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^{238}URadionuclides Series Ratios As A Guide For Alteration Processes Grades And Times of Different Rock Samples, Southwestern Sinai, Egypt

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Abstract: *The activity concentrations of the radionuclides ^{238}U, ^{232}Th and ^{40}K in twenty six sedimentary rock samples collected from four localities at southwestern Sinai, Egypt were measured using gamma-ray spectrometry with a high-purity germanium (HPGe) detector.The average activity concentration values of ^{238}U are approximately 42, 34, and 50 times the world average value at EL-Qur, GabalHomeirat, andGabalHomyerlocalities, respectively. As for the fourth locality;GabalAllouga, the average activity concentration value of ^{238}U is nearly two hundred and twenty times the world average value. ^{232}Th average activity concentrationvalues for GabalHomeiratand GabalHomyer localities are less than the world average activityconcentration value while it is nearly twice the world average value for El-Qur and GabalAllougalocalities.The ratio $^{226}Ra/^{238}U$is classified into four grades indicating of alteration processes for the examined samples.The uranium migration was recorded for 12 samples.Eight samples from GabalAllougawere chosen to calculate their alteration processgrades and duration history which ranged between106ka and 246ka.*

Keywords*:GabalAllouga, Gamma Spectroscopy,History of Alteration, Sedimentary Rocks.*

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I. Introduction

The earth's surface processes as weathering and alteration can be studied and interpreted by uranium series disequilibrium [1]. The differences in radionuclides mobility during the weathering processes can lead to the deviation from secular equilibrium. The relative mobility is believed to be $^{234}U>^{238}U>^{230}Th$, consequently the weathered rocks showed $^{234}U/^{238}U < 1$and $^{230}Th/^{238}U > 1$[2]. The extent of the disequilibrium depends mainly on the intensity and age of weathering processes.The sedimentary rocks either marine or continental usually contain radionuclides with different concentrations. Most of these radionuclides show disequilibrium between parents and their daughters due to the weathering and/or alterations. The ratios $^{234}U/^{238}U$and $^{230}Th/^{234}U$can be used to determine the weathering events up to 1 Ma and 300 Ka, respectively [1].The$^{226}Ra/^{238}U$ratios were studied in different rock types[3]and they clarified three categories: higher, around and lower than unity. These different categories are interpreted due to the alteration processes that affected these rock types. The alteration processes may lead to the migration of uranium and/or radium in or out of the rock, which may cause the differences of the ratios between uranium and its daughters.

II. Geologic Setting

The Sinai Peninsula covers an area of about 6.1% from the total surface of Egypt (1.01millionkm^2). This Peninsula is located between the Mediterranean Sea from the north, Gulf of Aqaba from the east and Gulf of Suez from the west. This study is concerned with the southwestern part of the Peninsula which is covered by different types of rocks which varied between granites and granodiorites (500-600 m.y) to sedimentary rocks mainly of lower Carboniferous (325 m.y)[4].The main types of the sedimentary rocks are dolomites, siltstone, shale and claystone which represent radioactive anomalies with uranium and low thorium. Four localities lies between longitude$33^0$15'to $33^0$35'E and latitude $29^0$00'to$29^0$05'N (Fig. 1)were chosen in this study to apply the fractionation of the uranium isotopes and their ratios with daughters during the alteration processes of different rock types.

III. Types Of Rocks

3.1Shalesand Siltstones

Blatt [5] estimates 69% of the continental sediments of the world to be shale. On the basis of certain geochemical considerations, shales should form 80% of all the sedimentsproduced through geologic time [6].Clark [7] defined shale as a rock whose particles have a diameter less than 1/16 mm. This definition embracingare both siltstone as well as shale. Shale is a fissile claystone.

3.2 Sandstones

Sandstones are produced by weathering and break down of preexisting rocks. These deposits reflect well the conditions prevailed during their deposition. Sandstones usually contain voids between their grains which can play as traps for water, oil and gas and also metals.

3.3 Limestones and Dolomites

The term limestone is applied to those rocks in which the carbonate fraction exceeds the non carbonate constituents [8]. If sand sized detrital quartz is present excess of 50 percent, the term calcareous sandstone would be more used. Also, rocks in which shaly matter exceeds the carbonate fraction are calcareous shales rather than limestones. The term dolomite is used for those rocks which are consisted mainly of the mineral dolomite even though dolomite is, of course a lime-bearing rock. The dolostone is the rock which contains less than 50% dolomite and the rest percent of other rocks.

IV. Materials And Methodology

4.1 SamplesCollectionand Preparation.

Twenty six rock samples were collected fromdifferent lithologies, i.e. sandstone,claystone, shales and siltstone sedimentary rocks, fromfour different localities named:El-Qur (8 samples),Gabal(mountain)Homeirat(4 samples), Gabal(mountain)Homyer (4 samples), and GabalAllouga(south site) (10 samples), southwestern, Sinai, Egypt (Fig.1 and Table1(a) to 1(c)**).**The samples should give labeling code that be used at all stages of storage, preparation and analysis. Each sample represents itself and collected by a method known as a grab sampling.

Samples were then subjected to air dried at room temperature in open air in order to remove excess moisture. These samples were packed in a polyethylene plastic Marinelli beakers of constant volume, so that there is geometric homogeneity around the detector and then the respective net weights were measured and recorded with a high sensitive digital weighing balance with a percent of ±0.01%. After that, the plastic Marinelli beakers were sealed with a PVC tape, and stored for about one month before counting, to allow secular equilibrium to be attained between ^{222}Rn and its parent ^{226}Ra in uranium chain.

4.2 Experimental Procedures

The samples were analyzed nondestructively, using gamma-ray spectrometry with a high-purity germanium (HPGe) detector. This detector has a relative efficiency of approximately 50% of the 3″x3″ NaI(Tl) crystal efficiency, with energy resolution (FWHM) 1.90 keVand a peak/Compton ratio of 69.91 at the 1.332 MeV gamma-ray line of ^{60}Co source. The detector is coupled to conventional electronics and connected to a multi-channel analyzer (MCA) card installed in a PC. The detector is shielded from the background radiation, using a 10 cm thickness lead cylinder shield, internally lined with a 2 mm thick copper foil[9].The energy and the efficiency calibrations are two steps to calibrate the gamma ray detection system. The energy calibration translates channel numbers to gamma-ray energy in MeV and the efficiency calibration aimed to determine the gamma ray counting efficiencies over the full energy range of measurement.

