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Gamma ray measurements of naturally occurring radioactive materials in various types of Egyptian rocks in Al-Qusair area

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Abstract: *Thirty rock samples from Al-Qusair area in Egypt have been investigated. The naturally occurring radionuclide contents have been measured by gamma-ray spectrometry employing a shielded HpGe detector. The activities of ^{238}U, ^{232}Th and ^{40}K have been determined in Bq Kg^{-1}. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K were ranged from (15.41±1.67) to (326.94±4.40), (2.05±0.98) to (25.58±3.40) and (39.02±3.98) to (654.94±14.42) Bq Kg^{-1} respectively. Radium equivalent activities, absorbed dose rate and the values of hazard indexes were calculated for the studied rock samples to determine the radiation hazards arising from those samples. The absorbed dose rate values were found to vary from 31.26 to 149.72 nGy h^{-1}. The radium equivalent values for the investigated samples are less than the international average value 370 Bq Kg^{-1}. The present results have revealed that the radiological parameters are normal and within the international levels. This study provides a base line map of radioactivity background levels in the Egyptian environment and could be used as reference information to assess any changes in the radioactive background level due to the geological process.*

Keyword: *NORM; Absorbed dose; Hazard index; Radium equivalent*

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I. Introduction

Radiation is present in all the Earth's surface environment, beneath the Earth and in the atmosphere. Human beings are exposed to external radiation and radiation from the naturally occurring radionuclides in their immediate surroundings from one side, and to internal radiation from the food, water and air they consume from the other side. Around the world, there are some areas with high background radiation levels. Natural radioactivity is composed of the Cosmogenic and radionuclides. Cosmogenic and radionuclides, such as ^{3}H, ^{7}Be, ^{14}C and ^{22}Na, are produced by the interaction of cosmic-ray particles (mainly high-energetic protons) in the Earth's atmosphere. Primordial radionuclides, also called terrestrial background radiations, are formed by the process of nucleosynthesis in stars. Only those radionuclides with half-lives comparable to the age of the Earth and their decay products can still be found today on Earth, e.g. ^{40}K and the radionuclides from ^{238}U and ^{232}Th series. Gamma radiation from these radionuclides represents the main external irradiation source of the human body. In the environment, the natural radionuclides, such as ^{40}K, ^{232}Th and ^{238}U are present in the Earth's crust [1]. Therefore,materials from the Earth's crust such as soil, building materials, and so on become a major source of external radiation exposure to humans in the environment. Everyone on the planet is exposed to some background level of ionizing radiation. External exposures will occur as a result of irradiation, and internal exposures will occur as a result of inhalation.The external radiation exposure arises mainly from cosmic rays and from terrestrial radionuclides occurring at trace levels in all soils. The absorbed dose rate in air resulting from cosmic radiation outdoors at sea level is about 30 nGy h^{-1} [2], while the specific levels due to terrestrial background radiation are related to the types of rock from which the soil originates. Therefore, the natural environmental radiation mainly depends on geological and geographical conditions [3]. Higher radiation levels are associated with igneous rocks, such as graniteand lower levels with sedimentary rocks.Shale and phosphate rocks can be considered an exception where shale and phosphate rocks have a relatively high content of radionuclides [1].

The present study aims to measure the naturally occurring radionuclide content in various types of Egyptian rocks in Al-Qusair area, and to initiate a map of radioactivity background levels in the surrounding environment around Al-Qusair area as well. This map could be used as reference information to detect any change in radioactivity background levels due to different geological processes or any effects on the radiation environment.

II. Material And Methods

Study area

Both of WadiQwieh and Wadi Abu Hamra are located in the central part of the Egyptian eastern desert. The studied area lies in the western side of the red sea, between Safaga and Qusair, between latitudes 26° 16′N and 26° 19′N and longitudes 34° 5′E and 34° 7′E respectively. Figure (1) shows the location map of studied area that is usually enriched with the Phosphate mining workers. Table (1), on the other hand, illustrates the description, location of different studied samples and their coordinates.

The area of study is bounded by the basement rocks in both the northern and western sides. It is also bounded by the quaternary deposits in the eastern side up to the coastal boundary. The area is fairly accessible; and the main paved road of Qusair – Safaga highway that extends from Safaga to Qusair is roughly parallel to the western boundary of the Red Sea, then to the entry of WadiQwieh which is located 55.5 kilometers far from Safaga and the entrance of Wadi Abu Hamra that is located 62.5 kilometers from Safaga. Safaga town lies in the northern part of the study area, but Qusair town is located in the southern part. The study area lies between the narrow coastal plain of the Red Sea and the topographically extended high area of WadiQwieh basement, which is about 360 meters high above sea level.

