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Uranium migration in Paleozoic lateritic Paleosol Samples, Southwestern Sinai, Egypt

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Abstract: This paper spotlights on determining the concentrations and distribution of radionuclides in paleozoic lateritic paleosol samples from El-Gor area, southwestern Sinai, Egypt, using high-resolution gamma spectrometry. The concentrations of eU (ppm), eTh (ppm), and K (in percentage) were determined. The obtained data of eU, eTh and their ratios were used for calculations of uranium migration tendency and its remobilization. The mean activity concentrations of ^{238}U, ^{235}U, ^{232}Th, ^{226}Ra and ^{40}K were 480.92 $Bqkg^{-1}$, 22.38 $Bqkg^{-1}$, 31.80 $Bqkg^{-1}$, 401.98 $Bqkg^{-1}$ and 222.44 $Bqkg^{-1}$, respectively. The migration study showed that the uranium is migrated-in in the paleosol and this is also confirmed by the mean ^{238}U / ^{226}Ra ratio which in the side of uranium enrichment than radium.

Key words: Paleosol**;** El-Gor area**;** Gamma ray spectroscopy; Uranium migration

INTRODUCTION

Paleosols are soils that formed on landscapes of the past in a non-marine stratigraphic record (Bronger and Catt,1989 and Kanhalangsy,1997). Soils were formed through long geologic time affected by the earth's atmosphere and life. The oldest paleosol recognized in about 3.1 billion years and is developed on granitic basement rocks (Edelman *et al*., 1983). Retallack (1986) pointed to ancient soils may even represent the oldest sedimentary rock record, formed approximately 3.8 billion years before present. The studied paleosol is enclosed in the lower Carboniferous (325 m.y) Um Bogma ormation. The mineral composition of this paleosol is mainly of calcite, dolomite, gibbsite and kaolinite in addition to halite, gypsum and iron-oxyhydroxides (Attia *et al*., 2011). Many features of this soil gave lights to complicated paragenetic history involving dissolutions, replacements and precipitations. These processes are the main factors which affecting in the uranium disequilibrium with its daughters.

The main aim of this study can be presented in the following:

(1) To calculate the uranium migration in the lateritic soil samples.
(2) An attempt to use the ^{226}Ra / ^{238}U ratios as an indication for the migration of uranium and or radium.
(3) The correlations between ^{238}U with ^{235}U, ^{226}Ra, ^{232}Th and ^{40}K.

Geologic Setting:

El Gor area is located in southwestern Sinai at the northwestern part of Um Bogma area (figure1). It is also within the environs of Abu Zeneima town on the eastern coast of the Gulf of Suez, southwestern Sinai Peninsula. These environs are bounded between longitudes 33° 18' 00" - 33° 21' 00" E and latitudes 28° 59' - 29° 01' N covering an area of about 700 km^2. El Gor area is located to the north of Wadi El Samra and Wadi Baba. The area is about 50 km^2 and can be reached by asphaltic and desert roads to the famous location of Um Bogma area.

The soil sediments in Farsh El Ebidiya location are hosted in a low land area (200 x 100 m) hosted in the dolomite of upper dolostone member of Um Bogma formation. The studied samples are collected from ten trenches (table1). This table (1) shows the field description of the collected samples.

Analytical Technique:

Seventeen paleosol soil samples were collected from El-Gor area, southwestern Sinai, Egypt. These samples were prepared for gamma (γ) ray spectrometric analyses using High Purity Germanium (HPGe) detector.

Methodology:

Gamma-ray spectrometry is an indirect technique which gives the U analysis as, equivalent uranium, eU and is equal to the uranium concentration only if there is radioactive equilibrium in the rock being analyzed (Asfahani and Abdul-hadi, 2001; Asfahani,2002). For radiometric analysis, each dried sample was splitted by quartering, weighed and transferred to 200 ml capacity polyethylene marinelli beakers then sealed and stored for 30 days to prevent the escape of the radiogenic gases ^{222}Rn and ^{220}Rn, and to allow the attainment of radioactive equilibrium in the decay chain. After the attainment of secular equilibrium between ^{238}U, ^{232}Th and their progenies, the samples were subjected to gamma ray spectrometric analysis. During an accumulating time approximately 24 hours, each sample was measured. An empty cylindrical plastic container (polyethylene

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marinelli beaker) was placed in the detection system, for a counting period of 48 hours in order to collect the background count rates. The samples were investigated using gamma-ray spectrometry with (HPGe) detector model (GEM-50210-P), a P-type crystal, from EG & G ORTEC. This detector has a resolution (FWHM) of 1.9 keV for the 1332 keV gamma-ray line of ^{60}Co and a relative efficiency of about 50%. The system was calibrated for energy to display gamma photopeaks between 63 and 3000 keV.

