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Evaluation of environmental radioactivity impacts and its associated radiological hazards with monitoring of radon-222 in rock and sand samples collected from some mountains and valleys in sinai area, Egypt

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ABSTRACT

Determination of the natural radionuclide (^{226}Ra, ^{238}U, ^{232}Th & ^{40}K) contents of soil and rock samples collected from various geological formations in some mountains and valleys in Sinai, Egypt has been carried out using gamma spectrometric techniques. The total average concentrations of radionuclides ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K were 51.37, 50.02, 19.34, and 286.66 Bq kg^{-1}, respectively. Correlations made among these radionuclides prove there is no existence of secular equilibrium in the investigated soils. The total average absorbed dose rate in the study areas is found to be 47.08 nGy h^{-1}, whereas the indoor and outdoor annual effective dose equivalent has an average value of 0.23 and 0.06 mSv y^{-1} respectively. The external and internal hazard indices and the radium equivalent activity associated with the investigated soils do not exceed the permissible limits. Moreover, the radio-elemental concentrations of uranium, thorium and potassium, evaluated for the various geological features in the study areas were calculated to indicate whether relative depletion/enrichment of radioisotopes had occurred. The results of the present study were discussed and compared with internationally recommended values.

In the present study some mathematical equations models are used to estimate the activity concentrations of ^{222}Rn in the air with the activities of ^{226}Ra that contents in vegetables, as well as the doses rates that results from the vegetables consumption and from inhalation of radon gas were considered. The range of ^{226}Ra activity was found from 11.15 ± 0.96 to 135.85 ±11.68 Bq/Kg. The concentrations of radon gas in the air was determined as well as the activity concentrations of ^{222}Rn in vegetables.

KEYWORDS

Radioactivity concentration;
Absorbed dose;
The activity concentrations of ^{222}Rn in the air and soil;
Gamma spectrometer;
The external and internal hazard indices;
Radium equivalent activity.

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INTRODUCTION

The external radiation exposure arises mainly due to cosmic rays and from terrestrial radionuclides. Gamma radiation arising from naturally occurring radioactive materials (NORMS), such as ^{238}U, ^{235}U, ^{232}Th and ^{40}K series and their decay products, existing at trace levels in all ground formations, is the principal external source of radiation to the human body[36]. Uranium has two primary isotopes ^{238}U (half-life: 4.5×10^9 years) and ^{235}U (half-life: 7.04×10^8 years) which occur in the proportion 99.27% (^{238}U) and 0.72% (^{235}U), respectively.

Both exhibit long and complex decay series. ^{232}Th (half-life: 1.4×10^{10} years) is another naturally occurring radionuclide with a complex decay chain. Potassium has three isotopes (^{39}K, ^{40}K and ^{41}K), ^{40}K (half-life: 1.28×10^9 years) being the only radioactive isotope having an abundance of 0.012% of total potassium. Absorbed dose rate in air, at sea level, from cosmic radiation was measured about 30 nGy/h for the Southern hemisphere[37]. The levels of terrestrial environmental radiation in an area are related to the geological condition of that area and also to the content of Th, U and K in the rocks from which the soils originate[26] Hence, the levels of natural environmental radioactivity and the associated external exposure due to gamma radiation are observed to be at different levels in the soil of different regions in the world[12].

The presence of natural occurring radioactive material (NORM) in the terrestrial composition of the natural background is dependent on the geological composition of the soil and rocks. Therefore, systematic and accurate measurements of the radioactivity level in soils are essential for understanding changes in the natural radiation background as a function of geographical location and time[21, 33, 6]. Radionuclides are present in rocks in varying amounts, and they are easily mobilized into the environment. Radioactivity in soil results from the rock from which they were derived. The distributions of naturally occurring radionuclide depend on the distribution of rocks from which they originate and the processes which result to their removal from the soil and migrate them. Therefore, the natural environmental radioactivity mainly depends on geological and geophysical conditions. The concentration of natural radionuclide in the rock varies considerably depending on the rock formation and litho logic character[1, 3, 4].

Even though radon was identified at the beginning of the 20th century, it was not until late in 1960 that a correlation between radon (daughter) exposure and lung cancer among uranium miners was established. A new analysis of published results also shows a slightly increased risk of lung cancer from household radon, consistent with the level of risk that has been estimated based on the studies of underground miners[23].

^{222}Rn is a daughter of ^{226}Ra and is in turn derived from the longer-lived antecedent ^{238}U. Since most materials contain ^{238}U, therefore, any material can be a potential radon emitter. However, some materials have higher concentrations of ^{238}U and ^{226}Ra such as alum shale and black shale. Radon is produced in the ground from the radioactive decay of uranium-238. Being a gas, it can move through the soil and is constantly released into outdoor air. Because of the natural pressure differential that exists between indoor and outdoor air, radon from the soil is preferentially 'sucked' into buildings through any gaps or imperfections that may exist in the foundations. Radon can be hazardous in the indoor environment as in some homes and workplaces it may build up to high concentrations. Quantification of background levels of radio nuclides is necessary to evaluate the potential environmental risk, to determine the boundary of a contaminated area and to establish it's clean up level[35].

The objective of this study is to determine the activity concentrations of natural radionuclides (^{226}Ra, ^{238}U, ^{232}Th and ^{40}K) in rock and soil samples collected from some Mountains and valleys in Sinai area of Egypt, and estimate the radiological hazard associated with them.

MATERIALS AND METHODS

Geology of the study area

Sinai, the triangular-shaped peninsula of Egypt, is situated between Asia and Africa. The separation of the two continents caused the form and geographical shape of Sinai the way it looks today. Sinai is approx. 380 km long (north - south) and 210 km wide (west - east). The surface area has an extension of

61.000 km², the coasts are stretching about 600 km on the west and on the east. On the western part there is the Gulf of Suez (with the Suez channel) and the eastern part of Sinai brings up the much deeper Gulf of Aqaba. The sea in the Gulf of Suez measures approx. 80 meters only, while the profile of the Gulf of Aqaba goes down to approx. 1.830 meters. The latter is a part of the big land rift that extends until Kenya. Big seismic activity and the tremendous eruptive phenomena have given Sinai its characteristic looks. The highest mountains are the Gebel Musa (Moses' mountain) with 2,285 meters, and the Sinai's highest mountain Mount St. Catherine (Gebel Kathrina) with 2,642 meters.