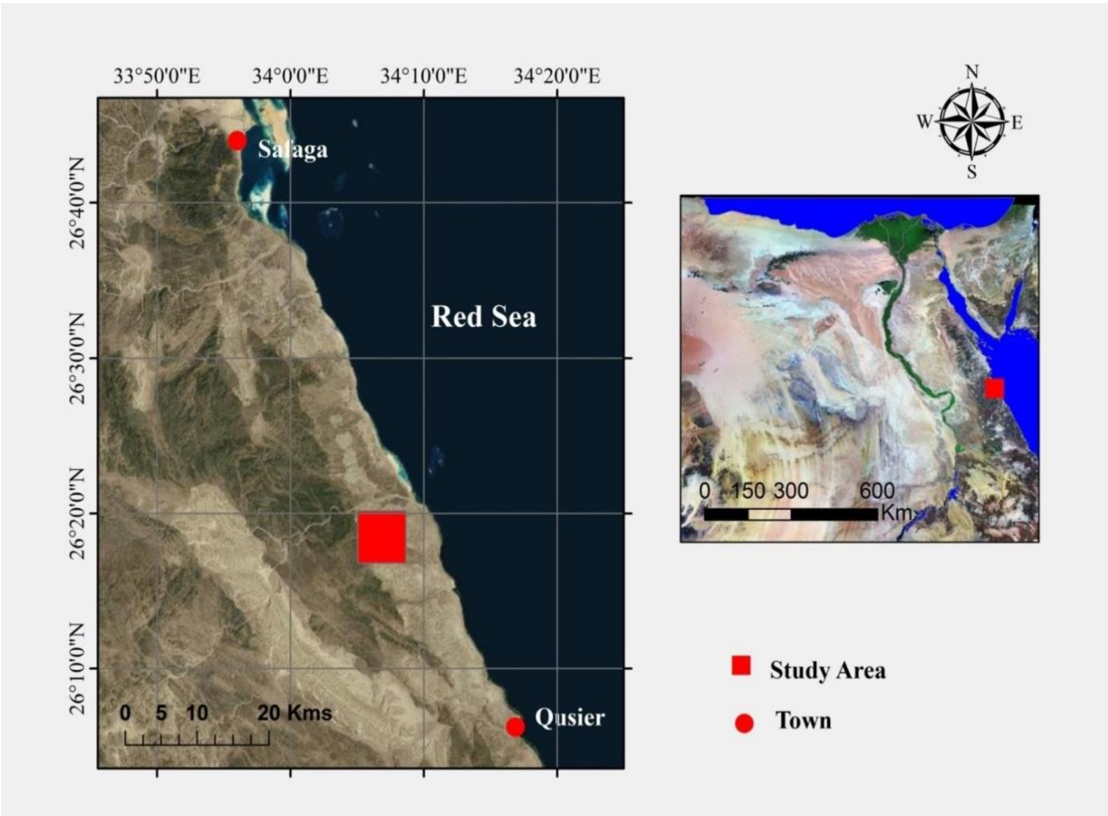

Figure no 1: location map and the distribution of Studied Sections in the study area

Table no 1: Sample description and location

Sample type	No. of samples	Thickness	Location (from west of the Qusair-Safaga road)	General contents	Coordinates
Qusair variegated shale	6	25.5 m	13 km	Sandstone, shale, gypsum, iron oxide, marl, siliceous limestone and manganese.	Latitudes 26° 17′ 53″N Longitudes 34° 5′35″E
Duwi Formation	6	55.9 m	9 km	Phosphate, phosphatic limestone, dolomatic limestone, shale, gypsum, iron oxide, marl, siliceous limestone and manganese veinlets.	Latitudes 26° 17′2″N Longitudes 34°6′ 23″E
Dakhla Shale	5	42.5 m	9 km	Phosphatic limestone, sandstone, shale, gypsum, iron oxide, marl, siliceous limestone and manganese	Latitudes 26°16′ 59″N Longitudes 34° 6′ 8″E
Tarawan Chalk	2	9 m	8 km	Chalky limestone, siliceous limestone, manganese staining and chert lenses.	Latitudes 26° 18′ 52″N Longitudes 34° 5′35″E
Esna Shale	6	30 m	10 km	Shale, gypsiferous, iron oxides, crystalline gypsum veinlets and manganese patches.	Latitudes 26° 18′ 39″N Longitudes 34°5′ 32″E
Thebes Formation	5	17.5 m	10 km	Limestone, chalky limestone, dolomitic limestone, marly limestone, iron oxides patches, dendritic manganese and chert nodules.	Latitudes 26° 18′ 39″N Longitudes 34° 5′32″E