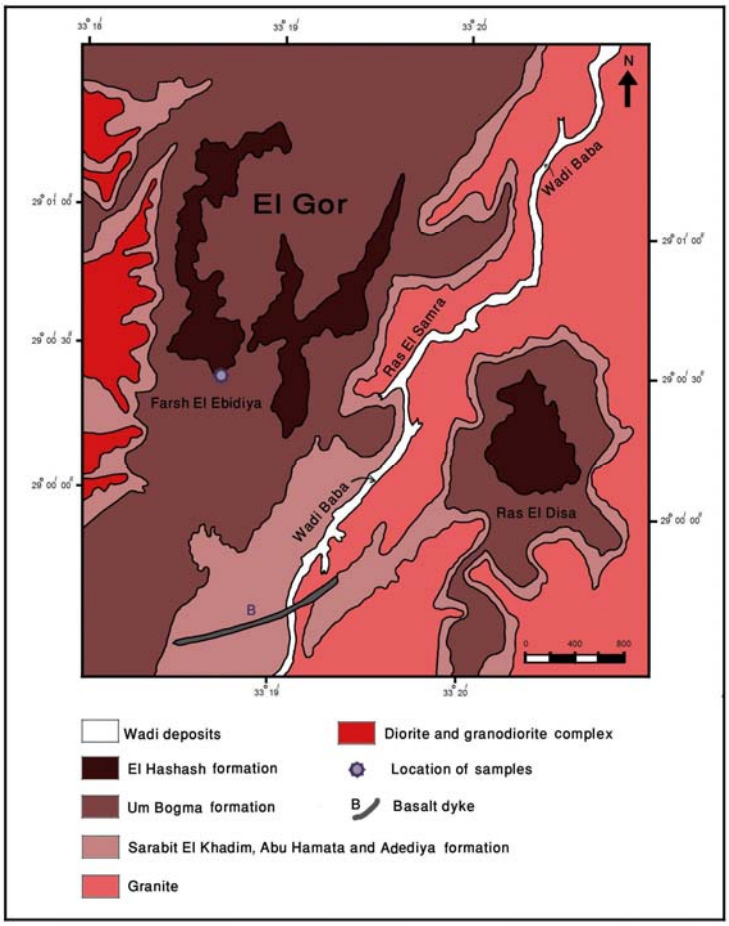


Fig. 1: Geologic map of El Gor Area, Southwestern Sinai, Egypt (After Attia *et al*., 2011)

Table 1: Field description of the collected samples

Number of Trench	Symbol of bed	Field Description
1	a	At the base of the first trench, black, medium hard to soft soil sediments
	b	Reddish brown soil, powdery as ocher.
	C	Gravellitic shales, brown and white (may be alunite or kaolinite) pebbles.
2	b	At the middle of trench No. 2, black soil with whitish patches, salty.
3	a	It is soft to medium hard shaley, black soil.
	b	Brown soil,soft, partially compact.
4	a	It is black, soft, soil, very light mainly gibbsite.
	b	Black, mottled, brown soil.
5	a	Dark gray, soft, friable soil with whitish patches.
6	b	Black, soft to weakly cemented very light soil, mainly gibbsite.
	c	Pebbly cemented with carbonate, light brown, reddish to whitish soil.
7	b	Dark gray to black soil.
8	a	Highly weathered dolomite or calcrete (dolocrete), light brown, the exposed part 0.3 m.
	b	Brown to yellow soil with black patches.
9	a	Black soil with yellow patches.
	b	Yellowish brown, soil, soft, fissile shale.
10	c	Black, mottled with white patches soil.

