.2 \times 0{:}7\ \text{Sv/Gy} \times 0{:}001 \quad (7)$$

$$\text{Deff (mSv/y)}_{\text{in}} = D \times 24\ \text{hour} \times 365.25\ \text{days} \times 0.8 \times 0{:}7\ \text{Sv/Gy} \times 0{:}001 \quad (8)$$

Whereas, D is dose rate in (nGy/h), (0.2, 0.8) are the outdoor and indoor occupancy factors and (0.7) was transformation coefficient from the absorbed dose in air to an effective dose received by adults in (Sv/Gy) [28]. The external annual effective dose is given by the equation (9):

$$D_{eff\,(EX)} = \left(\sum D_{eff\,(out)} + D_{eff\,(in)}\right)\ mSv\ /\ y \quad (9)$$

The absorbed dose and annual effective dose rates can be total calculated of samples are shown in TABLE 2. It is observed that the calculated absorbed dose rate varied from 54.05 to 205.77 nGy/h, with an average value of 130.76 nGy/h. The weighted mean value of 130.76 nGy/h represents 238% of the world average outdoor exposure due to terrestrial gamma radiation (55 nGy/h, according to UNSCEAR, 1993, 2000) [28]. Thus, the radioactive impact and the additional external radiation exposure for population due to rocks were not negligible, and consequently, the recorded value in the study area for most samples are important for health, which indicates high hazard effects to the people living there. The corresponding outdoor and indoor annual effective doses rate range from 0.07 to 0.25 mSv/y and 0.27 to 1.01 mSv/y with an average value of 0.16 and 0.64 mSv/y respectively. The similar type of direction is observed in all the samples and no steady direction in the different in the annual effective dose and absorbed dose rate is observed from the rock samples.

The calculated external annual effective dose varies from 0.33 to 1.26 mSv/y with an average value of 0.80 mSv/y and these results lie within the worldwide average values reported by UNSCEAR 2000 [4], although it remains within the dose criterion of 1 mSv/y recommended by ICRP [5]. In this work too, the least absorbed dose rate in the air surrounded by absolute thicknesses value of study area was found to be 20.22 (10−8 Gy/h), while the highest value was found to be 68.55 (10−8 Gy/h) for all rock samples. D4π (10−8 Gy/h) refers to the total absorbed dose rate. The International Commission on Radiological Protection (ICRP) [5] has recommended the annual effective dose equivalent limit of 1 mSv/y for the individual members of the public and 20 mSv/y for the radiation workers [29]. These results for mean annual effective dose are in the range of worldwide mean value.

Excess lifetime cancer risk (ELCR)

From annual effective dose value, calculated the excess lifetime cancer risk (ELCR) was using the following equation;

$$\text{ELCR)\ outdoor (= Deff (out)} \times \text{LE} \times \text{RF} \quad (10)$$

$$\text{ELCR) indoor (= Deff (in)} \times \text{LE} \times \text{RF} \quad (11)$$

where Deff (out) and Deff (in)are the external annual effective doses, LE life expectancy (70 years) and RF (Sv-1) is a fatal risk factor per Sievert, which is 0.05 as per ICRP-60 [30]. The (ELCR) for outdoor exposure, given in TABLE 2 ranged from 0.23×10^{-3} to 0.88×10^{-3} with an average value of 0.56×10^{-3}. For indoor exposure, it is 0.93×10^{-3} to 3.53×10^{-3} with an average of 2.20×10^{-3}. The total external ELCR ranges from 1.16×10^{-3} to 4.42×10^{-3} with an average value of 2.81×10^{-3}. Average ELCR for all samples is higher than the world average (0.29×10^{-3}) [31].

External and internal hazard indexes (Hex and Hin) for finite thickness of walls

External hazard index (Hex): (Hex) appear the external radiation exposure related with gamma irradiation from radionuclides of concern. Hex value must not exceed the maximum acceptable value than one in order to maintain the considerable danger. The

external hazard index definition (Hex) through [32]:

$$Hex = (CRa/370 + CTh/259 + CK/4810) \leq 1 \quad (12)$$

Internal hazard index (H_{in}): H_{in} is used to control the internal exposure to ^{222}Rn and its radioactive progeny [20]. The internal exposure to radon and its daughter products is quantified by the internal hazard index (H_{in}), [23] which is given by the following equation:

$$Hin = CRa/185 + CTh/259 + CK/4810 \leq 1 \quad (13)$$

Where: CTh, CRa and CK are the activity concentrations of 232Th, 226Ra and ^{40}K in Bq/kg.