The peninsula of Sinai may be divided into three geological districts, named from the granitic, limestone, and sandstone rocks of which they are composed. The central part of South Sinai is a mountainous region of Precambrian igneous and metamorphic rock, which includes Egypt's highest peak, Saint Catherine's Mountain. Mount Sinai's rocks were formed in the late stage of the Arabian-Nubian Shield's (ANS) evolution. Mount Sinai displays a ring complex[31] that consists of alkaline granites intruded into diverse rock types, including volcanic. The granites range in composition from syeno granite to alkali feldspar granite. The volcanic rocks are alkaline to per alkaline and they are represented by sub aerial flows and eruptions and sub volcanic porphyry. Generally, the nature of the exposed rocks in Mount Sinai indicates that they originated from different depths. North of the Sinai Mountains there is a series of valleys in which the Nubian sandstone is clearly exposed. At the centre of the peninsula is the Tih Plateau which is mainly composed of Cretaceous and Eocene limestone. A wide plain with sand dunes flanks the Mediterranean coastline. The southern part has been affected by many tectonic movements forming spectacular folds and dark mineral-rich veins.

Sample collection and preparation

A total of 21 surface rocks and soil samples were collected from 18 mountains and 3 valleys in Sinai region. Multiple samples were collected from 0-5 cm depth at a location and homogenized to make one representative sample. As shown in Figure 1. A total of 21 rock and soil samples, each about 2 kg in weight, were crushed, homogenized and sieved to about 100 mesh by a crushing machine. The samples were then placed for drying at 105 ºC for 24 h to ensure that moisture is completely removed. The samples were transferred to beakers polyethylene container of 250 cm^3 volume. Each sample was carefully sealed for four weeks to reach secular equilibrium between ^{232}Th and ^{238}U and its short lived daughter products[5].

Instrumentation and calibration

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 30% counting efficiency. The resolution of this spectrometer was 1.89 keV at 1332 keV γ -rays of ^{60}Co. The efficiency calibration of the gamma-ray spectrometer was performed with the radionuclide specific efficiency method in order to avoid any uncertainty in gamma-ray intensities as well as the influence of coincidence summation and self-absorption effects of the emitting gamma photons. A set of high quality certified reference materials (IAEA, RG-set) was used, with densities similar to the collected samples after pulverization. Measurements were carried out in Marinelli. The obtained spectra were analyzed with the use of Canberra Genie 2000 software version 3.0. The measurement duration was up to 80 000 sec. The activity concentrations of ^{238}U and ^{232}Th were calculated assuming secular equilibrium with their decay products. The gamma ray transitions of energies 63.3 keV (^{234} Th), 186.1 keV (^{226}Ra), 295.1, 352.1 keV (^{214}Pb) and 609.3,768.4, 934.1 and 1120.3 keV (^{214}Bi) were used to determine the concentration of the ^{238}U that were in secular equilibrium with ^{226}Ra following four weeks storage, due to the difficulty of direct measurement caused by the overlap with ^{235}U at 185.7 keV. Gamma ray lines at 185.70 and 186.1 keV, they include two different sources: ^{235}U ($I\gamma$ = 53.1 %), ^{226}Ra ($I\gamma$ = 3.3%) respectively. These lines can be used assuming the hypothesis that the ^{226}Ra is in secular equilibrium with ^{238}U and the isotopic ratio between ^{235}U and ^{238}U is 0.72 %. The gamma-ray lines at 209.4, 338.4, 462.1, 911.2, and 966.6 keV (^{228}Ac), and 583.1 keV (^{208}Tl) were used to determine the concentration of the ^{232}Th series. The 1460 keV gamma-ray transition was used to de-

termine the concentration of ^{40}K. The activity levels of the samples obtained for ^{226}Ra, ^{214}Pb, ^{214}Bi, ^{228}Ac, ^{208}Tl and ^{40}K are expressed in Bq/kg.

In order to determine the background distribution due to naturally occurring radionuclide in the environment around the detector, an empty polystyrene container was counted in the same manner as the samples. The activity concentrations were calculated after measurement and subtraction of the background. The activities were determined from measuring their respective decay daughters[27]. The activity concentrations were calculated from the intensity of each line taking into account the mass of the sample, the branching ratios of the ã-decay, the time of counting and the efficiencies of the detector[19, 13]. The activity concentrations of the investigated samples were calculated from equation (1):

$$A = (CPS)_{net} / I. E_{ff}. M \quad (1)$$

Where A is the activity concentration in Bq/kg, $(cps)_{net}$ is the (count per second). I is the intensity of the ³-line in a radionuclide, E_{ff} is the measured efficiency for each ³-line observed and M is the mass of the sample in kilograms. The correction for the contribution of ^{232}Th via its daughter nuclide ^{228}Ac (1459.2 keV peak) to the 1460.8 keV peaks of ^{40}K was made according to[22]:

$$\text{The error in } ^{40}K \text{ activity (\%)} = 9.3(A_{Th}/A_K) \quad (2)$$

A_{Th} and A_K are the activity concentration of ^{232}Th and ^{40}K, respectively, in Bq·kg^{-1}, as in TABLE (3).