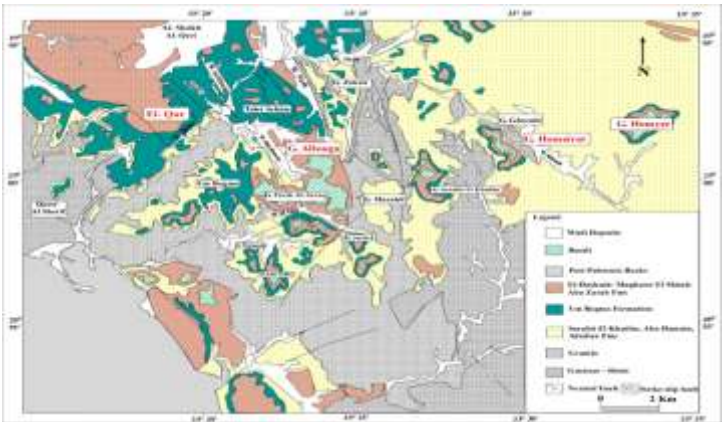

Figure1: Geologic map of the study area with the location of the collected samples (After[10]).

A set (RG-set) of high quality certified reference sources [9] used to achieve the energy and efficiency calibrations of the detection system. For calibrations, the reference source was placedin the same place as the samples when measuring their gamma-ray spectra. The system is calibrated for energy to display gamma-ray photo-peaks between 63 and 3000 keV. The software program MAESTRO-32 was used to accumulate and analyze the data.Activity of several nuclides for uranium-238 (^{234}Th, ^{234m}Pa, ^{234}U, ^{230}Th, ^{226}Ra, ^{214}Pb, ^{214}Bi and ^{210}Pb) decay series have been measured through gamma spectrometry.Uranium-238 activity was determined indirectly from the gamma rays emitted by its daughter products (^{234}Th and ^{234m}Pa) whose activities are determined from the 63.3 and 1001 *keV*photopeaks respectively [11].

The uranium-234 activity was determined directly from the gamma rays emitted from this nuclide at energy of 53.2 *keV*[12]. For the measurement of the thorium-230 activity, the γ-ray emission at 67.7 keVis used[13] and Pb-210 was determined from its own gamma 46.5 keV. The specific activity of radium-226 was measured using the 186.1 keVfrom its own gamma-ray after the subtraction of the 185.7 keVof uranium-235 [3]. The specific activity of lead-214 was measured using the 295.1 and 352 keVphotopeaks, whereas the specific activity of bismuth-214 was measured using the 609.3, 1120.3 and 1764.5 keVphotopeaks.

Table 1(a): Samples Description of El-Qur locality.

locality	Sample ID	Description
El-Qur	**Qur-1** **Qur-1a** **Qur-3** **Qur-5** **Qur-8** **Qur-8a** **Qur-9** **Qur-11**	• Siltstone to very fine grained sandstone, light brown, soft to medium hard. • Siltstone to very fine grained sandstone, light brown, soft to medium hard, more compact. • Variegated shale, yellow, brown and reddish, highly ferruginous, soft and fissile. • Sandy siltstone, medium hard, brownish. • Variegated silty shale, lateral extension of Qur-3. • Sandy claystone, grey to brown, medium hard. • Siltstone, grey with brown patches, soft. • Sandstone, fine to medium grained, medium hard, light brown.

Table 1(b): Samples Description of GabalHomeirat and GabalHomyer localities.

Locality	Sample ID	Description
GabalHomeirat	**M-11** **M-12** **M-13** **M-14**	• Sandstone, ferruginous, red, fine to medium grained, medium hard to semi friable. • Sandstone, ferruginous, red, fine-grained. • Siltstone, ferruginous, yellowish red, laminated, at the distance between GabalGhorabi and GabalHomeirat. • Siltstone, ferruginous, yellow, laminated, 10m to the west from the previous sample.
GabalHomyer	**M-21** **M-22** **M-23** **M-24**	• Ferruginous sandstone, red, coarse grained, massive, from the base of Adediya Formation in the NE-SW trend. • Sandstone, ferruginous, reddish brown, coarse to medium grained from the SW direction. • Ferruginous sandstone, reddish brown, coarse grained, massive. • Ferruginous sandstone, brown, coarse grained, medium hard.

Table 1(c):Samples Description of GabalAllouga locality.

Locality	Sample ID	Description
GabalAllouga	**A-1** **A-2** **B-1** **B-2**	• Siltstone,pale yellow to brown, ferruginous, soft to medium hard. • Siltstone, pale yellow to brown, ferruginous, soft to medium hard. • Variegated shale, brown, yellow and reddish, fissile, soft to medium hard. • Variegated shale, 2m north of the previous sample, highly ferruginous.

	B-3 **B-4** **C-1** **C-2** **D-1** **D-2**	• Variegated shale, 3m to the north of sample B-2. • Variegated shale, yellowish brown with yellow secondary uranium mineral as encrustation, 2m to the north of sample B-3. • Shale, brownish, fissile, ferruginous, soft. • Shale, yellowish brown, 4m to the north from sample C-1. • Marly shale, yellow to brown, fissile, ferruginous. • Marly shale as the previous, 3m to the north.

The uranium-235 activity was determined by its gamma ray photopeaks: 143.8, 163.4, 185.7 and 205.3keV. The specific activity ofthorium-232 was measuredfrom the gamma rays emitted by its daughter products (Ac-228 and Tl-208) whose activities are determined from the 338.4 and 911.2keVfor Ac-228, 583 and 860.3keVfor Tl-208. K-40was measured from its own gamma1460.8keV.