The calculated external hazard values Hex are between 0.43 and 1.14 (TABLE 3). The mean value of the external hazard index (0.81) is lower than the recommended limit. Seven locations such as (L2, L4, L5, L10 and L11) exceed the recommended limit. This encroachment in these destinations is because of the higher concentration of radionuclides. Also, mean relative help to the gamma-index due to the ^{238}U is higher go after by the contributions due to ^{232}Th and ^{40}K. The values for Hin in this study ranged between 0.70 and 1.92 with a mean value of 1.33. The internal hazard index Hin exceeds the permissible value in the most rock samples; this means that ^{222}Rn and its progeny plays a significant role in the expected internal hazard due to radiation from the samples under consideration. The values derived from the second model of (Hex) for finite thickness of walls ranged between 0.16 and 0.61, with a mean value of 0. 39. Since these values are lower than unity. Therefore, according to the Radiation Protection 112 report [33,34], soil from these regions is safe and can be used as a construction material without posing any significant radiological threat to the population.

Representative level index (I γr): A hazard index so-called agent level index is studied by using the formula of [35]:

$$I\gamma r = (CRa/150 + CTh/100 + CK/1500) \quad (14)$$

Where: CTh, CRa and CK are the specific activities (Bq/kg) of ^{232}Th, ^{226}Ra and ^{40}K. The value of these indexes must be less than unity in order to keep the radiation hazard insignificant. The index Iγr is associated with the annual dose due to the spare external gamma radiation caused by a superficial material. Values of index $I\gamma r \leq 1$ correspond to 0.3 mSv/y, while $I\gamma r \leq 3$ correspond to 1 mSv/y. Thus, the activity concentration index should be used only as a screening tool for identifying materials which might be of concern to be used. According to this dose criterion, materials with $I\gamma r \leq 3$ should be avoided, since these values correspond to dose rates higher than 1 mSv/y [34] which is the highest value of dose rate in air recommended for population [4,28].

The calculated Iγr values for all the samples are presented in TABLE 3. The values range from 0.80 to 3.08 with an average of 1.97. The calculated values for most samples were higher than the international values ($I\gamma r < 1$), which corresponds to an annual effective dose < 0.3 mSv/y. The recorded averages of the radiological hazards (Iγr) in the locality were higher than the average of rock global average [4] except for the S30 samples which is lower than unity. TABLE 3 shows the obtained values for S8 and S10 are $I\gamma r \geq 3$, which is a very far from the acceptable values indicating a very high radiation risk in the study area.

TABLE 3. **Representative level index Iγ, Iα, External hazard index (H_{ex}), and Internal hazard index (H_{in})**

Sample	Location	$I_{\gamma r}$	$I_{\alpha r}$	H_{ex}	H_{in}	Hex for finite thickness walls
1	L1	1.94	0.63	0.76	1.10	0.37
2		2.25	0.88	0.84	1.26	0.44
3	L2	2.62	1.25	1.14	1.92	0.52
4		2.54	1.09	1.00	1.59	0.50
5	L3	1.78	0.66	0.82	1.31	0.35
6		2.72	1.52	0.96	1.66	0.54
7	L4	2.49	1.20	1.10	1.86	0.49
8		3.08	1.48	1.03	1.65	0.61
9	L5	2.63	1.44	0.84	1.42	0.52
10		3.08	1.43	1.07	1.71	0.61
11	L6	1.90	0.67	0.89	1.41	0.37
12		1.89	0.81	0.78	1.26	0.37
13	L7	1.45	0.63	0.63	1.03	0.28
14		1.39	0.54	0.57	0.88	0.27

15	L8	1.60	0.63	0.64	1.01	0.31
16		1.50	0.72	0.85	1.50	0.29
17	L9	1.30	0.49	0.78	1.32	0.25
18		2.19	1.08	0.91	1.53	0.43
19	L10	2.47	1.30	1.04	1.81	0.49
20		2.14	1.17	0.92	1.62	0.43
21	L11	2.43	1.03	1.01	1.62	0.48
22		1.44	0.58	0.60	0.95	0.28
23	L12	1.29	0.49	0.53	0.82	0.25
24		2.12	0.80	0.94	1.48	0.41
25	L13	2.38	1.12	0.99	1.65	0.47
26		1.65	0.58	0.66	0.99	0.32
27	L14	1.53	0.63	0.61	0.96	0.30
28		1.06	0.45	0.43	0.70	0.21
29	L15	1.35	0.81	0.57	1.05	0.27
30		0.80	0.45	0.44	0.81	0.16
Min		0.80	0.45	0.43	0.70	0.16
Max		3.08	1.52	1. 14	1.92	0.61
Mean + SD		1.97	0.89	0.81	1.33	0.39