TABLE 1 gives the energies used to determine the concentrations of different radionuclide's and their yields[9]. The background spectrum was used to correct the areas of gamma rays of measured isotopes. The lowest limits of detection (LLD) of the measuring system, which is required to estimate a minimum detection level for appropriate determination of radionuclides using analytical technique in each sample, were obtained following environmental measurement laboratory procedure using the expression[8, 2].

$$LLD = 4.66\ S_b / \zeta \cdot I\gamma, \quad (3)$$

Where S_b is the standard error of the net back-ground count rate in the spectrum of the radionuclide; ¶ is the counting efficiency; I³ is the abundance of gamma emission per radioactive decay. The LLD of a measuring system measures its operating capability without the influence of the sample. The LLD values for ^{40}K, ^{232}Th and ^{238}U are obtained in TABLE (2).

RESULTS AND DISCUSSION

Activity Concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K in (Bq/Kg) and (PPm), and radium equivalent activity

The mean values of measured activity concentrations of selected radionuclides of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K in rock and soil samples from all 21 sites in Sinai are shown in TABLE 3. The activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K are in the range of 11.15± 0.096 – 135.85 ± 11.68 Bq kg^{-1}, 13.20 ±1.13 –98.30 ± 8.45Bq kg^{-1}, 4.70 ± 0.40 - 45.56 ±3.91 and 46.95 ± 4.03–573.11± 49.28 Bq kg^{-1} with a mean value of 51.37, 50.02, 19.34 and 286.66 Bq kg^{-1}, respectively. The results shown in TABLE 3 also indicate that the mean value of ^{232}Th (19.34 Bq kg^{-1}) < ^{226}Ra (51.37 Bq kg^{-1}) < ^{40}K (286.66 Bq kg^{-1}). Clearly, the

TABLE 1 : Photo energies and branching ratios for different radionuclides

Nuclide	Energy KeV	Photon per disintegration %	Nuclide	Energy KeV	Photon per disintegration %	Nuclide	Energy KeV	Photon per disintegration %
	^{238}U series			^{232}Th series			^{235}U series	
^{234}Th	63.3	7	^{228}Ac	209.4	4.1	^{235}U	143.8	10.5
^{226}Ra	186.1	3.3		338.4	12.4		163.3	4.7
^{214}Pb	295.1	19.2		462.1	4.6		185.7	53.1
	352.1	37.1		911.2	29.1		205.3	4.7
^{214}Bi	609.3	46.1		966.6	23.2			
	768.4	5.1				^{40}K	1460.8	10.7
	934.1	3.2	^{208}Ti	583.1	30.9			
	1120.3	15						

TABLE 2 : The lowest limit of detection (LLD) for the radionuclide ^{40}K, ^{238}U and ^{232}Th

(LLD) Lowest detection limit (Bq/kg)	Nuclide
9.36	^{40}K
1.32	^{238}U
1.35	^{232}Th

average results 51.37, 50.02, 19.34 and 286.66 (Bq/Kg) samples are higher than the permissible levels (35, 35) for radium, uranium, and less than (30, 400 Bq/kg)for thorium and potassium.

As it was mentioned above for all samples A_{Th}/A_U ratio was calculated. This ratio varies in the range

TABLE 3 : Mean Activity concentrations of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K (Bq.kg^{-1}), P- Factor (e_U/e_{Ra}), ($^{232}Th/^{238}U$) Ratio and the error in ^{40}K for all samples under study

Location	A_{Ra} (Bq/Kg)	A_U (Bq/Kg)	A_{Th} (Bq/Kg)	A_K (Bq/Kg)	A_{Th}/A_U	P-Factor e_U/e_{Ra}	The error in ^{40}K activity (%)
Jebel Katherina	118.9 ± 10.17	98.30 ± 8.45	45.56 ± 3.91	573.11 ± 49.28	0.46	0.83	4.31
Jebel Umm Shaumar	62.25 ± 5.35	77.30 ± 6.64	34.96 ± 3.00	332.10 ± 28.56	0.45	1.24	4.21
Jebel Serbal	85.25 ± 7.33	81.00 ± 6.96	40.60 ± 3.49	256.50 ± 22.05	0.50	0.95	4.66
Jebel Abbas Basha	77.33 ± 6.65	66.40 ± 5.71	27.08 ± 2.32	297.67 ± 25.59	0.41	0.86	3.79
Jebel el Bab	135.85 ± 11.68	81.90 ± 7.04	41.90 ± 3.60	441.65 ± 37.98	0.51	0.60	4.76
Jebel Bab el Donya	55.75 ± 4.79	32.80 ± 2.82	15.26 ± 1.31	231.62 ± 19.91	0.47	0.59	4.33
Jebel Naja	45.80 ± 3.94	77.50 ± 6.66	29.92 ± 2.57	494.06 ± 42.48	0.39	1.69	3.59
Jebel el Banat	32.25 ± 2.77	66.30 ± 5.70	11.50 ± 0.98	518.78 ± 44.61	0.17	2.06	1.61
Jabal ?ur Sina	34.65 ± 2.98	58.80 ± 5.05	16.24 ± 1.39	46.95 ± 4.03	0.28	1.70	2.57
Jabal Musa	49.40 ± 4.25	55.50 ± 4.77	8.30 ± 0.71	103.29 ± 8.88	0.15	1.12	1.39
Jabal Thabt	11.15 ± 0.96	13.20 ± 1.13	6.90 ± 0.59	209.71 ± 18.03	0.52	1.18	4.86
Jabal Sabbah	48.20 ± 4.15	55.40 ± 4.76	13.68 ± 1.17	225.36 ± 19.38	0.25	1.15	2.30
Jabal Mileihis	23.50 ± 2.02	22.10 ± 1.90	6.84 ± 0.58	287.64 ± 24.73	0.31	0.94	2.88
Jabal Matamir	29.95 ± 2.58	43.90 ± 3.77	19.32 ± 1.66	462.60 ± 39.78	0.44	1.47	4.09
Jabal Guna	90.40 ± 7.77	44.00 ± 3.78	11.50 ± 0.98	343.82 ± 29.56	0.26	0.49	2.43
Jabal Barqa	48.20 ± 4.15	44.30 ± 3.80	24.96 ± 2.14	274.34 ± 23.59	0.56	0.92	5.24
Jabal Maharum	33.45 ± 2.88	55.20 ± 4.74	19.02 ± 1.63	46.95 ± 4.03	0.34	1.65	3.20
Jabal Fuga	21.60 ± 1.86	13.54 ± 1.16	5.56 ± 0.47	408.50 ± 35.13	0.41	0.63	3.82
Wadi El Shallal	16.55 ± 1.42	14.30 ± 1.22	7.14 ± 0.61	237.80 ± 20.45	0.50	0.86	4.64
Wadi Sahu	24.90 ± 2.14	22.12 ± 1.90	4.70 ± 0.40	58.50 ± 5.03	0.21	0.89	1.98
Wadi Firan	34.10 ± 2.93	26.60 ± 2.28	15.30 ± 1.31	168.86 ± 14.52	0.58	0.78	5.35
Mean	51.37	50.02	19.34	286.66	0.36	1.08	3.36
Worldwide*	35	35	30	400		= 1	