a): base, b): middle and c): top

The efficiency calibration was performed by using three well-known reference materials obtained from the International Atomic Energy Agency for U, Th and K activity measurements: RGU-1, RGTh-1 and RGK-1 (IAEA (1987), Anjos, *et al*., 2005). Absolute efficiency calibration of the gamma spectrometry system was carried out using the radionuclide specific efficiency method in order to reduce the uncertainty in gamma-ray intensities, as well as the influence of coincidence summation and self-absorption effects of the emitting gamma photons (Stoulos, *et al*., 2003). The sample containers were placed on top of the detector for counting. The same geometry and size were used for both the samples and the reference materials (Turhan and Gu¨ndu¨z L, 2008). The uranium standard (RGU-1) is U-ore diluted with silica with 4940 $Bqkg^{-1}$ of ^{238}U, 228 $Bqkg^{-1}$ of ^{235}U, a negligible amount of ^{40}K (less than 0.63 $Bqkg^{-1}$) and some traces of ^{232}Th (less than 4 $Bqkg^{-1}$). The thorium standard (RGTh-1) is Th-ore diluted with silica having 3250 $Bqkg^{-1}$ of ^{232}Th, but containing some ^{238}U (78 $Bqkg^{-1}$) and ^{40}K (6.3 $Bqkg^{-1}$). The potassium calibration standard (RGK-1) is produced from high purity (99.8%) potassium sulphate with 14000 $Bqkg^{-1}$ of potassium with uranium and thorium contents lower than 0.001 and 0.01 parts per million (ppm), respectively (Anjos, *et al*., 2005).

Uranium Migration:

It is worth noting that the uranium and thorium hold similar characteristic in their ionic radii which is why they both behave isochemically during the process of magmatic (Abu Deif *et al*., 2001). However, during weathering and other crustal processes, U^{4+} gets oxidized easily and turn to U^{6+} which is known to be soluble in ground water, while Th does not, as it maintain insoluble in the oxidization zone. Hence, for the original concentration of U in the rock, eTh can be used as proxy (Aswathanaray, 1985).

A good indicator for the migration of uranium is the eU/eTh ratio, whereas if the ratio remains approximately constant in rocks of the same type so the migration of uranium has not occurred. The half-lives of uranium and thorium are long enough to consider the ratio of eU/eTh measured to represent the original ratio in the rock (NMA, 1999). We can calculate the value of uranium migration (U_m) for a given rock unit by subtracting the original uranium content (U_o) from the present uranium content (U_p) (NMA, 1999), as follows:

$$U_m = U_p - U_o \quad (1)$$

The following equation can be used to compute theoretically the original uranium concentration (U_o)

$$U_o = eTh \times (\, regional\ eU / eTh \,) \quad (2)$$

Where eTh is the average thorium content (in ppm) in a certain rock unit, and the regional eU/eTh is the average regional eU/eTh ratio in different rock units (Abu-Deif *et al*., 2001; Asfahani *et al*., 2007).

The extent of uranium migration rate (U_{me} %) can be computed through the knowledge of both U_o and U_p using the following equation:

$$U_{me}\ \% = (U_m / U_p) \times 100 \quad (3)$$

A good indicator for the determination of the migration degree and its direction is the U_{me}%. The migration is into the unit if the U_{me}% is positive, while if it is negative then the migration is out of the unit.

RESULTS AND DISCUSSIONS

For the actinium series, ^{235}U gamma energies of 143.8, 163.4 and 185.7 keV were taken to represent the ^{235}U activity (Yu¨cel, 1998). The specific activity of ^{40}K was measured directly by its own gamma-ray at 1460.8 keV, while activities of ^{226}Ra and ^{232}Th were calculated based on the weighed mean value of their respective decay products in secular equilibrium. The specific activity of ^{226}Ra was measured using the 186.1 keV from its own gamma-ray (after the subtraction of the 185.7 keV of ^{235}U). The specific activity of ^{232}Th was measured using the 338.4 and 911.2 keV from ^{228}Ac and 583 keV and 2614.4 keV from ^{208}Tl. ^{238}U activity was determined indirectly from the gamma rays emitted by its daughter products (Sutherland, 1990) while ^{235}U activity was determined directly by its gamma ray peaks (Po¨lla¨nen, *et al*., 2003). The activity of ^{238}U cannot be directly determined, since the isotope emits only a weak (0.064%) gamma-ray at 49.55 keV. However, any of the gamma-emitting daughter nuclides in equilibrium with ^{238}U such as ^{234}Th or ^{234m}Pa could be used for this purpose (Papachristodoulou 2003, Zhang W 2011). The daughter radionuclides (^{234}Th and ^{234m}Pa) were selected as an indicator of ^{238}U activity. Progeny of ^{238}U are relatively short-lived, and thus it is reasonable to calculate gamma radiations of this sequence as ^{238}U. Therefore, secular equilibrium was assumed between ^{238}U and ^{234}Th

and ^{234m}Pa, and thus ^{234}Th and ^{234m}Pa activities determined from the 1001 keV photopeaks, were assumed to represent actual ^{238}U activities.