Representative level index (Iα r): Several Iα have been suggested to value the exposure level due to radon inhalation product from rock materials [29]. The alpha index was determined using the following formula:

$$I\alpha r = CRa / 200 \ (Bq/ kg) \tag{15}$$

Where: CRa (Bq/kg) is the activity concentration of ^{226}Ra assumed in equilibrium with 238U. The recommended exception and higher level of ^{226}Ra activity concentrations in building materials are 100 and 200 Bq/kg, respectively, as suggested by [34]. These considerations are reflected in the alpha index. The recommended upper limit concentration of ^{226}Ra is 200 Bq/kg, for which Iαr =1. The mean computed Iαr values for the studied samples are given in TABLE 3 for the different rock types and the locations where they were collected. The values of Iαr ranged from (0.45 to 1.52), with the mean value of 0.89. The alpha-indexes were lower than unity as it is seen in TABLE 3. Thus radon inhalation from investigated rock samples was below the upper level and the study area is safe from the view of environmental radiation hazard.

Correlation studies

To find the rate the existence of these nuclides jointly at a particular place, correlation studies were completed between the collections of radionuclides like ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K. A search was carried out to detect the presence of a statistically significant correlation between the measured radionuclides in the present rock samples. In fact knowing the conditions of the secular equilibrium was necessary in order to make the correct assumptions to assess the dose [36,37]. In this context and considering all samples, regarding (FIG. 2) which, shows linear regression of the activity concentrations of ^{238}U versus ^{226}Ra for all samples. As can be seen in (FIG. 2) concentrations of ^{238}U and ^{226}Ra showed a statistically significant. Since the P-value in the ANOVA table is less than 0.05, there is a statistically significant relationship between activity concentration of ^{226}Ra and activity concentration of ^{238}U at the 95.0% confidence level. The R-Squared statistic indicates that the adjusted model explains 67.591% of the variability in activity concentration of ^{226}Ra. The correlation coefficient is equal to 0.822138, indicating a moderately strong relationship between the variables. This value can be used to construct prediction limits for new observations by selecting the forecasts option from the text menu. Furthermore, the good correlation coefficient of the $^{238}U/^{226}Ra$ activity ratio indicates a common source of the parent materials [38].

Other correlations among measured radionuclide were also investigated which may provide information on the relative depletion or enrichment of the natural radioelement's. FIG. 3 also shows a moderate correlation between (^{232}Th, ^{238}U,) with N = 30 and R2 = 8.28803% which means that the two elements accompanied each other. The correlation coefficient equals 0.287819, indicating a relatively weak relationship between the variables, which disagreed with a previous study in rock samples obtained at Red Sea coast Area [39,40]. In general, most sites have the ratio ^{232}Th /^{238}U higher than one and this agrees with the reported mean activity concentration ratios of ^{232}Th /^{238}U in sandstone and shale areas, which are 1.7 and 2.5, respectively [41]. This coincides with the same ratio derived from FIG. 3, which is equal to 1.13.

On the other hand, weak correlations were also observed between (^{238}U, ^{40}K) and (^{232}Th, ^{40}K) in the collected samples, with N = 30 and R^2 = 2.09366% and 17.9579% of the variability in ^{40}K, respectively. The correlation coefficient equals 0.144695 and 0.423767