***UNSCEAR (2000)**.

from 0.15 to 0.58 with an overall mean value of 0.36. The highest value refers to the Wadi Firan, whereas the lowest value was noted in the sample collected from Jabal Musa. Obtained A_{Th}/A_U concentration ratios are very low, strongly lower than data published in literature concerning rocks and sands collected from different location in Sinai[30]. Finally, the $^{226}Ra/^{40}K$ ratio ranged from 0.05 to 0.33 (mean 0.1). Also; in this case the ratios for various soils are consistent with each other.

Values of $A_{Ra,}$ $A_{u,}$ A_{Th} and A_K are the activities concentration in Bq kg^{-1}, were converted to elemental activity concentration of eU, eTh and eRa, in ppm, as well as K, in %,, using the conversion factor given by Polish Central Laboratory for Radiological Protection[24]. The specific parent activity of a sample containing 1 ppm, by weight, of U is 12.35 Bq kg^{-1}, 1 ppm of ^{226}Ra is 11.1 Bq kg^{-1}, 1 ppm of ^{232}Th is 4.06 Bq kg^{-1} and 1% of ^{40}K is 313 Bq kg^{-1}. These data were used for calculation of some radiological parameters to estimate the environmental radioactivity impacts of the radionuclides.

The equilibrium factor, which was defined by[14] as P-factor and expressed as the ratio between radiometrically measured equivalent uranium and equivalent radium (e_U/e_{Ra}) was calculated in all rock and soil samples. This factor is more or less than unity indicating state of disequilibrium, while P-equal unity indicates the state of equilibrium. From the estimated values, which illustrated in TABLE 3, the mean values of the (e_U/e_{Ra}) elemental activity ratio, obtained in this study, were varied from 0.49 in Jabal Guna soils to 2.06 in Jabal el Banat soils, with mean value 1.08. In addition to that, as the specific activities of ^{238}U and ^{226}Ra in the samples under investigation were at the environmental level, the slight deviation from the equilibrium condition in uranium series could also be attributed to the separation between ^{230}Th (parent of ^{226}Ra) and ^{238}U. Thus, on the base of the above presented results, one can state that in the examined soils a state of radioactive equilibrium within the uranium series is most probable. However, as the soil samples examined in this study are taken from the top soil layer, further detailed studies, especially measurements of depth profiles, are needed for a better assessment of the secular equilibrium conditions in the uranium series. We note that the composition (Rocks & Soil) of Sinai area in equilibrium state within experimental error, except (Jebel Umm Shaumar, Jebel Naja, Jebel el Banat, Jabal lur Sina, Jabal Musa, Jabal Thabt, Jabal Matamir, Jabal Maharum and Jabal Sabbah), p- factor was >1. While p- factor >1 for (Jebel Katherina, Jebel Abbas Basha, Jebel el Bab, Jebel Bab el Donya, Jabal Fuga, Wadi El Shallal, Wadi Sahu and Wadi Firan).

To compare the specific activity of the samples containing ^{226}Ra, ^{232}Th and ^{40}K, the radium equivalent index ($^{226}Ra_{eq}$) has been used to obtain the total amount of these activities[5, 35].

$$^{226}Ra_{eq} = 1.43A_{Th} + A_{Ra} + 0.077A_K \quad (4)$$

A_{Ra} A_{Th} and A_K are the activities concentration in (Bq/kg) of ^{226}Ra (U series), ^{232}Th and ^{40}K respectively. The ^{232}Th and 13 Bq kg^{-1} of ^{40}K produce the same gamma-ray dose. As reference, the permissible dose limit for public which is recommended by[17] are 1.5 mSv y^{-1} or equivalent to 370 Bq kg^{-1}. The radium equivalent activities of samples under investigation were calculated on the basis of the above equation and are shown in TABLE 4. For all soil samples under investigation, The radium equivalent values are lower than the acceptable value of 370 Bq kg^{-1}[38] ranging from 33.35 Bq kg^{-1} to 207.58 Bq kg^{-1}.

Correlation studies

In order to find the extent the existence of these radioactive nuclides together at a particular place, correlation studies were performed between the combinations 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 and soil samples. In fact, the knowledge of the secular equilibrium conditions is necessary in order to make correct assumptions for the dose assessments[29].