Sample preparation

Thirty samples have been collected from Al-Qusair area. Table no1 represents the sedimentary rock formations of the collected samples that were prepared in Central Laboratory for Environmental Radioactivity Measurements, Inter-comparison and Training (CLERMIT). Measured samples of 100 cm^3 in volume were homogenized and sieved through 0.15 mm mesh size by a crushing machine and then dried in oven at 50°C for twenty four hours so that moisture can be removed from samples. Then packed and sealed in standard 1000 ml Marinelli beakers, and stored for four weeks to reach secular equilibrium between ^{232}Th and ^{238}U and there short lived daughter products [4, 5].

Gamma spectrometry

The radionuclide activity concentrations in the studied samples were measured using an n-type coaxial high-purity germanium (HPGe) detector, Canberra model in the Central Laboratory for Environmental Radioactivity Measurements, Inter-comparison and Training (CLERMIT) - Egypt. The detector has an efficiency of about 40%, energy resolution of 1.9 keV, full width at the half maximum (FWHM) for the 1332.3 keV gamma line of ^{60}Co and MCA with 8000 channels. An empty Marenilli beaker was used to determine the background concentrations of the γ- rays. The background spectra were used to correct the net γ- ray peak areas for the studied isotopes. To constrict the γ- rays background from building and cosmic rays, a cylindrical lead shield of 100mm thickness is used to protect the detector from the surrounding environment. This shield is composed of three inner concentric shells of lead, cadmium and copper [6]. The γ- ray transitions used to measure the concentration of the assigned nuclides in the series were as follows: ^{226}Ra (186.1 KeV), ^{214}Pb (295.1and 352.0 KeV), ^{214}Bi (609.3, 1120.3 and 1765 KeV) for uranium series; ^{208}Tl (583.0 KeV) ^{212}Bi (39.86, 288.07, 727.33 and 1620.50KeV) and ^{228}Ac (338.5 and 911.2 KeV) for thorium series; ^{40}K (1460 KeV) for potassium.

III. Result and Discussion

Table no 2illustrates the average concentration of radionuclides, ^{238}U, ^{232}Th and ^{40}K which have been calculated for thirty rock samples from six different rock types in Al-Qusair area.

The concentration for ^{238}U, ^{232}Th and ^{40}K varies from (15.41±1.67) to (326±4.40), (2.05±0.98) to (25.58±3.40) and (39.02±3.98) to (654.94±14.42) Bq kg^{-1} respectively in rock samples used in this study. The activity found for ^{238}U is the lowest for Qusair Variegated Shale and the highest for Duwi Formation. The results showed that the specific activity values of ^{238}U were higher than the permissible limit except Qusair Variegated Shale type as the specific activity value of ^{238}U was in the permissible level 35 Bq Kg^{-1} [2]. The high concentration values of ^{238}U in the studied samples may be indicating that the samples are rich by phosphate ore.

^{232}Th concentration for Tarawan Chalk has been found to be the lowest while Qusair Variegated Shale has marked the highest. Also, ^{40}K concentration in Qusair Variegated Shale has been found to record the maximum while in Thebes Formation ranked the minimum. The results showed that the specific activity values of ^{232}Th were in permissible limit 30 Bq Kg^{-1} [2]. Also, the specific activity values of ^{40}K were in the permissible limits 370 Bq Kg^{-1} except for the samples of Qusair Variegated Shale where the specific activity value of ^{40}K was (654.94±14.42) Bq Kg^{-1}.

Table no 3 shows the comparison between the results obtained in the present study and the levels of the natural radionuclides in rock samples of various other countries.