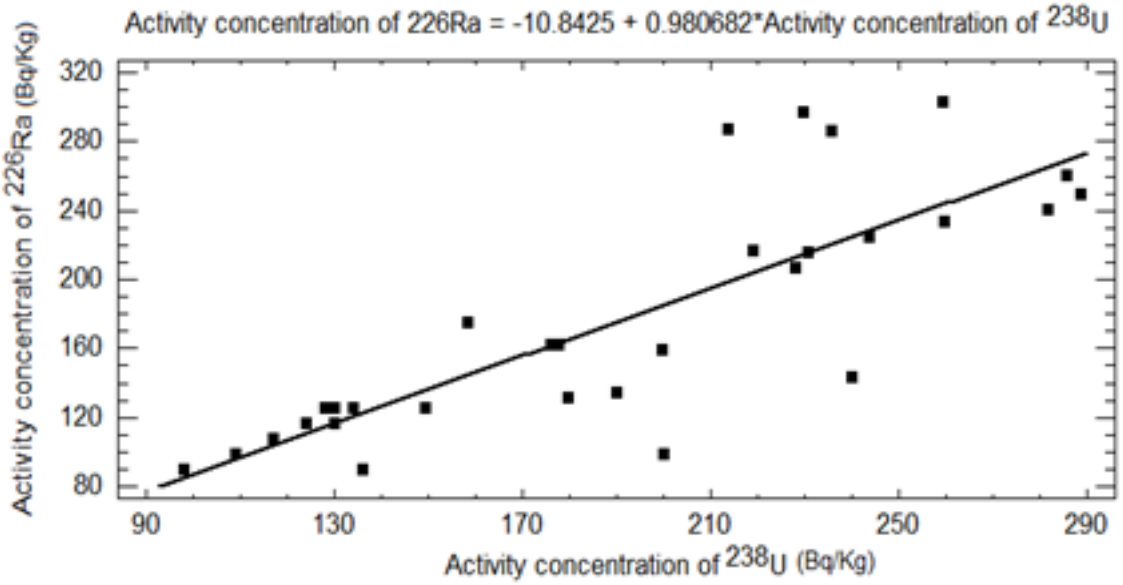

FIG. 2. **Linear regression of the activity concentration of ^{238}U versus ^{226}Ra for all rock samples under study.**

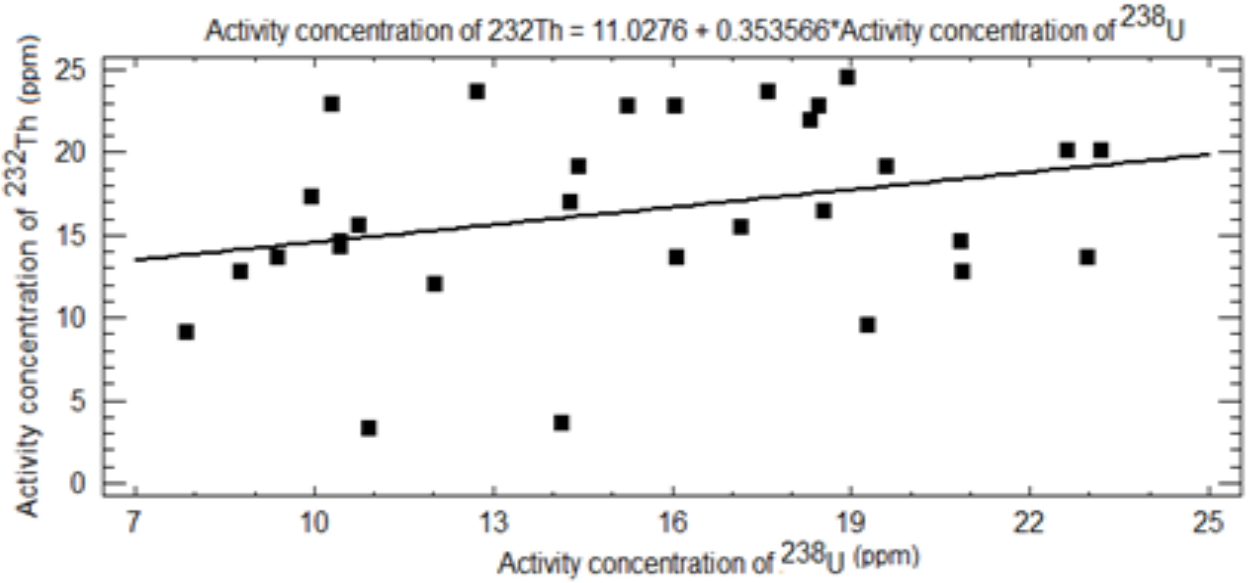

FIG. 3. **Linear regression of the activity concentration of ^{238}U (ppm) versus ^{232}Th (ppm) for all rock samples under study.**

respectively, indicating a relatively weak relationship between the variables (FIG. 4 and 5). Weak correlation may be due to rock processes that affect differently the mobility of the two radionuclides. It appears in most figures that the number of points less than thirty because points having the same activity concentration in more than one sample (with little difference) coincidence with each other. The levels of detected radionuclide in all samples indicated wide variations and this may be attributed to the diversity of formations and textures of the rock in the studied area. However, the variability among levels of ^{238}U and levels ^{232}Th are frequently associated with the type of geological minerals. Therefore, detailed mineralogical investigations are needed for more interpretations.