In this context and considering all samples, regarding Figure 2. which shows linear regression of the activity concentrations of ^{238}U versus ^{226}Ra for all samples. As can be seen in Figure 2, concentrations of ^{238}U and ^{226}Ra showed a statistically significant positive correlation ($P < 0.05$ there is a statistically significant relationship between Ra and U at the 95.0% confidence level. The R-Squared statistic indicates that the model as fitted explains

TABLE 4 : The values of radium equivalent (Bq/kg), elemental activity concentration of eU, eTh and eRa, in ppm, K, in %, and external, internal hazards for all samples under study

H_{in}	H_{ex}	A_k (%)	A_{th} (ppm)	A_U (ppm)	A_{Ra} (ppm)	Ra_{eq} (Bq/Kg)	Location
0.93	0.61	1.83	11.25	7.90	9.50	207.58	Jebel Katherina
0.54	0.37	1.06	8.64	6.21	5.00	152.86	Jebel Umm Shaumar
0.67	0.44	0.82	10.03	6.51	6.85	158.81	Jebel Serbal
0.58	0.38	0.95	6.69	5.33	6.21	128.04	Jebel Abbas Basha
0.99	0.62	1.41	10.35	6.58	10.91	175.82	Jebel el Bab
0.41	0.26	0.74	3.77	2.63	4.48	72.46	Jebel Bab el Donya
0.47	0.34	1.58	7.39	6.23	3.68	158.33	Jebel Naja
0.33	0.24	1.66	2.84	5.33	2.59	122.69	Jebel el Banat
0.26	0.17	0.15	4.01	4.72	2.78	85.64	Jabal ?ur Sina
0.32	0.19	0.33	2.05	4.46	3.97	75.32	Jabal Musa
0.13	0.10	0.67	1.7	1.06	0.90	39.21	Jabal Thabt
0.36	0.23	0.72	3.38	4.45	3.87	92.32	Jabal Sabbah
0.21	0.15	0.92	1.69	1.78	1.89	54.03	Jabal Mileihis
0.33	0.25	1.48	4.77	3.53	2.41	107.15	Jabal Matamir
0.60	0.36	1.10	2.84	3.53	7.26	86.92	Jabal Guna
0.41	0.28	0.88	6.17	3.56	3.87	101.12	Jabal Barqa
0.26	0.17	0.15	4.7	4.43	2.69	86.01	Jabal Maharum
0.22	0.16	1.31	1.37	1.09	1.74	52.95	Jabal Fuga
0.17	0.12	0.76	1.76	1.15	1.33	42.82	Wadi El Shallal
0.16	0.10	0.19	1.16	1.78	2.00	33.35	Wadi Sahu
0.28	0.19	0.54	3.78	2.14	2.74	61.48	Wadi Firan
0.41	0.27	0.93	4.45	3.82	4.13	96.83	Mean
1 =	1 =	1.3	7.4	2.8	2.8	370	Worldwide*

***UNSCEAR (2000)**.

54.8612% of the variability in Ra. The correlation coefficient equals 0.740684, indicating a moderately strong relationship between the variables. Furthermore, the good correlation coefficient of the $^{238}U/^{226}Ra$ activity ratio indicates a common source from the parent materials[11].

The software used has been Statgraphics version 17.1.06. The output shows the results of fitting a linear model to describe the relationship between ^{238}U and ^{226}Ra. The equation of the fitted model is:

$^{238}U = 20.8417 + 0.568061*^{226}Ra$

Other correlations among measured radionuclide were also investigated. The correlation between the activity concentrations of ^{238}U and ^{232}Th was strong, since the P-value is less than 0.05; there is a statistically significant relationship between ^{232}Th and ^{238}U at the 95.0% confidence level. The R-Squared statistic indicates that the model as fitted explains 73.0193% of the variability in ^{238}Th. The correlation coefficient equals 0.854514, indicating a strong relationship between the variables, which disagreed with a previous study in soil samples obtained at Wadi Sahu Area[30].

On the other hand, weak correlations were also observed between ^{238}U and ^{40}K in the collected samples, since the P-value is greater or equal to 0.05, there is not a statistically significant relationship between ^{40}K and ^{238}U at the 95.0% or higher confidence level. The R-Squared statistic indicates that the model as fitted explains 17.1876% of the variability in ^{40}K. The correlation coefficient equals 0.414579, indicating a relatively weak relationship between the variables. Similarly, weak correlations were also observed between ^{232}Th and ^{40}K. Although the correlation coefficient equal to

0.454129, pointing to the presence of relatively weak relationship between variables, but the P-value less than 0.05, there is a statistically significant relationship between Th and K at the confidence level of 95.0%. R-Squared statistic indicates that the model as fitted explains 20.6233% of the variation in Th. Weak correlation may be due to soil processes that affect differently the mobility of the two radionuclides.

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 soil in the studied area. The soil in most of studied valleys is layer plates of clay that resulted from runoff of rain water while other samples from mountains were mainly sands of different grain sizes and colors. 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.

Radiological hazard indices

Estimation of the absorbed gamma dose rate

The absorbed dose rate at one meter above the ground due to the radioactivity in the samples are calculated using the following equation

$$D_R = K_k A_k + K_{Th} A_{Th} + K_{Ra} A_{Ra} \quad (5)$$

Where D_R is the absorbed dose rate (nGy/h), also K_k, K_{Th} and K_{Ra} is the conversion factors (or dose rate coefficients) expressed in (nGy.hr^{-1} per Bq.kg^{-1}) for Potassium (0.043), Thorium (0.662) and Radium (0.427), respectively[16].

TABLE 5 shows the lowest absorbed dose rate

TABLE 5 : Absorbed dose rate D_R (nGy/h), The annual effective dose equivalent (D_{eff}) and activity indices (I_γ, $I_\pm$) for all samples under study