V. Results And Discussion

The measured activitiesand concentrations (BqKg^{-1}, ppm andppt) of the radionuclides ^{238}Uand its daughters, ^{232}Thseries and ^{40}K of twenty sixsamples collected from four various localities at southwestern Sinai, Egypt presented in Table 2(a) to 2(d).Activity concentrations, (Bqkg^{-1}) were converted to the values of AU and AThinppm as well as AK in %, using the conversion factors given by the International Atomic Energy Agency [14]. The activity concentration of a sample containing 1 ppm by weight of ^{238}U is 12.4 Bqkg^{-1}, 1 ppm of ^{232}Th is 4.1 Bqkg^{-1} and 1 % ^{40}K is 313 Bqkg^{-1}. Essentially all naturally occurring radium is present as radium-226. Radium exists naturally in soil, rocks, surface water, groundwater, plants, and animals in generally low concentrations on the order of one or more parts per trillion, or 1 picocurie (pCi)/g. The average activity concentrations of uranium (Bqkg^{-1} andppm), thorium (Bqkg^{-1} and ppm) and Potassium % are represented in Table 2(a) to 2(d).

The averageactivity concentration values are comparable to the world average activity concentrations [15]which are 412, 32, and 45 Bqkg^{-1}for ^{40}K, ^{238}Uand^{232}Threspectively. The average activity concentration values of^{238}Uare approximately 42, 34, and 50 times the world average value for EL-Qur, GabalHomeiratandGabalHomyerlocalities respectively.For GabalAllouga, the average activity concentration value of^{238}Uis nearly two hundred and twenty times the world average value.^{232}Th average activity concentrations for GabalHomeirat(30.12Bqkg^{-1})andGabalHomyer (22.43Bqkg^{-1}) localities are less than the world average activity concentration value (45Bqkg^{-1}), while it is nearly twice the world average value for El-Qur (93.02Bqkg^{-1}) and GabalAllouga (100.84Bqkg^{-1}) localities.^{40}Kaverage activity concentrations for four localities are varied from the less or more than one and half the world average value. Average activity concentrations of the radionuclides at EL-Qur, GabalHomeirat and GabalAllouga localities increase according to the order ^{238}U > ^{40}K > ^{232}Th, while at GabalHomyer increases according to the order ^{238}U > ^{232}Th > ^{40}K.

^{238}U/^{232}Thratios ranged between 19.2:21.5, 19.2:21.7, 20.1:21.8 and 18.7:22.1 for El-Qur, G.Homeirat, G.Homyer and G.Allouga respectively, this show there is disequilibrium between U and Th for all samples.

^{238}Ucontribution in G.Homyer(Fig. 2) with percentage of 31%, followed by G.Homeriat and El-Qur (29%) and the least in G.Allouga (24%). ^{226}Racontribution with percentage of (37%) in both G.Homyer and G.Allouga. ^{234}U contributionwith percentage of (35%) in G.Allouga followed by G.Homyer (31%), G.Homeriat (30%) and the least in El-Qur (27%). From the previously mentioned data, it can be concluded that the most probable situation in the last alteration process led to migration out of uranium from G.Allouga to the east in both G.Homyer and G.Homeriat and to the west to El-Qur locality. This situation can be imagined if the path-ways were connected at that time.

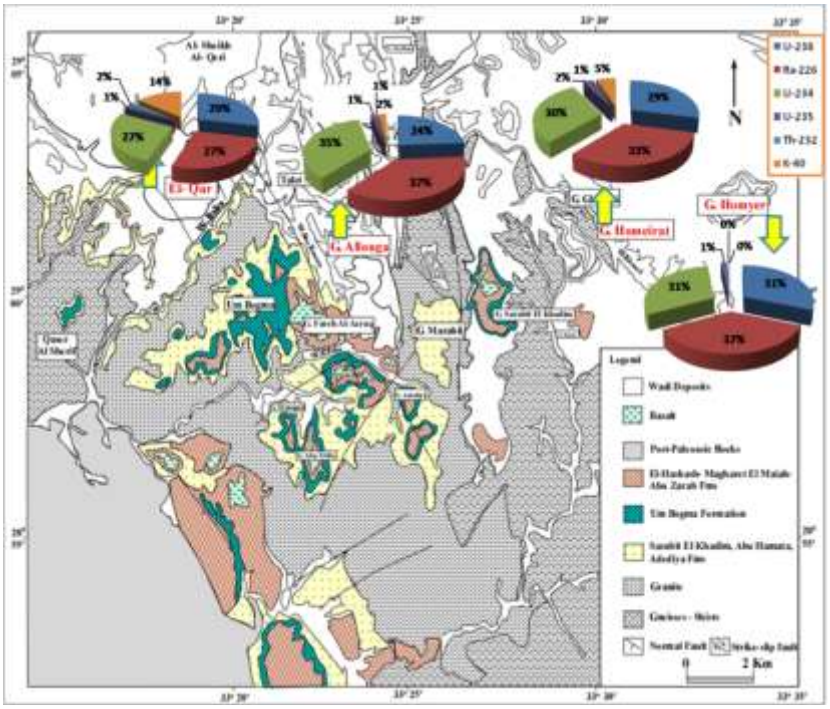

Figure 2: Relative contribution of average activity concentrations for U-238, Ra-226,U-234,U-235, Th-232 and K-40 for different localities at southwestern Sinai, Egypt.

Table 2(a): The activity concentrations (Bqkg^{-1}, ppm, ppt) of U-238 series, Th-232 series, U-235 and K-40 (**%)** radionuclides for samples collected from El-Qur locality.