Atmospheric ^{222}Rn concentrations can be evaluated by measuring ^{214}Pb and ^{214}Bi by gamma spectrometry [42]. The radon emanation coefficient of samples was calculated based on two γ- measurements.

Radon emanation coefficient and radon mass exhalation

The first measurement was load out right away after sealing of samples, and the second measurement was carried out after attainment of secular equilibrium between radon and its short-lived decay daughters (after 30 days). This particular method may be appropriate where there is temperature inversion leading to very little or no vertical movement of air masses. Based on these measurements, the

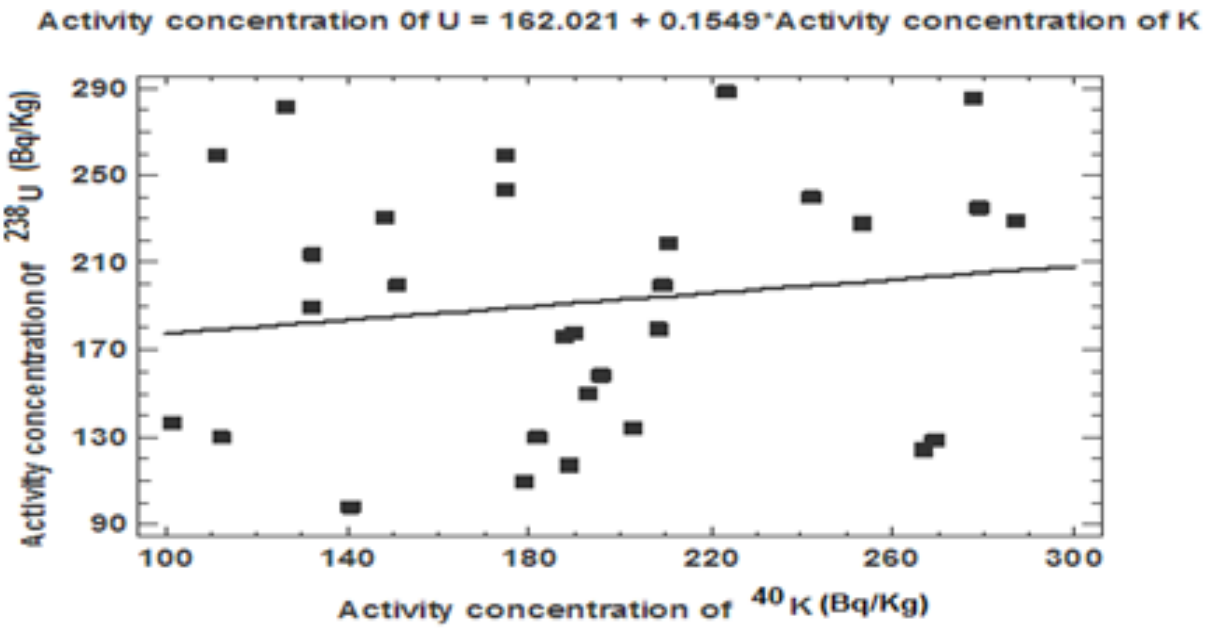

FIG.4. **Linear regression of the activity concentration of ^{238}U (Bq/kg) versus ^{40}K (Bq/kg) for all rock samples under study.**