I_α	I_γ	D_{eff} outdoor (mSv\y)	D_{eff} indoor (mSv\y)	D_R (nGy|\h)	Location
0.49	1.63	0.12	0.48	97.61	Jebel Katherina
0.39	0.99	0.09	0.35	71.27	Jebel Umm Shaumar
0.41	1.15	0.09	0.36	73.33	Jebel Serbal
0.33	0.98	0.07	0.29	59.91	Jebel Abbas Basha
0.41	1.62	0.10	0.40	82.27	Jebel el Bab
0.16	0.68	0.04	0.17	34.29	Jebel Bab el Donya
0.39	0.93	0.09	0.37	74.99	Jebel Naja
0.33	0.68	0.07	0.29	59.41	Jebel el Banat
0.29	0.42	0.05	0.19	39.21	Jabal ?ur Sina
0.28	0.48	0.04	0.17	35.10	Jabal Musa
0.07	0.28	0.02	0.09	19.13	Jabal Thabt
0.28	0.61	0.05	0.21	43.49	Jabal Sabbah
0.11	0.42	0.03	0.13	26.45	Jabal Mileihis
0.22	0.70	0.06	0.25	51.57	Jabal Matamir
0.22	0.95	0.05	0.21	41.81	Jabal Guna
0.22	0.75	0.06	0.23	47.41	Jabal Barqa
0.28	0.44	0.05	0.19	39.27	Jabal Maharum
0.07	0.47	0.03	0.13	26.74	Jabal Fuga
0.07	0.34	0.03	0.10	20.96	Wadi El Shallal
0.11	0.25	0.02	0.08	15.58	Wadi Sahu
0.13	0.49	0.04	0.14	28.83	Wadi Firan
0.25	0.73	0.06	0.23	47.08	Mean
1 =	1 =	0.3 - 1.0	0.3 - 1.0	55	Worldwide*

***UNSCEAR (2000)**.

was 15.58 nGy/h for the soil represented by Wadi Sahu sample, while the highest dose rate was 97.61 nGy/h for the soil represented by sample of Jabal Katherina. The published maximal admissible dose rate is 55nGy/h. The values determined for Samples (Jebel Katherina, Jebel Umm Shaumar, Jebel Serbal, Jebel Abbas Basha, Jebel el Bab,Jebel Bab el Donya and Jebel el Banat) in the current work is higher than the worldwide value.

The annual effective dose equivalent (D_{eff})

The annual effective dose equivalent received by a member due to soil radioactivity has been calculated from the absorbed dose rate by applying the dose conversion factor of 0.7 Sv/Gy and the occupancy factor for outdoor and indoor as 0.2(5/24) and 0.8(19/24), respectively[39] using the following equations:

D_{eff} (Outdoor) (mSv/y) = (Absorbed dose)

nGy/h x 8760h x 0.7 Sv/Gy x 0.2 x10^{-6} (6)

D_{eff} (Indoor) (mSv/y) = (Absorbed dose)

nGy/h x 8760h x 0.7 Sv/Gy x 0.8x10^{-6} (7)

TABLE 5 shows the absorbed dose rate D_R (nGy h^{-1}) and the annual effective dose D_{eff} (mSv y^{-1}) estimated for all the examined samples. The annual effective dose limit was considered to be 1 mSv. Some samples have ^{40}K activity concentrations higher than the regular soil values, for example Jebele Katherina with activity concentrations of 573.11Bq/Kg. Moreover, Jebele Katherina, Jebel Umm Shaumar, Jebel Serbal, Jebel Abbas Basha, Jebel el Bab and Jebel Naja has ^{232}Th, ^{238}U and ^{226}Ra, activity concentrations higher than of the regular soil concentrations.

Finally, an attempt to find a relation between terrestrial radiation doses in air outdoors and the nature of the rock, studying dose contributions of each natural series, ^{232}Th and ^{238}U,and ^{40}K that occur with different concentrations on rock and soil composition. For rocks and soil, the highest dose rate is 97.61 nGy h^{-1} (0.12 mSv y^{-1}) and was registered for Jebele Katherina, where the relative weight to dose is 573.11± 49.28 Bq/Kg for ^{40}K, 98.30 ± 8.45 Bq/Kg for ^{238}U series and 45.56 ± 3.91 Bq/Kg for ^{232}Th series. The lowest dose rate that registered was 15.58 nGy h^{-1}(0.02 mSv y^{-1}) for Wadi Sahu,where the relative weight to dose is 58.50 ± 5.03 Bq/Kg for ^{40}K, 22.12 ±1.90 Bq/Kg for ^{238}U series and 4.70 ± 0.40 Bq/Kg for ^{232}Th series. The world average annual effective dose equivalent (D_{eff}) from outdoor terrestrial gamma radiation is 70 ìSv/year[11]. Therefore, the obtained values from this preliminary study are all lower than the accepted, average worldwide value, except for Samples (Jebel Katherina, Jebel Umm Shaumar, Jebel Serbal, Jebel Abbas Basha, Jebel el Bab,Jebel Bab el Donya and Jebel el Banat) in the current work is higher than the worldwide value.

The calculated outdoor and indoor D_{eff} values are given in TABLE (5). The minimum, the maximum and the average values for outdoor are 0.02 mSv/y, 0.12 mSv/y and 0.06 mSv/y, respectively and the corresponding indoor values are 0.08 mSv/y, 0.48 mSv/y and 0.23 mSv/y respectively.

External and internal hazard index

The external hazard index (H_{ex}) is given by a model proposed by[20] as:

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_K/4810 < 1 \quad (8)$$

The internal exposure to ^{222}Rn and its radioactive progeny is controlled by internal hazard index, (H_{in}) which is given by.

$$H_{in} = A_{Ra}/185 + A_{Th}/259 + A_K/4810 < 1 \quad (9)$$

A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively. The mean external and internal radiation hazard indices H_{ex} and H_{in} are 0.27and 0.41 for all studied samples respectively, TABLE 4. As can be seen, the average values of the radiation hazard indices do not, in general, exceed the permissible recommended limits, indicating that the hazardous effects of these radiations are negligible.

Activity Indices {Gamma-index (I_γ) and Alpha Index ($I_\pm$)}

A number of indices dealing with the assessment of the external and internal radiations originating from building materials and gamma concentration indices have been proposed by several investigators[34, 25]. In this study, the gamma-index was calculated as proposed by the European Commission[10]:

$$I\gamma = A_{Ra}/150 + A_{Th}/100 + A_K/1500 \quad (10)$$

A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq·kg^{-1}, respectively. The mean values of I_γ calculated from the measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K are presented in TABLE 5 for different all samples and all the regions

from where they were collected. The calculated values of I_γ for the studied samples varied in the range between 0.25 –1.63. The mean calculated values of I_γ for the studied samples values (0.73) varied in the range between Wadi Sahu and Jebele Katherina. While the values of I_α for the studied samples are less than unity except for Jebel Katherina, Jebel Serbal and Jebel el Bab as its value reaches 1.63, 1.15 and 1.62 (TABLE 5),which were higher than the critical value of unity. $I_\gamma < 1$ corresponds to a dose creation of 1 mSv y^{-1}, while $I_\gamma < 0.5$ corresponds to 0.3 mSv y^{-1}. Depending on the dose criterion and the manner in which it used, and how much it safe for human to carry out their activities in the area, regulatory control should be considered for materials that give rise to doses between 0.3 mSv and 1 mSv per year.