	U-238 series						U-235		Th-232 series		K-40	
ID	U-238 (Bqkg^{-1})	U-238 (ppm)	U-234 (Bqkg^{-1})	Ra-226 (Bqkg^{-1})	Ra-226 (ppt)	Pb-210 (Bqkg^{-1})	U-235 (Bqkg^{-1})	U-235 (ppm)	Avg.(Bqkg^{-1}) [Ac-228, Tl-208]	Th-232 (ppm)	K-40 (Bqkg^{-1})	K (%)
Qur-1	1554.60±76.26	125.37	1357.81±77.61	1362.92±25.02	36.84	635.03±14.53	79.43±5.55	0.99	80.07±3.52	19.53	692.99±18.86	2.21
Qur-1a	1393.16±62.60	112.35	1399.77±72.18	1437.42±22.81	38.85	744.64±16.80	71.28±3.11	0.89	85.68±3.50	20.90	671.65±17.31	2.15
Qur-3	2268.02±75.80	182.90	2129.72±63.09	1624.18±22.31	43.90	1274.93±16.48	116.80±5.76	1.46	122.06±2.77	29.77	821.04±16.00	2.62
Qur-5	1586.83±88.80	56.97	526.85±35.87	537.64±22.83	14.53	333.20±14.44	32.83±3.13	0.41	125.20±5.73	21.67	789.98±22.80	2.52
Qur-8	1329.59±72.07	127.97	1480.13±98.40	1369.17±31.89	37.00	763.93±18.39	76.13±6.35	0.95	134.37±8.82	30.54	1016.01±29.98	3.25
Qur-8a	706.42±77.38	72.16	681.36±38.59	801.97±15.30	21.67	453.95±11.46	41.75±3.21	0.52	88.83±3.90	21.50	917.21±15.01	2.93
Qur-9	894.81±35.75	107.23	ULD	1920.87±27.65	51.92	965.74±22.77	66.02±5.71	0.83	88.14±2.88	32.77	174.52±25.47	0.56
Qur-11	927.61±48.87	74.81	ULD	1069.62±15.39	28.91	567.37±11.65	46.61±2.48	0.58	19.81±1.85	4.83	207.51±08.69	0.66
Minimum	706.42±77.38	72.16	ULD	537.64±22.83	14.53	333.20±14.44	32.83±3.13	0.41	19.81±1.85	4.83	174.52±25.47	0.56
Maximum	2268.02±75.80	182.90	2129.72±63.09	1920.87±27.65	51.92	1274.93±16.48	116.80±5.76	1.46	134.37±8.82	30.54	1016.01±29.98	3.25
Average	1332.63±67.19	107.47	1262.61±64.29	1265.48±22.90	34.20	717.35±15.81	66.36±4.41	0.83	93.02±4.12	22.69	661.36±19.26	2.11

Table 2(b): The activity concentrations (Bqkg^{-1}, ppm, ppt) of U-238series, Th-232 series,U-235 and K-40(**%)** radionuclides for different sedimentary samples collected from GabalHomeirat locality.

	U-238 series						U-235		Th-232 series		K-40	
ID	U-238 (Bqkg^{-1})	U-238 (ppm)	U-234 (Bqkg^{-1})	Ra-226 (Bqkg^{-1})	Ra-226 (ppt)	Pb-210 (Bqkg^{-1})	U-235 (Bqkg^{-1})	U-235 (ppm)	Avg. (Bqkg^{-1}) [Ac-228, Tl-208]	Th-232 (ppm)	K-40 (Bqkg^{-1})	K (%)
M-11	156.25±031.99	12.60	104.72±23.54	167.37±08.43	4.52	43.58±07.55	7.20±1.55	0.09	22.97±3.04	5.60	385.09±15.76	1.23
M-12	397.29±070.41	32.04	379.76±51.30	393.80±16.93	10.64	206.86±15.97	ULD	ULD	25.83±4.77	6.30	ULD	ULD
M-13	216.77±031.13	17.48	ULD	358.30±11.85	9.68	153.87±06.69	10.40±1.10	0.13	20.43±1.68	4.98	16.04±05.67	0.05
M-14	3536.74±112.26	285.22	2925.40±73.43	3985.63±40.66	107.72	2601.34±37.30	184.02±7.91	2.30	51.24±4.06	12.50	ULD	ULD
Minimum	156.25±031.99	12.60	ULD	167.37±08.43	4.52	43.58±07.55	ULD	ULD	20.43±1.68	4.98	ULD	ULD
Maximum	3536.74±112.26	285.22	2925.40±73.43	3985.63±40.66	107.72	2601.34±37.30	184.02±7.91	2.30	51.24±4.06	12.50	385.09±15.76	1.23
Average	1076.76±061.45	86.84	1136.63±49.42	1226.27±19.47	33.14	751.41±16.88	67.20±3.52	0.84	30.12±3.39	7.35	200.56±10.72	0.64

Table 2(c): The activity concentrations (Bqkg^{-1}, ppm, ppt) of U-238 series, Th-232 series, U-235 and K-40 (**%)**radionuclides for different sedimentary samples collected from GabalHomyer locality.

	U-238 series						U-235		Th-232 series		K-40	
ID	U-238 (Bqkg^{-1})	U-238 (ppm)	U-234 (Bqkg^{-1})	Ra-226 (Bqkg^{-1})	Ra-226 (ppt)	Pb-210 (Bqkg^{-1})	U-235 (Bqkg^{-1})	U-235 (ppm)	Avg.(Bqkg^{-1}) [Ac-228, Tl-208]	Th-232 (ppm)	K-40 (Bqkg^{-1})	K (%)
M-21	1785.54±109.79	143.99	1727.75±058.05	3054.79±34.35	82.56	1590.47±30.01	82.04±4.57	1.03	18.44±2.90	4.50	ULD	ULD
M-22	1758.64±076.27	141.83	2090.09±118.81	2168.98±29.69	58.62	1357.92±28.01	85.34±4.85	1.07	17.50±2.91	4.27	ULD	ULD
M-23	2756.76±102.95	222.32	2466.78±153.42	2317.43±37.41	62.63	1939.06±36.77	137.13±9.41	1.71	33.14±4.84	8.08	ULD	ULD
M-24	89.87±016.31	7.25	64.85±012.86	57.19±03.53	1.55	52.23±03.80	4.47±0.55	0.06	20.65±0.92	5.04	ULD	ULD
Minimum	89.87±016.31	7.25	64.85±012.86	57.19±03.53	1.55	52.23±03.80	4.47±0.55	0.06	17.50±2.91	4.27	ULD	ULD
Maximum	2756.76±102.95	222.32	2466.78±153.42	3054.79±34.35	82.56	1939.06±36.77	137.13±9.41	1.71	33.14±4.84	8.08	ULD	ULD
Average	1597.70±076.33	128.85	1587.37±085.63	1899.60±26.25	51.34	1234.92±24.65	77.24±4.84	0.97	22.43±2.90	5.47	ULD	ULD

Table 2(d): The activity concentrations (Bqkg^{-1}, ppm, ppt) of U-238 series, Th-232 series, U-235 and K-40 (**%)** radionuclides for different sedimentary samples collected from a site of GabalAllouga locality.