The results represented by the specific activity (Bqkg^{-1}) of different radionuclides in the studied samples are presented in table (2). Table (2) shows the activity concentration of ^{238}U ranging from 95.84 Bqkg^{-1} to 1169.08 Bqkg^{-1} with an average 480.92 Bqkg^{-1}, which is higher than the recommended value which is documented at 35 Bqkg^{-1} (UNSCEAR, 2000). ^{235}U is ranging between 4.65 Bqkg^{-1} and 52.57 Bqkg^{-1} with an average of 22.38Bqkg^{-1}. ^{232}Th is ranging between 10.90 and 76.13 Bqkg^{-1} with an average of 31.80 Bqkg^{-1} which is in agreement with the world wide average which is documented at 30 Bqkg^{-1} (UNSCEAR, 2000). ^{226}Ra is ranging between 48.43 Bqkg^{-1} and 863.97 Bqkg^{-1} with an average of 401.98 Bqkg^{-1} which is higher than the recommended value which is documented at 35 Bqkg^{-1} (UNSCEAR, 2000), but ^{40}K is ranging from 65.38 Bqkg^{-1} to 526.53 Bqkg^{-1} with an average 222.44 Bqkg^{-1} which is in agreement with the world wide average (400 Bqkg^{-1} UNSCEAR, 2000).

Table 2: The activity concentrations of the ^{238}U, ^{235}U, ^{232}Th, ^{226}Ra and ^{40}K in Bqkg^{-1} for the studied samples.

Sample No.	^{238}U Bqkg^{-1}	^{235}U Bqkg^{-1}	^{232}Th Bqkg^{-1}	^{226}Ra Bqkg^{-1}	^{40}K Bqkg^{-1}
1*a**	1169.08	52.57	51.44	863.97	291.57
1b	656.40	31.04	37.73	624.04	281.32
1c	241.96	11.39	17.71	267.78	108.35
2b	280.69	13.39	13.47	216.95	72.04
3a	646.11	32.17	15.79	435.14	163.52
3b	434.51	20.83	14.67	374.71	124.06
4a	733.80	32.74	45.68	587.77	215.79
4b	515.77	23.88	31.85	374.80	319.12
5a	203.52	9.50	39.09	97.44	332.79
6b	550.10	26.38	13.85	369.58	139.22
6c	575.26	27.06	10.90	726.35	65.38
7a	229.35	10.48	32.92	122.98	205.65
8a	95.84	4.65	12.14	48.43	73.04
8b	459.94	20.74	76.13	425.00	470.31
9a	258.77	12.11	35.88	231.26	170.75
9b	428.43	19.13	64.72	351.20	526.53
10c	696.06	32.36	26.63	716.26	221.97
average	480.92	22.38	31.80	401.98	222.44