So far, several alpha indices have been proposed to assess the exposure level due to radon inhalation originating from soil materials European Commission[10]. The alpha index was determined using the following formula:

$$I_\gamma = A_{Ra}/200 \ (Bq\ kg^{-1}) \quad (11)$$

A_{Ra} (Bq kg^{-1}) is the activity concentration of ^{226}Ra assumed in equilibrium with ^{238}U. The recommended exemption and upper level of ^{226}Ra activity concentrations in soil are 100 and 200 Bq kg^{-1}, respectively, as suggested by[18]. These considerations are reflected in the alpha index. The recommended upper limit concentration of ^{226}Ra is 200 Bq kg^{-1}, for which $I_\gamma = 1$. The mean computed I_γ values for the studied samples are given in TABLE 5 for the different rock and soil samples and the regions where they were collected. The values of I_γ ranged from (0.07 to 0.49), with the mean value of 0.25.

Evaluation of ^{222}Rn

As shown in TABLE 3, in most samples under investigation, it was found that the activity concentration of ^{238}U and ^{232}Th significantly elevated which due to the natural state and creation of soil. The radioactive radon gas that arises from the disintegration of ^{238}U and ^{232}Th in the earth´s crust is considered the main source of exposure to ionizing radiation for humans that representing 40% of the annual accumulated dose[36]. Therefore, in the present study some of mathematical equations models are used to estimate the activity concentrations of ^{222}Rn in the air with the activities of ^{226}Ra that contents in vegetables, as well as the annual effective dose that results from the vegetables consumption and from inhalation of radon gas were considered.

Determination the activity concentration of ^{226}Ra inside the soil

The magnitude of the concentration of ^{226}Ra inside the soil was calculated by[7]:

$$CRa(n) = C(E_n) - B(E_n)/ m.f.t.P(E_n) \quad (12)$$

Where: n : is the number of soil sample, 1,2,3…..etc; CRa(n) : is the radioactive concentration of ^{226}Ra in soil sample (n) in (Bq/Kg); $C(E_n)$: is the net ã-counts above continuum at the characteristic energy (E_n); $B(E_n)$: is the background counts at (E_n); m : is the mass of the sample in (Kg); f : is the branching ratio of the γ-emission at the energy considered; t : is the measuring live time in (sec); $P(E_n)$: is the absolute efficiency at energy (E_n).

Determination of the radioactive concentration of ^{222}Rn in air

The formula that used to measuring the radioactive concentration of ^{222}Rn in air as follows[28]:

Firstly it must be estimate the radioactive concentration of ^{222}Rn inside the soil samples by;

$$Gs(n) = F_r . p . CRa(n) \quad (13)$$

Where; Gs (n): concentration of radon gas inside the soil for sample (n) in (Bq/m^3); F_r : the constant of emission of ^{222}Rn from the soil that is equal to (o.1); ñ : is the soil density that is equal to (1800 Kg/m^3); CRa(n) : is the radioactive concentration of ^{226}Ra in soil sample (n) in (Bq/Kg).

Now we can calculate the concentration of ^{222}Rn in the air by equation:

$$Ca(n) = Gs(n) \ (d_{soil} / D_{air})^{1/2} \quad (14)$$

Where; Ca (n): is the concentration of ^{222}Rn in the air for sample (n) in (Bq/m^3); Gs (n) : concentration of radon gas inside the soil for sample (n) in (Bq/m^3); d_{soil} : is the diffusion rate constant of Rn^{222} in the soil ($o.5 \times 10^{-4}$ m^2/sec); D_{air} : is the diffusion rate constant of Rn^{222} in the air (5 m^2/sec).

Determination the concentration of ^{226}Ra in vegetables

Expense of radioactivity in vegetables was determined by using the following equation[28]:

$$C_P = A_n . CRa(n) \quad (15)$$

Where; C_P: is the concentration of ^{226}Ra in vegetables in (Bq/Kg); A_n: is the transfer coefficient of ^{226}Ra from soil to Vegetables that is equal to (o.o4)[15]; CRa (n): is the radioactive concentration of ^{226}Ra in soil sample (n) in (Bq/Kg).

Determination the annual effective dose resulting from inhalation of radon gas or vegetables consumption

The annual effective dose that coming from inhalation of radon gas and vegetables consumption was determined by using the below equation[18]:

$$Hp = Cp . Ip . DCF \quad (16)$$

Where; Hp: is the annual effective dose resulting from inhalation of radon gas or vegetables consumption in (sv/y); Cp: is the concentration of ^{226}Ra in vegetables (Bq/Kg) or the concentration of ^{222}Rn in the air (Bq/m^3); Ip : is the amount of consumption of vegetables in year (90Kg/y) and for air outside the home (1600 m^3/y)[15]; DCF: is the dose conversion coefficient: for ^{226}Ra equal to (2.8×10^{-7} sv/Bq) and for ^{222}Rn equal to (1.3×10^{-9} sv/Bq),[28].

TABLE 6 represents the estimated activity concentration of ^{222}Rn in soil and in air with its annual effective dose resulting from consumption of vegetables (^{226}Ra) and inhalation of ^{222}Rn gas (μsv/y) respectively. Results of radon concentrations in air, revealed relatively high radon levels, ranging from 6.35 to 77.33 Bq/m^3; with mean value 28.31 Bq/m^3.