	U-238 series						U-235		Th-232 series		K-40	
ID	U-238 (Bqkg^{-1})	U-238 (ppm)	U-234 (Bqkg^{-1})	Ra-226 (Bqkg^{-1})	Ra-226 (ppt)	Pb-210 (Bqkg^{-1})	U-235 (Bqkg^{-1})	U-235 (ppm)	Avg.(Bqkg^{-1}) [Ac-228, Tl-208]	Th-232 (ppm)	K-40 (Bqkg^{-1})	K (%)
A-1	1818.64±056.50	146.66	ULD	4719.08±27.94	127.54	2545.29±23.40	89.57±04.82	1.12	80.65±02.97	19.67	447.79±15.84	1.43
A-2	2922.30±080.49	235.67	ULD	6778.81±37.83	183.21	3578.81±32.73	147.94±06.43	1.85	68.15±03.28	16.62	542.96±21.46	1.73
B-1	5556.15±160.02	448.08	6285.54±113.85	13833.59±74.77	373.88	8373.56±68.75	272.91±13.98	3.41	110.36±07.35	26.92	510.09±46.17	1.63
B-2	7955.74±145.69	641.59	9386.78±126.92	18162.73±55.42	490.88	11180.85±50.68	378.86±08.81	4.74	108.53±04.70	26.47	578.85±23.46	1.85
B-3	5502.42±183.13	443.74	14808.33±244.47	14036.93±66.11	379.38	10375.32±58.59	257.71±11.40	3.22	110.78±06.02	27.02	780.65±34.07	2.49
B-4	16816.28±200.33	1356.15	19463.30±205.99	22576.47±77.74	610.17	14414.71±70.10	843.26±15.81	10.54	72.69±11.26	17.73	672.41±36.05	2.15
C-1	5691.64±121.13	459.00	10685.61±162.38	7935.41±41.83	214.47	5365.48±33.96	257.37±06.88	3.22	108.31±07.05	26.42	805.85±23.09	2.57
C-2	6402.65±121.35	516.34	6816.80±128.60	5188.50±39.98	140.23	4655.56±34.57	294.33±07.17	3.68	94.36±03.95	23.02	612.14±23.42	1.96
D-1	9918.85±159.25	799.91	11063.38±189.01	9036.57±63.17	244.23	5536.11±39.81	529.96±12.84	6.62	126.87±05.10	30.94	836.39±32.19	2.67
D-2	7816.27±189.38	630.34	8916.30±197.85	7247.67±68.54	195.88	4344.41±37.80	404.38±12.96	5.05	127.72±06.87	31.15	799.93±36.23	2.56
Minimum	1818.64±056.50	146.66	ULD	4719.08±27.94	127.54	2545.29±23.40	89.57±04.82	1.12	72.69±11.26	16.62	447.79±15.84	1.43
Maximum	16816.28±200.33	1356.15	19463.30±205.99	22576.47±77.74	610.17	14414.71±70.10	843.26±15.81	10.54	127.72±06.87	31.15	836.39±32.19	2.67
Average	7040.09±141.73	567.75	10928.26±171.13	10951.58±55.33	295.99	7037.01±45.04	347.63±10.11	4.35	100.84±05.85	24.60	658.70±29.20	2.10

Looking at the values reported in the four different localities understudy, and considering longitude N33^{0}25' is the separation line which represent G.Allouga locality, the average activity concentration values for ^{238}U, ^{234}Uand^{226}Ra are decreasing east and west of that longitude (Fig. 2).
Figure 3 shows GabalAllouga has the highest activity concentration values of ^{235}U, ^{238}U, ^{234}U, ^{226}Raand ^{232}Thin the four localities at this time.

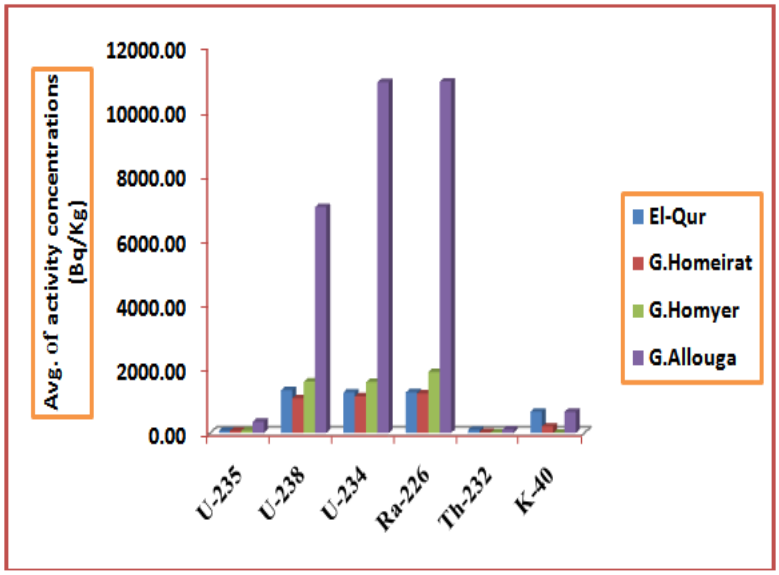

Figure 3: Averageactivityconcentrations(Bq/kg) for U-235, U-238, U-234,Ra-226, Th-232andK-40 for four different localitiesatsouthwestern Sinai, Egypt.