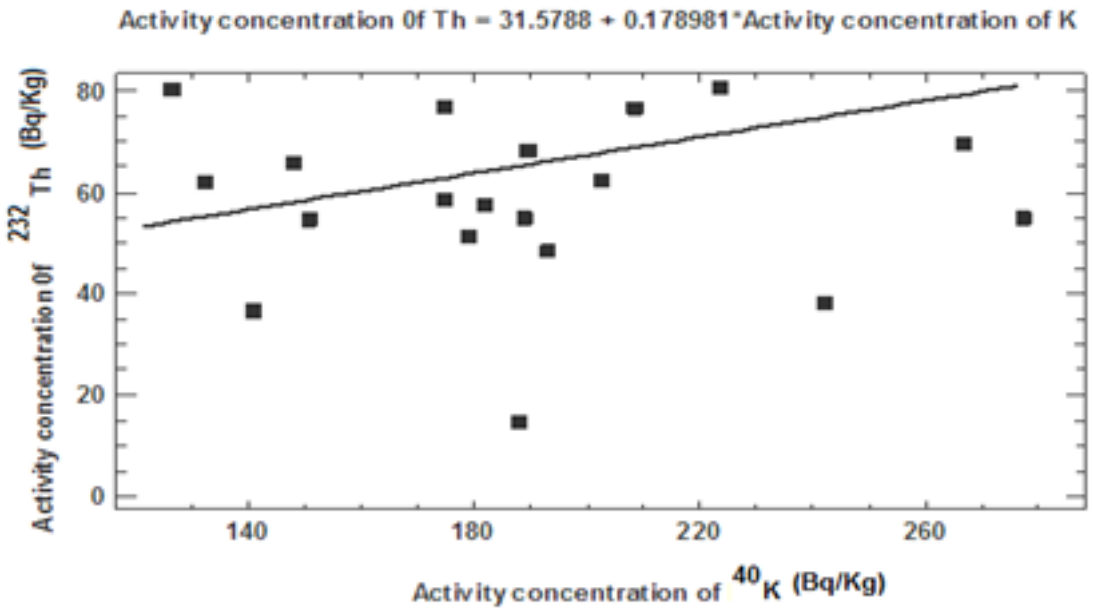

FIG.5. **Linear regression of the activity concentration of ^{232}Th (Bq/kg) versus ^{40}K (Bq/kg) for all rock samples under study**

radon emanation coefficient was calculated according to the following expression:

$$Rn_{EC} = \frac{N}{(N_o + N)} \quad (17)$$

Where, RnEC is the radon emanation coefficient, No is the net count rate of ^{222}Rn at the time of sealing the sample container, N is the net count rate of ^{222}Rn emanated at the radioactive equilibrium with 226Ra and its progeny. The mass exhalation rate of radon is the product of the emanation coefficient of radon (ERa) and production rate of radon [43]. The mass exhalation rate (ERn in Bq/kg•s) is determined using the following equation:

$$Ex = CRa \times RnEC \times \lambda Rn \quad (18)$$

Where CRa is the specific activity of ^{226}Ra (in Bq/kg) and λRn is the decay constant of ^{222}Rn (λRn = 2.1 × 10−6 s−1). The radon emanation coefficient CRn and ^{222}Rn mass exhalation rate of rock samples under the current study have been shown in TABLE 4. It

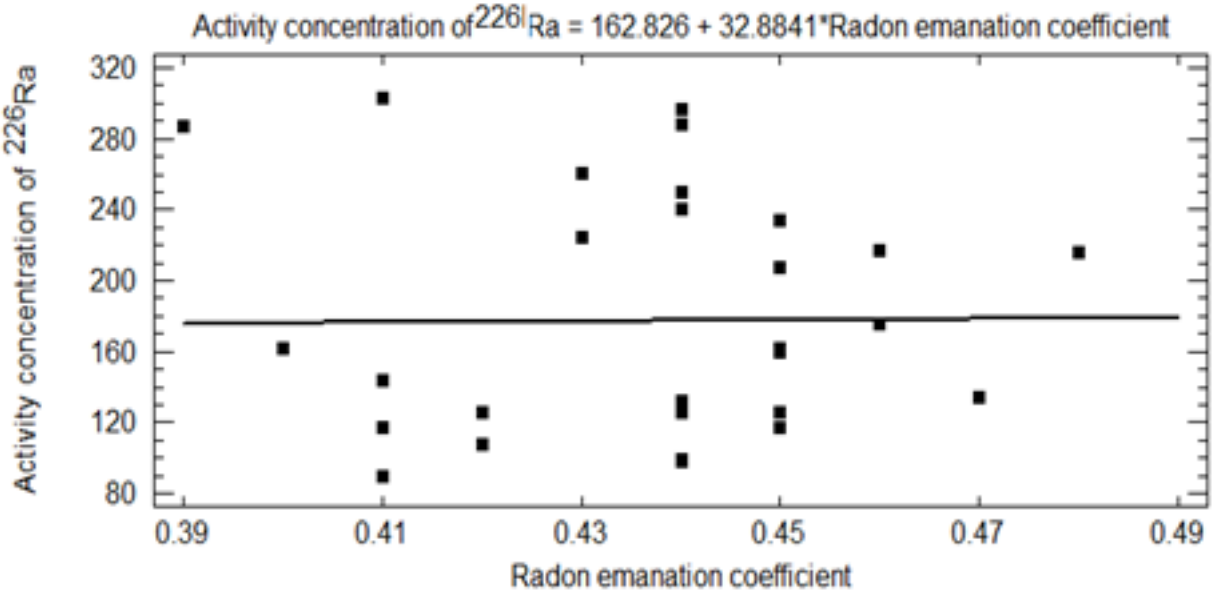

FIG. 6. **Correlations between the specific activity of ^{226}Ra and ^{222}Rn radon emanation coefficient.**