The data of the annual effective dose from the vegetables consumption and inhalation of radon gas that given in Table 6 within the allowed limits that equal (1msv/y)[20, 34] in all regions that selected in the present search. Additionally, high radon concentrations to some extent in air may be explained by assuming the presence of uranium-bearing rocks.

It is worth mentioning, due to the high concentration of radium higher than the world average in most of the samples under study, particularly in Jebel Katherina, Jebel Serbal, Jebel el Bab and Jabal Guna samples, the radon concentration in the soil and in the air were also high, this led to higher effective dose rate in these samples, this is due to the natural state and creation of soil.

CONCLUSION

The activity levels and distribution of natural terrestrial radionuclides of ^{226}Ra, ^{238}U, ^{232}Th, and ^{40}K were measured by gamma-ray spectrometry system for surface soil and rock samples collected from populated areas in Sinai, Egypt. The activity and elemental concentrations of thorium, uranium and potassium in the studied soils are found to be normal. In soils at Sinai region, the uranium and radium contents exceeded values typical of the upper part of the Earth's crust due to the existence of phosphate rocks. In this context, a comparison of median activities of ^{226}Ra, ^{232}Th and ^{40}K indicates that ^{226}Ra is the dominant gamma-emitting source in the soil. Moreover; correlations made among measured radionuclides prove the existence of secular equilibrium in the investigated soils.

A search was carried out to detect the presence of a statistically significant correlation between the measured radionuclides in the present rock and soil samples. In fact, the knowledge of the secular equilibrium conditions is necessary in order to make correct assumptions for the dose assessments. Linear regression of the activity concentrations of ^{238}U versus ^{226}Ra for all samples indicating a moderately strong relationship between the variables.

The obtained values of natural radioactivity and ^{3}-absorbed dose rates due to the activity concentrations of soil samples and in the air show that none of the studied samples is considered a radiological hazard. In addition to that, estimations of the radium equivalent activity, external hazard index, internal hazard index and annual effective dose equivalent associated with the investigated soils have been made. In regard to the results of the above radiation hazard parameters, except for soils from regions where phosphate rocks exist (Jebel Katherina, Jebel Serbal and Jebel el Bab), do not exceed internationally recommended values, hence soils can be safely used in construction of buildings and exploits for the agriculture without posing any significant radiological threat to population. It is important to point out that these values were not the representative values for the Sinai area mentioned, but for the regions from where the samples were collected. Further investigation is still needed both to measure soils from deep layers and vegetations grown by these soils.

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TABLE 6 : The activity concentrations of ^{226}Ra in soil samples (Bq/Kg), evaluation of the activity concentrations of ^{222}Rn in soil and in air (Bq/m^3), the annual effective dose resulting from consumption of vegetables(^{226}Ra)and inhalation of ^{222}Rn gas (µsv/y), and the activity concentrations of ^{226}Ra in vegetables (Bq/Kg).

Location	A_{Ra} (Bq/Kg)	concentration of ^{222}Rn inside the soil Gs(n) (Bq/m3)	Activity ^{222}Rn gas in Air Ca(n) (Bq/m3)	Concentration of ^{226}Ra in vegetables Cp (Bq/Kg)	Annual effective dose from ^{222}Rn inhalation Hp (µsv/y)	Annual effective dose from ^{226}Ra consumption in vegetables Hp (µsv/y)
Jebel Katherina	118.9 ± 10.17	21276.00	67.28	3.93	139.94	99.09
Jebel Umm Shaumar	62.25 ± 5.35	11205.00	35.43	3.09	73.70	77.92
Jebel Serbal	85.25 ± 7.33	15345.00	48.53	3.24	100.93	81.65
Jebel Abbas Basha	77.33 ± 6.65	13919.40	44.02	2.66	91.56	66.93
Jebel el Bab	135.85 ± 11.68	24453.00	77.33	3.28	160.84	82.56
Jebel Bab el Donya	55.75 ± 4.79	10035.00	31.73	1.31	66.01	33.06
Jebel Naja	45.80 ± 3.94	8244.00	26.07	3.10	54.23	78.12
Jebel el Banat	32.25 ± 2.77	5805.00	18.36	2.65	38.18	66.83
Jabal ?ur Sina	34.65 ± 2.98	6237.00	19.72	2.35	41.02	59.27
Jabal Musa	49.4 ± 4.25	8892.00	28.12	2.22	58.49	55.94
Jabal Thabt	11.15 ± 0.96	2007.00	6.35	0.53	13.20	13.31
Jabal Sabbah	48.2 ± 4.15	8676.00	27.44	2.22	57.07	55.84
Jabal Mileihis	23.5 ± 2.02	4230.00	13.38	0.88	27.82	22.28
Jabal Matamir	29.95 ± 2.58	5391.00	17.05	1.76	35.46	44.25
Jabal Guna	90.4 ± 7.77	16272.00	51.46	1.76	107.03	44.35
Jabal Barqa	48.2 ± 4.15	8676.00	27.44	1.77	57.07	44.65
Jabal Maharum	33.45 ± 2.88	6021.00	19.04	2.21	39.60	55.64
Jabal Fuga	21.6 ± 1.86	3888.00	12.29	0.54	25.57	13.65
Wadi El Shallal	16.55 ± 1.42	2979.00	9.42	0.57	19.59	14.41
Wadi Sahu	24.9 ± 2.14	4482.00	14.17	0.88	29.48	22.30
Wadi Firan	34.1 ± 2.93	6138.00	19.41	1.06	40.37	26.81
Mean	51.37	8953.97	28.31	1.95	58.90	49.15

It is worth mentioning, due to the high concentration of radium higher than the world average in most of the samples under study, particularly in Jebel Katherina, Jebel Serbal, Jebel el Bab and Jabal Guna samples, the

radon concentration in the soil and in the air were also high, this led to higher effective dose rate in these samples, this is due to the natural state and creation of soil.

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