The average activity concentrations of U-238, Th-232 and K-40 for twenty six sedimentary samples in the studied area compared with the other countries are represented in Table 3 which showed variations between the different countries and wide variations between different localities in the same country. The activity ratio $^{226}Ra/^{238}U$ is used as indicator to assess the alteration process for the studied four localities. Uranium-238 mobility is more than radium-226 and thorium-232. Each locality contains different type of rocks, although for the same type of rock for the same or for different locality, the ratio $^{226}Ra/^{238}U$has different values (Table (4)). There are four ID Grades for alteration processes: I-1, I, I+1 and I+2 corresponding to $^{226}Ra/^{238}U$ activity ratios < 0.8, 0.8:1.2, 1.3:2 and > 2 respectively.

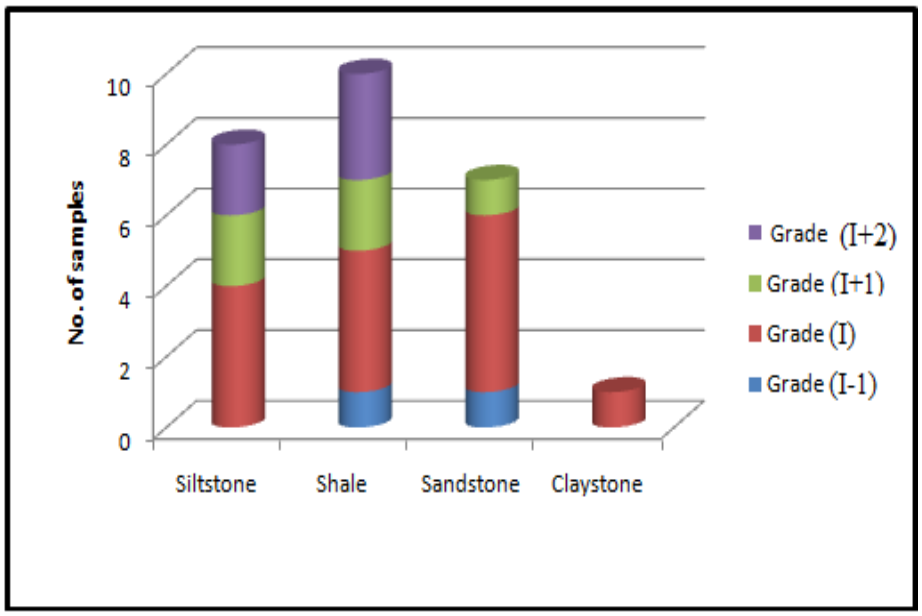

Figure 4: Classification of different rock samplesaccordinglytofour ID Grades.

Table 3: Average activity concentrations (Bqkg^{-1}) for **s**edimentary samples comparison between the present study and other previous studies in the world.

Country		U-238 (Bqkg^{-1})	Th-232 (Bqkg^{-1})	K-40 (Bqkg^{-1})	References
S-W Sinai, Egypt	G.Homyer	1597.70	22.43	ULD	**Present study**
	G.Homeirat	1076.76	30.12	200.56	
	G.Allouga	7040.09	100.84	658.70	
	El-Qur	1332.63	93.02	661.36	
Bangladesh		60.00	113.00	1002.00	[16]
S-W Nigeria		28.52	30.32	426.03	[17]
India		19.16	48.56	1146.88	[18]
Turkey (İkizdere Valley)		124.21	32.71	811.68	[19]
Turkey (Kaptanpaşa Valley)		19.46	24.48	609.66	[19]
Sharm El-Sheikh south Sinai, Egypt		48.20	60.70	1278.00	[20]
El Gor area southwestern, Sinai,Egypt		480.92	31.80	222.44	[21]

Siltstone sedimentary samples (Table (4) and Fig.4) have three Grades (I, I+1 and I+2) which express variation from no to slightly alteration in the direction of migration out of uranium. From Table (4), it can be concluded that most of the samples (14 samples) of Grade (I) are representing the whole rock types while two samples of Grade (I-1) are representing shale and sandstone. Five samples of Grade (I+1) are representing siltstone, shale and sandstone and five samples of Grade (I+2) are representing siltstone and shale.There is no migration-in or out for uranium (Grade I) in only claystone samples at El-Qur (Table (4)).

Table 4: The activity ratios $^{226}Ra/^{238}U$for twenty six sedimentary rock samples and their grades for all localities southwestern Sinai, Egypt.

Type of sedimentary rock	Locality	Sample ID	Ra-226/U-238 [<0.8] Grade (I-1)	[≃ 1] (0.8:1.2) Grade (I)	[>1] (1.3:2) Grade (I+1)	[>1] (>2) Grade (I+2)
Siltstone	El-Qur	Qur-1		0.9		
		Qur-1a		1		
		Qur-5		0.8		
		Qur-9			1.4	
	GabalHomeirat	M-13			1.7	
		M-14		1.1		
	GabalAllouga	A-1				2.6
		A-2				2.3
Shale	El-Qur	Qur-3	0.7			
		Qur-8		0.9		
	GabalAllouga	B-1				2.5
		B-2				2.3
		B-3				2.6
		B-4			1.3	
		C-1			1.4	
		C-2		0.8		
		D-1		0.9		
		D-2		0.9		
Sandstone	El-Qur	Qur-11		1.2		
	GabalHomeirat	M-11		1.1		
		M-12		1		
	GabalHomyer	M-21			1.7	
		M-22		1.2		
		M-23		0.8		
		M-24	0.6			
Claystone	El-Qur	Qur-8a		0.9		

5.1 Alteration Process Duration History

GabalAllouga has the highest value of activity concentration of uranium ($^{238}U + {}^{235}U$) for all localities, therefore more measurements and investigations will assist to give some information about the alteration processes history of this locality. Eight samples were taken within 3.5 meters thickness and isorad

contour map for uranium($^{238}U + {}^{235}U$) represented in Fig. 5. The sample B-4 has the highest uranium ($^{238}U + {}^{235}U$) value with 17659.5 Bqkg-1. The reason may because it is variegated shale, yellowish brown with yellow secondary uranium mineral as encrustation.