Table no 2: The average activity concentrations of ^{238}U, ^{232}Th and ^{40}K radionuclides for different rocks samples of Al-Qusair area in Egypt

Sample Type	**No. of samples**	**^{238}U (Bq Kg^{-1})**	**^{232}Th (BqKg^{-1})**	**^{40}K (Bq Kg^{-1})**
Qusair Variegated Shale	6	15.41±1.67	25.60±3.40	654.94±14.42
Duwi Formation	6	326.94±4.4	10.55±2.48	72.93±7.02
Dakhla Shale	5	80.84±2.88	22.38±2.48	251.45±10.04
Tarawan Chalk	2	69.77±2.95	2.21±1.35	69.41±6.75
Esna Shale	6	50.09±2.42	13.41±2.1	169.29±8.10
Thebes Formation	5	66.09±2.8	2.05±0.98	39.20±3.98

Table no 3: Comparison between the activity concentrations of our rock samples with that of other countries of the world

Country	^{238}U	^{232}Th	^{40}K	Reference
Egypt (Abu-Zaabal)	514	37	19	[14]
Egypt (El-Quseir)	358	38	N.F	[15]
Egypt (El-Mahamid)	567	217.3	217.3	[16]
Egypt (W. El-Mashash)	666	329.4	329.4	[16]
Egypt (Bir El-Sid)	57.4 ±4.5	53.4 ±5.4	1041.4 ±76.7	[17]
Egypt (Wadi El-Gemal)	39 ±3.2	47.9±5.1	1031.0±75.6	[17]
Germany	76.1 ±5.1	70 ±6.5	1465.4±106.8	[17]
Egypt (Eastern desert)	23 ± 5	30 ± 6	563± 24	[18]
Egypt (Sinia)	17.2-85.9	26.1-121.1	940.6-1322.9	[19]
Egypt (Minia)	42	2.8	97	[20]
Egypt (Aswan)	163	40	49	[20]
Pakistan (Hazara)	440	50	207	[21]
Finland	10	10	110	[21]
Sudan (Uro)	4131	7.5	62.3	[22]
Sudan (Kurun)	393	6.9	141.3	[22]
Pakistan	9-40	4-63	7-105	[23]
Brazil	257	284	45	[24]
Turkey	12.0-48.9	8.2-53.3	144.0-452.3	[25]
Egypt (Al-Qusair)	15.41-326.94	2.05-25.60	39.02-654.94	The present work

Estimation of γ- radiation dose

The γ radiation doses of the sample content of radionuclides can be estimated by employing the convenient formula [7, 8]. The conversion factors used for (D) calculations were 0.427 nGy h^{-1} for ^{238}U,0.662 nGy h^{-1} for ^{232}Th and 0.043 nGy h^{-1} for ^{40}K respectively.

$$D\ (\text{nGy h}^{-1}) = 0.427\ C_U + 0.662\ C_{Th} + 0.043\ C_K \qquad (1)$$

Where, C_U, C_{Th} and C_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively.

As shown in Table no 4 the absorbed dose rate values were found to vary from 31.26 to 149.72 nGy h^{-1}. The lowest value was found in Thebes Formation type and the highest value in Duwi Formation type. The absorbed dose rate values are below the permissible level 59 nGy h^{-1} [2] except for the samples of Dakhala Shale type and Duwi Formation type where the values are 60.14 and 149.72 nGy h^{-1} respectively.

Calculation of radium equivalent

The radium equivalent index is generally introduced as the weighed sum of ^{226}Ra, ^{232}Th and ^{40}K based on the assumption that 10 Bq $Kg^{-1}$of ^{226}Ra, 7 Bq $Kg^{-1}$of ^{232}Th and 130 Bq $Kg^{-1}$of ^{40}K will produce the same dose rates of gamma rays. Values of Ra_{eq} were calculated using the following equation [6, 9]:

$$Ra_{eq}\ (Bq/Kg) = C_{Ra}+1.43C_{Th}+0.077C_{K} \qquad (2)$$

where, C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively.

Table no 4 reveals the radium equivalent values which were found to vary from 72.04 to 347.64 Bq Kg^{-1}. The lowest value was found in Thebes Formation type and the maximum value was found in Duwi Formation type. The radium equivalent values for the investigated samples are less than the international average value 370 Bq Kg^{-1} [2].

External hazard index

The external hazard index due to the emitted γ- rays of samples was calculated by using the following equation:

$$H_{ex} = C_{Ra} / 370 + C_{Th} / 259 + C_K / 4810 \ \leq 1 \qquad (3)$$

where, C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively.

The value of the external hazard should be below one which corresponds to the upper limit of Ra_{eq} (370 Bq Kg^{-1}) to keep the radiation hazard insignificant [10].Table no 4 illustrates the values of external hazard index that have varied between 0.20 in Thebes Formation type to 0.94 in Duwi Formation type. The values of external hazard index have been found to be under the permissible levels [2].