is clear that the values of the emanation coefficient and the ^{222}Rn and exhalation rate for all samples under investigation was ranged from 0.39 to 0.48 and 83.52 to 265.42 (μBq/kg•s), respectively. This variation in radon concentration confirms an earlier the position that the uranium content in the earth crust is different at different locations. FIG. 6 shows a weak correlation between the specific the activity of ^{226}Ra and ^{222}Rn radon emanation coefficient with (R^2 = 0. 0102137, N = 30) for rock samples, which means that ^{222}Rn and ^{226}Ra not accompanied each other and that the individual result for any one of the radionuclide concentration is not a good predictor of the concentration of the other. FIG. 7 shows a moderately strong correlation between radon mass exhalation rate (E_{Rn}) and activity of ^{226}Ra (C_{Ra}) with correlation coefficient is equal to 0.797704, R^2 = 63.6331, which means that E_{Rn} and C_{Ra} accompanies with each other.

Conclusion

Radioactivity levels of the environment depend on geological aspects of rock samples, where they are found in different concentrations. The physical and chemical change plays their part in the redistribution of radionuclides in different rock types which were subjected to these alteration processes. This distribution of radionuclide was reflecting its effect on the environment. Thirty kinds of rocks

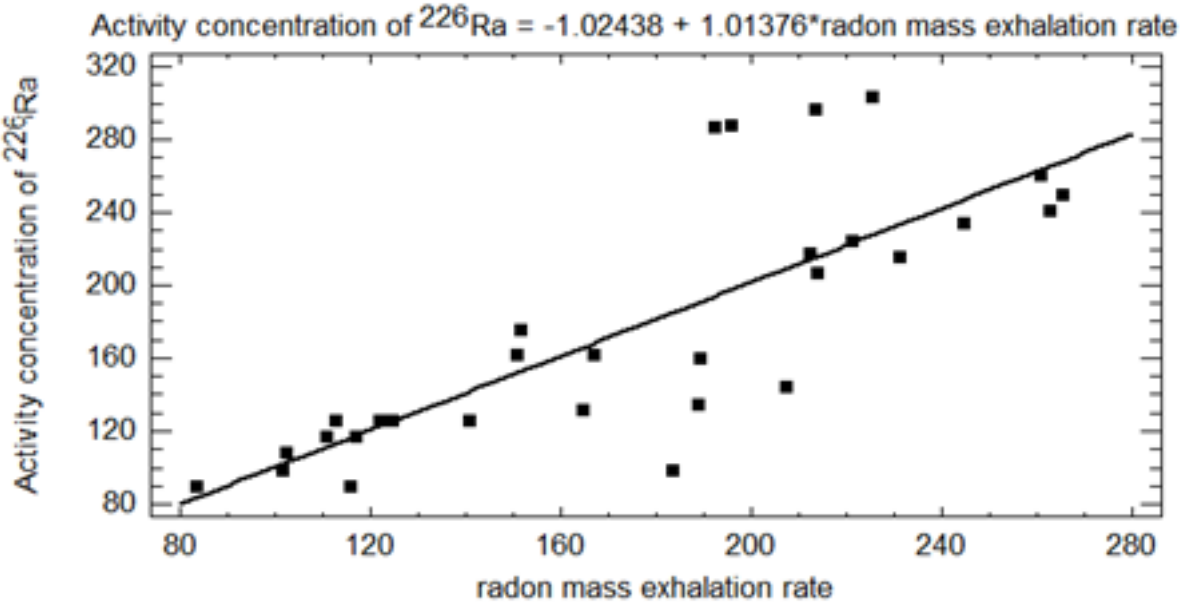

FIG. 7. **Mass exhalation rate for ^{222}Rn verses specific activity of ^{226}Ra for all samples under investigation.**

TABLE 4.**The specific activity of ^{226}Ra, activity of ^{238}U before and after sealing time, the emanation coefficient and the radon mass exhalation from the rock samples used in the study area.**