The Th-230/U-234 dating method is based on the radioactive decay series of U-238. The daughter isotope U-234 ($T_{1/2}$ = 2.45x10^5 a[22]) decays into the daughter isotope Th-230 ($T_{1/2}$ = 7.7x10^{4}a[23]) with the emission of alpha particle. The half-life ($T_{1/2}$) of U-238 ($T_{1/2}$ = 4.46x10^9 a,[22])is considerably longer than that ofU-234 and Th-230. The decay of uranium 234 to thorium 230 is part of the much longer decay seriesbegin in uranium 238 and ending in lead 206.As a consequence, radioactive equilibrium is almost approached after about one million years in most geologically old rocks and sediments. A characteristic of radioactive equilibrium is that all members of the decay series have the same specific activity. Thus, the activity ratios of any two of the series is equal to one. The samples which are not suitable for Th-230/ U-234dating may have behaved as open system for uranium or might have contained more than one detrital component.

The uranium dissolved in groundwater may be incorporated into a new system. The most simple ^{230}Th/^{234}U dating model presupposes that the new system contains solely uranium. Then, the numerical ^{230}Th/^{234}U clock starts at zero and a ^{230}Th/^{234}U AR (Activity Ratio) = 0. The radioactive disequilibrium between ^{230}Th and ^{234}Umaintains during ageing until the radioactive equilibrium is approached after about seven times the half-life of Th-230 (350-500 ka). This process is described by equation (1)[24].

$$\frac{^{230}\text{Th}}{^{234}\text{U}} = \left[\frac{^{238}\text{U}}{^{234}\text{U}}\right]\left(\mathbf{1} - \mathbf{e}^{-\boldsymbol{\lambda_{230}t}}\right) + \left(\frac{\boldsymbol{\lambda_{230}}}{\boldsymbol{\lambda_{230}-\lambda_{234}}}\right)\left[\mathbf{1} - \left(\frac{^{238}\text{U}}{^{234}\text{U}}\right)\right][\mathbf{1} - \mathbf{e}^{-(\boldsymbol{\lambda_{230}-\lambda_{234}})\mathbf{t}}] \qquad (1)$$

Where t is the alteration's age of the sample, (Th-230/U-234) and (U-238/U-234) are the measured activity ratios, and λ_{230} and λ_{234} are the decay constants of Th-230 and U-234, respectively. The last equation was evaluated using (Maple 12) program.

Different correction methods have been developed in U-series dating to deal with detrital contamination. The purpose of all correction methods is to subtract from the total measured activities of Th-230, U-234 and U-238, which have originated in detritus. A review of several correction methods can be found in [25]. One of those methods, an isochron solution was first proposed to date fossil shells[26] to estimate ages of marine terraces.The detrital Th-230 decays during ageing while the activity of the radiogenic Th-230* increases as a result of the decay of U-234. Hence the Th-230/U-234 age error due to detrital contamination decreases with increasing age. The two or more sources of Th-230 (one radiogenic and at least one detrital) have to be distinguished and quantified to enable a correction of the Th-230/U-234 age {equation (2)}[24].

$$\left[^{230}\text{Th}_{\text{total}}\right] = \left[^{230}\text{Th}^{*}\right] + \left[\frac{^{230}\text{Th}}{^{232}\text{Th}}\right]\left[^{232}\text{Th}\right] = \left[^{230}\text{Th}^{*}\right] + \text{f}\left[^{232}\text{Th}\right] \qquad (2)$$

wheref is the thorium index. Standard deviation □(equation 3{a,b}) of the detritus-corrected 230Th*/U and 230Th/U age can be calculated[27].

$$+\sigma t = \left[^{234}\text{U} - \sigma^{234}\text{U}\right]/[\,^{230}\text{Th} + \sigma\,^{230}\text{Th}]\text{a} \qquad (3)$$

$$-\sigma t = \left[^{234}\text{U} + \sigma^{234}\text{U}\right]/[\,^{230}\text{Th} - \sigma\,^{230}\text{Th}]\text{b}$$

The slope of the straight mixing line (Fig. 6) in the plot of (230Th/232Th) versus (234U/232Th) equals the actual 230Th*/234U which is the important parameter to calculate (230Th*/U) ages. The intersection of the isochron on the Y axis yields the present (230Th*/232Th) or the present thorium index f. This factor is then used to correct each sample separately [28].Any correction of radiometricallydetermined $^{230}\text{Th}/\text{U}$ages for detrital ^{230}Th is negligible if the measured $^{230}\text{Th}/^{232}\text{Th}$of any sample is smaller than 20 because in this case the detrial ^{230}Th activity is very low [27].

The isochron-corrected ages of six samples (B-1, B-3, B-4, C-2, D-1 and D-2) with a (230Th/232Th) ratio of 0.744 as shown in (Fig. 6) and then calculate the actual radiogenic 230Th and age of alteration process{ equation (2)}.

Eight samples from GabalAllouga were chosen to calculate their alteration's processes age (Table (5)). The alteration's processes time ranged between 106.31 ka for sample C-1 (Shale, brownish, fissile, ferruginous and soft) and 246.11 ka for sample C-2 (Shale, yellowish brown, 4 m to the north from sample C-1). The upper (D) bed has Grade (I) and its alteration processes duration time ranged between 231.52 ka and 222.08 ka. The alteration processes duration time for (D) bed has Grade I is above 200 ka. The (C) bed has Grade (I+1) and Grade (I) which mean there is migration out and no migration in or out for uranium with alteration processes time ranged between 106.31 ka and 246.11 ka respectively. The (B)bed has Grades (I+1) and (I+2), that is mean there is only migration out with alteration processes time ranged between 109.94 ka and 228.92 ka.