Internal hazard index

The internal exposure to ^{222}Rn and its daughter products are controlled by an internal hazard index H_{in} then calculated and exanimated according to the following criterion [11, 12]:

$$H_{in} = C_{Ra} / 185 + C_{Th} / 259 + C_K / 4810 \ \leq 1 \qquad (4)$$

Table no 4presents the values of internal hazard index, where the lowest value 0.34 was found in QusairVariegated Shale type while the maximum value 1.83 was found in Duwi Formation type. The internal hazard index for all studied samples is below the permissible levels except that of Duwi Formation typeexceeds the permissible levels [1].

Radioactivity level index

The radioactivity Level index has been used to estimate the level of gamma radiation hazard associated with different concentrations of some specified radionuclides [6, 9, 13]. The radioactivity Level index was calculated by using the following equation:

$$\mathbf{I\gamma} = \frac{\mathbf{C_{Ra}}}{\mathbf{150}} + \frac{\mathbf{C_{Th}}}{\mathbf{100}} + \frac{\mathbf{C_k}}{\mathbf{1500}} \quad (5)$$

Table no 4 represents the radioactivity level index, where the lowest value 0.49 was found in Thebes Formation type while the maximum value 2.33 was found in Duwi Formation type. The radioactivity level index for all studied samples was under the permissible levels except samples of Duwi Formation type were higher than the permissible levels [1]. If the external hazard exceeds unity, it could be concluded that the external doses that individuals are exposed to will exceed the acceptable levels.

Table no 4: Radium equivalent (Ra_{eq}), absorbed dose rate (D), external hazard index (H_{ex}), internal hazard index (H_{in}) and radioactivity level index (Iγ)for different rocks samples

Sample type	No. of samples	Ra_{eq} (Bq Kg^{-1})	D (nGy h^{-1})	H_{in}	H_{ex}	I_γ
Qusair Variegate Shale	6	102.42	51.68	0.34	0.30	0.80
Duwi Formation	6	347.64	149.72	1.83	0.94	2.33
Dakhla Shale	5	132.20	60.14	0.58	0.37	0.93
Tarawan Chalk	2	78.28	34.24	0.40	0.21	0.53
Esna Shale	6	82.31	37.55	0.36	0.23	0.58
Thebes Formation	5	72.04	31.26	0.37	0.20	0.49

Annual effective dose rates

To estimate the annual effective doses (AED), one has to take into account the conversion coefficient from absorbed dose in air to effective dose 0.7 Sv Gy^{-1}, the outdoor occupancy factor 0.2 and the indoor is 0.8 [2]. The annual effective dose rates are determined as follows [10]:

$AED_{outdoor}$ (mSv y^{-1}) = D (mGy h^{-1}) × 24 h × 365.25 d× 0.2 × 0.7 ×10^{-6} (6)

and

AED_{indoor} (mSv y^{-1}) = D (mGy h^{-1}) × 24 h × 365.25 d× 0.8 × 0.7 ×10^{-6} (7)

where, D is the absorbed dose rate

Table no 5 presents the values of indoor, outdoor and the total annual effective dose rate for different collected samples. It can be seen that the total values for each formation type are less than the corresponding worldwide value of 1 mSv y^{-1}.

Table no 5: The outdoor ($AED_{outdoor}$), indoor (AED_{indoor}) annual effective doses and total annual effective doses (AED_{tot}) for different rocks samples

Sample type	No. of samples	$AED_{outdoor}$ (mSv y^{-1})	AED_{indoor} (mSv y^{-1})	AED_{total} (mSv y^{-1})
QusairVarigated Shale	6	0.06	0.25	0.32
Duwi Formation	6	0.18	0.74	0.92
Dakhala Shale	5	0.07	0.30	0.37
Tarawan Chalk	2	0.04	0.17	0.21
Esna Shale	6	0.05	0.18	0.23
Thebes Formation	5	0.04	0.15	0.19

IV. Conclusion

High- resolution gamma ray spectroscopy is a good experimental tool to study natural radioactivity and determine activity concentrations as well as dose rates in different rock types. The specific activity values of ^{238}U are found to exceed the permissible levels for all types of formation as this formation is rich by phosphate ore except for Qusair Variegated Shale. The specific activity values of ^{232}Th and ^{40}K are normal and within the international levels except for the sample of Al-Qusair formation type which is rich by shale and known by Qusair Variegated Shale where the specific activity of ^{40}K was higher than the permissible level. The activity concentrations were obtained to calculate several radiological parameters that help to conduct quantitative and qualitative designation of the radiological hazard associated with the studied rock samples. The radiological parameters obtained are normal and within the international levels. The obtained results are considered modernized base line data and a radiological map for the studied area as well.

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