*: The numbers of trenches

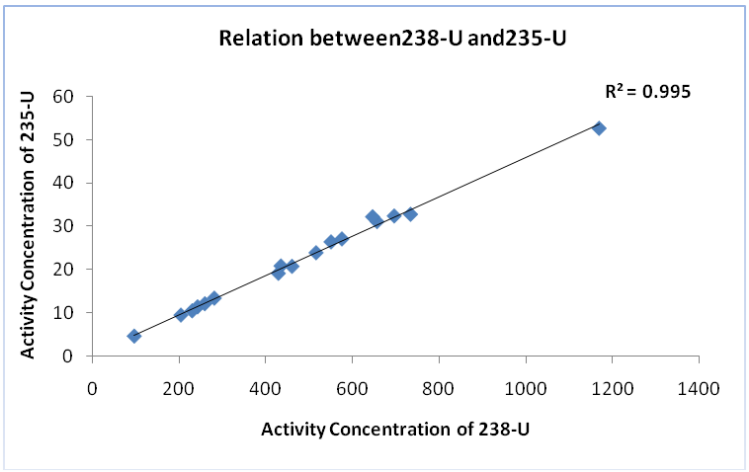
**: Symbols of beds in the section

The relation between ^{235}U and ^{238}U activities (figure 2a) shows a good correlation (0.995) because the activity ratios between them are natural. The relation between ^{226}Ra and ^{238}U activities (figure 2b) shows a strong correlation (0.844), which clarifies the behavior of ^{226}Ra with its parent ^{238}U. The ratio ^{238}U / ^{226}Ra in the whole samples is in the direction of uranium enrichment than radium except in the three samples 1c, 6c and 10c which show radium higher than uranium. The well noticed thing is that the three beds represent the surface beds (top) in their sites which may be subjected to later radium mobilization with different degrees. ^{40}K makes a very weak relation (0.039) with ^{238}U (figure 2 c), while it is a good relation (0.828) with ^{232}Th (figure3a) as both (^{232}Th and ^{40}K) are less mobile during soil formation in opposite to U^{6+} which is mobile in these conditions. The same case is noticed between ^{238}U and ^{232}Th which have very low correlation (0.064) as in figure (3b).

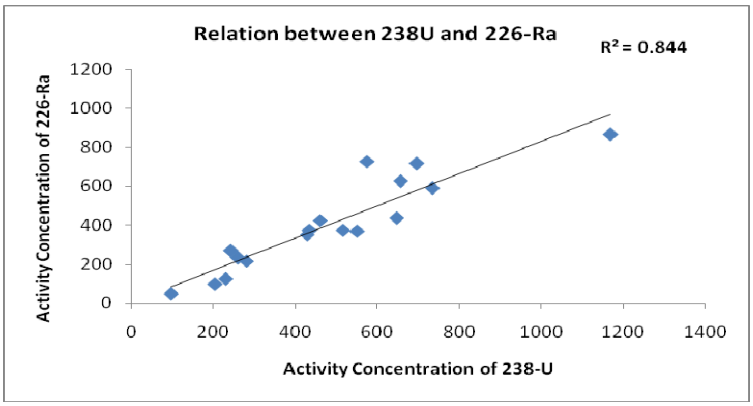
With respect to the uranium migration the values of the present uranium, original uranium content, uranium migrated, and uranium migration rate for 17 paleosol samples in El-Gor area are shown in table (3). The present uranium content (U_p) for the 17 paleosol samples ranging from 7.73 to 94.28 ppm with an average of 38.78 ppm. The original uranium content (U_o) was calculated by Equation (2) for the studied samples, which ranging from 3.37 to 23.56 ppm with an average of 9.84 ppm.

The uranium migrated content (U_m) can be calculated with the formula $U_m = U_p - U_o$, these values ranging from 3.97 to78.36 ppm with an average of 28.94 ppm. The uranium migration rate (U_m %), which was given by equation (3) is ranging from 26.31% to 92.73% with an average of 68.30%. It can be noticed that, the minimum value of the present uranium content coincide with the minimum value of uranium migration for the sample 8a {At the base of trench number 8, it is highly weathered light brown dolomite or calcrete (dolocrete)}. On the other hand, the maximum value of the present uranium content coincide with the maximum value of uranium migrated but it have not the highest migration rate in the sample 1a which is black, medium hard to soft soil sediments.

(a)



(b)



(c)

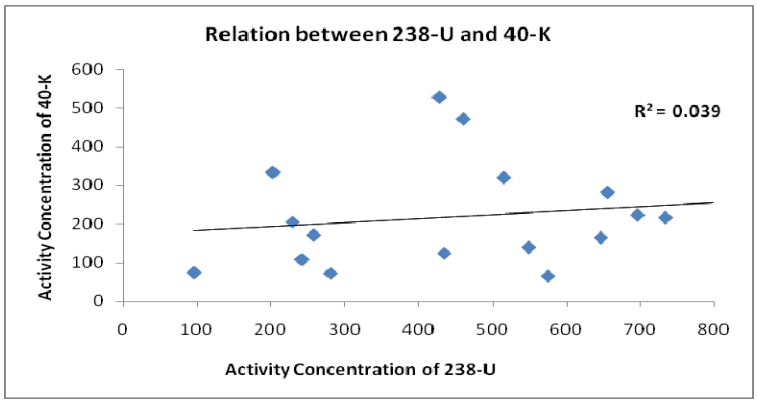
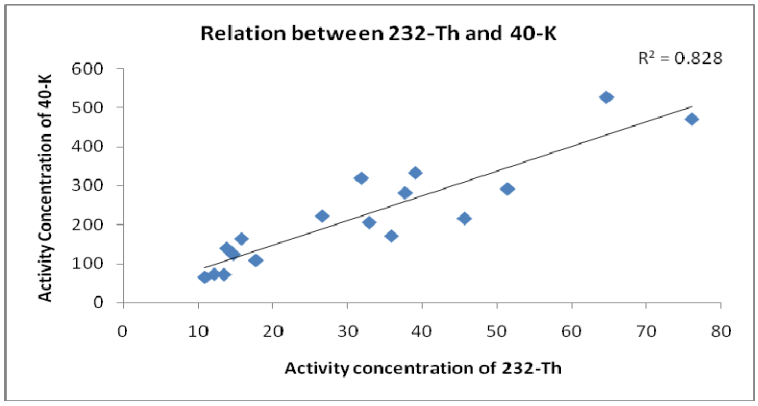