Sample	Location	C_{Ra} (Bq/Kg)	238U-series (Bq/kg) before C0	^{238}U-series (Bq/kg) after C	Emanation coefficient of Radon CRn	Mass exhalation rate for ^{222}Rn (µBq/kg·s)
1	L1	128.13 ± 17.45	107.46	77.38	0.42	112.64
2		158.47 ± 17.34	138.19	115.41	0.46	151.45
3	L2	288.61 ± 38.71	180.5	140.63	0.44	265.42
4		218.98 ± 24.34	176.7	151.56	0.46	212.32
5	L3	179.52 ± 19.53	106.5	82.53	0.44	164.59
6		259.44 ± 28.50	276.71	195.49	0.41	225.56
7	L4	281.51 ± 39.03	172.31	137.81	0.44	262.70
8		229.67 ± 22.08	218.95	174.1	0.44	213.64
9	L5	213.68 ± 24.59	231.49	179.35	0.44	195.89
10		235.57 ± 28.05	273.74	174.01	0.39	192.26
11	L6	189.95 ± 18.18	101.7	91.35	0.47	188.75
12		177.82 ± 17.64	153.77	104.19	0.40	150.83
13	L7	149.45 ± 9.99	97.58	79.44	0.45	140.84
14		116.88 ± 20.34	42.51	30.47	0.42	102.48
15	L8	133.86 ± 13.35	92.5	73.63	0.44	124.59
16		239.94 ± 26.68	121.49	84.83	0.41	207.17
17	L9	199.9 ± 17.67	69.81	54.24	0.44	183.55
18		230.76 ± 31.62	144.42	131.56	0.48	231.01
19	L10	285.71 ± 39.10	185.79	142.83	0.43	260.78
20		259.74 ± 36.61	177.72	144.4	0.45	244.52
21	L11	227.99 ± 18.20	160.85	129.86	0.45	213.87
22		129.87 ± 16.85	107.61	73.5	0.41	110.68
23	L12	108.89 ± 23.84	88.23	70.44	0.44	101.52
24		199.6 ± 17.56	139.35	114.88	0.45	189.41
25	L13	243.75 ± 25.11	177.05	134.56	0.43	221.04
26		123.87 ± 18.95	96.21	78.78	0.45	117.11
27	L14	129.99 ± 16.80	106.08	85.78	0.45	122.05
28		97.92 ± 18.34	90.83	62.14	0.41	83.52
29	L15	175.92 ± 25.67	128.93	106.34	0.45	166.98
30		135.92 ± 34.33	17.61	12.04	0.41	115.91
Min		97.92 ± 18.34	17.61	12.04	0.39	83.52
Max		288.61 ± 38.71	276.71	195.49	0.48	265.42
Mean + SD		191.71± 23.55	139.42	107.78	0.44	175.77

were collected from natural mountains in Red Sea coast Egypt, considered as the most popular ones, and were calculated for their natural radioactivity in order to value the radiological impact when they are used as building materials. The activities concentration of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K of most the rocks samples exceed the average level of these radionuclides in regular 288.61 ± 38.71, 303.16 ± 46.24, 98.46 ± 13.47 and 286.90 ± 31.29 Bq/kg respectively, while the concentration of ^{40}K is lower than world figures. The corresponding absorbed dose rate from all those radionuclides also higher than the average value of 55 nGy/h from these terrestrial radionuclides in regular rocks, and the annual effective dose is based on the standard room model, less than the dose limit of 1 mSv/y for all samples understudies, and according to the dose criteria recommended by European Commission Radiation Protection 112, in 1999. ^{238}U/^{226}Ra ratios for most of the 30 rock samples are higher than unity, reflecting a state of radioactive disequilibrium between U and its daughter, ^{226}Ra. The disequilibrium state is linked to high U-enrichment. Thus radon inhalation from investigated rock samples was below the upper level and the study area is safe from the view of environmental radiation hazard.

Furthermore, the good correlation coefficient of the ^{238}U/^{226}Ra activity ratio indicates a common source of the parent materials. Other correlations among measured radionuclide were also investigated between (^{238}U, ^{40}K) and (^{232}Th, ^{40}K) in the collected samples. The correlation coefficient indicating was a relatively weak relationship between the variables. Weak correlation may be due to rock processes that affect differently the mobility of the two radionuclides. In this figures the number of points lower than thirty because this points having the same activity concentration in more than one sample (with little difference) coincidence with each other. The

calculation from all samples values are, in general, comparable to the corresponding ones obtained from other studies in Egypt, and they all fall within the average worldwide ranges. These results can be given basic values for distribution of natural radionuclides in the area and will be used as reference information for determining any future changes. The levels of detected radionuclide in all samples indicated wide variations and this may be attributed to the diversity of formations and textures of the rock in the studied area. However, the variability among levels of ^{238}U and levels ^{232}Th are frequently associated with the type of geological minerals. Therefore, detailed mineralogical investigations are needed for more interpretations.

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