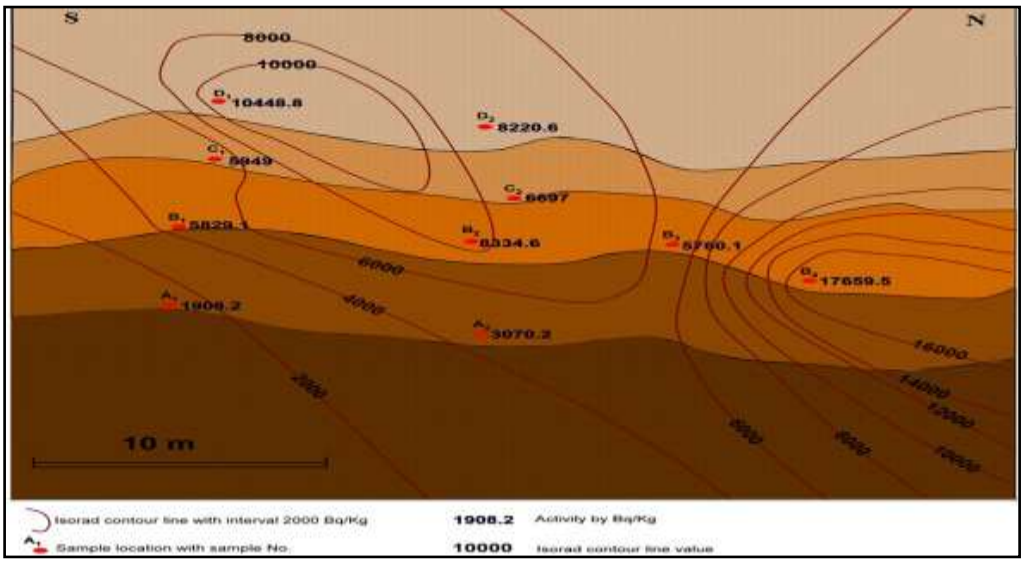

Figure 5: Isorad contour map for samples collected from a site ofGabalAllouga locality.

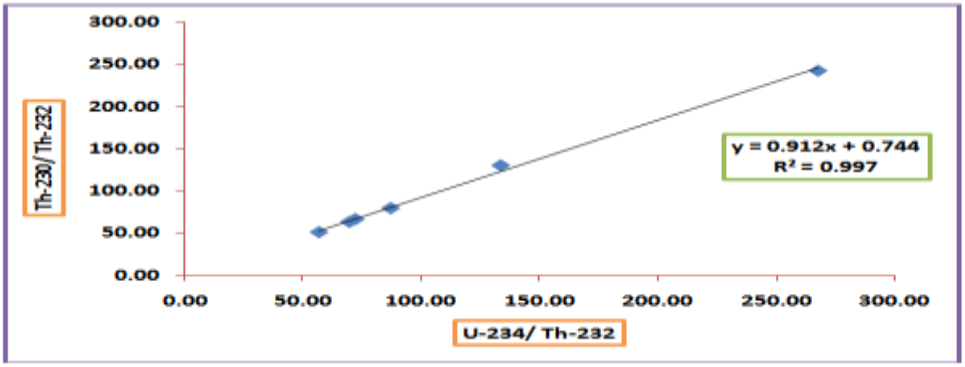

Figure 6: The relation between Th-230/Th-232and U-234/Th-232.

Table 5: Specific activities (Bqkg^{-1}) of U-238, U-234, Th-230 and Th-232, isotopic activity ratios and alteration process duration history for sedimentary samples at a site of GabalAllouga locality.

ID	U-238 (Bqkg^{-1})	U-234 (Bqkg^{-1})	Th-230 (Bqkg^{-1})	Th-232 (Bqkg^{-1})	U-234/ U-238	Th-230/ Th-232	Th-230/ U-234	Age(ka) Measured	Age(ka) Corrected
B-1	5556.15±160.02	6285.54±113.85	5603.88±207.19	110.36±07.35	1.131	50.78	0.892	223.25 +1.06 -1.19	213.16 +1.08 -1.20
B-3	5502.42±183.13	14808.33±244.47	14415.93±328.98	110.78±06.02	2.691	130.13	0.974	206.57 +0.99 -1.07	203.74 +0.99 -1.07
B-4	16816.28±200.33	19463.30±205.99	17647.00±330.66	72.69±11.26	1.157	242.77	0.907	231.27 +1.07 -1.14	228.92 +1.07 -1.14
C-2	6402.65±121.35	6816.80±128.60	6248.54±216.27	94.36±03.95	1.065	66.22	0.917	257.18 +1.03 -1.15	246.11 +1.05 -1.16
D-1	9918.85±159.25	11063.38±189.01	10057.76±331.13	126.87±05.10	1.115	79.27	0.909	238.22 +1.05 -1.16	231.52 +1.06 -1.17
D-2	7816.27±189.38	8916.30±197.85	8050.86±305.28	127.72±06.87	1.141	63.03	0.903	230.52 +1.04 -1.18	222.08 +1.06 -1.19
B-2	7955.74±145.69	9386.78±126.92	6102.57±144.70	108.53±04.70	1.180	56.23	0.650	112.23 +1.48 -1.60	109.94 +1.50 -1.62
C-1	5691.64±121.13	10685.61±162.38	7087.00±195.54	108.31±07.05	1.877	65.44	0.663	108.06 +1.44 -1.57	106.31 +1.46 -1.59

VI. Conclusion

The studiedtwenty six different rock samples from four localities at southwestern, Sinai, Egypt shown that:

(1) The average activity concentration values of ^{238}U were approximately 42, 34 and 50 times the world average value for EL-Qur, GabalHomeirat, andGabalHomyerlocalities respectively.

(2) For GabalAllouga, the average activity concentration value of ^{238}U was nearly two hundred and twenty times the world average value.
(3) Th-232 average activity concentrations for GabalHomeiratand GabalHomyer localities were less than the world value while it is nearly twice the world average value for El Qur and GabalAllougalocalities.
(4) Ra-226 average activity concentration was the most contribution forGabalHomeirat, GabalHomyer and GabalAllouga.
(5) U-238averageactivity concentration was the most contribution forEL-Qur.
(6) There is no migration in or out of uranium recorded for fourteen samples according to $^{226}\mathrm{Ra}/^{238}\mathrm{U}$ ratio.
(7) The uranium migration was recorded for twelve other samples.
(8) There are four Grades for alteration process according to the activity ratio $^{226}\mathrm{Ra}/^{238}\mathrm{U}$.
(9) The alteration's processestime ranged between 106ka and 246ka for eight samples from GabalAllouga.

These further confirm that the sediments of the four localities are unsuitable for use as building materials. Some precautions and recommendations should be taking into consideration for the public in this area.

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