Fig. 2: (a) The relation between ^{235}U and ^{238}U activities, (b) the relation between ^{226}Ra and ^{238}U activities, (c) the relation between ^{40}K and ^{238}U activities.

(a)



(b)

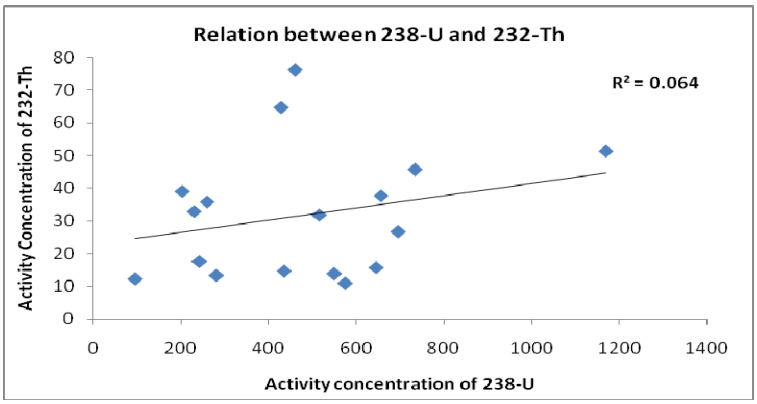


Fig. 3: (a) The relation between ^{232}Th series and ^{40}K activities, (b) The relation between ^{232}Th series and ^{238}U activities.

Table 3: The values of the present uranium, uranium migrated and uranium migration rate for 17 paleosol soil samples in El-Gor area.

Sample No.	Thickness (m)	eTh ppm	Present uranium (U_p) ppm	Original uranium (U_o) ppm	Uranium migrated (U_m) ppm	Uranium migration rate %
1*a**	0.50	12.73	94.28	15.92	78.36	83.12 %
1b	0.50	9.34	52.94	11.67	41.27	77.95 %
1c	0.75	4.38	19.51	5.48	14.03	71.91 %
2b	0.50	3.33	22.64	4.17	18.47	81.59 %
3a	0.70	3.91	52.11	4.89	47.22	90.62 %
3b	1.50	3.63	35.04	4.54	30.50	87.04 %
4a	0.60	11.31	59.18	14.13	45.05	76.12 %
4b	1.20	7.88	41.59	9.85	31.74	76.31 %
5a	0.60	9.68	16.41	12.09	4.32	26.31 %
6b	0.60	3.43	44.36	4.28	40.08	90.34 %
6c	1.20	2.70	46.39	3.37	43.02	92.73 %
7b	0.30	8.15	18.50	10.19	8.31	44.93 %
8a	0.30	3.00	7.73	3.76	3.97	51.40 %
8b	1.50	18.84	37.09	23.56	13.54	36.49 %
9a	1.00	8.88	20.87	11.10	9.77	46.81 %
9b	1.00	16.02	34.55	20.03	14.52	42.04 %
10c	0.70	6.59	56.13	8.24	47.89	85.32 %

*: The numbers of trenches.
**: Symbols of beds in the section

Conclusions:

1) The average activity concentrations for both ^{238}U and ^{226}Ra of El-Gor area are higher than the permissible value.
2) The ratios ^{238}U / ^{226}Ra are higher than unity in the whole samples except three samples.
3) The average activity concentrations of ^{235}U, ^{232}Th and ^{40}K are in agreement with the permissible value.
4) There is a natural relation between ^{235}U and ^{238}U.

5) There is a good relation between ^{238}U and ^{226}Ra.

6) There is a weak relation between ^{238}U and for both ^{232}Th and ^{40}K, and this normalized in these types of sediments.

7) There is a good relation between ^{232}Th and ^{40}K, as both are low mobile.

8) The uranium migrated content (U_m) for the studied samples is a positive value with different migration rates, which means the migration in of uranium.

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