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Research Article

DISTRIBUTION OF NATURAL RADIONUCLIDES FOR SEDIMENTRY ROCK SAMPLES FROM SOUTHWESTERN SINAI AND THEIR ENVIRONMENTAL IMPACTS, EGYPT

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ABSTRACT

Some sedimentary rock samples were collected from Um-Bogma area, Sinai for investigating by γ-ray spectrometry. The main goal of the current study was estimation the specific activities of natural radionuclides; analyze the data by statistical calculations to get the correlation ratios between radioelements and their ratios, to give information about enrichment and depletion as a result of alteration processes. The exposure risk was determined through dose, equivalent radium and annual effective dose calculations. This study reveals in general that all samples are exceeding the world permissible safe criteria and consider a risk source for human environment. Fractionation is noticed in one sample which showed migration out of uranium.

INTRODUCTION

Some sedimentary rocks represent the raw materials as they are the basics of most industries. There are many continuous investigations for determining natural series and non-series radionuclides all over the world. Radionuclide is a source of risk to humanity. Human beings are directly exposed to external and internal ionizing radiation from radionuclides through several processes (Khan *et al*., 2010; UNSCEAR, 2000; IAEA 1999; Kovler, 2016). The distribution of radionuclides varies from area to another due to many reasons like the geological localities and natural chemical reactions as well as man activities (Augustine *et al*., 2010). It is expected that radionuclides variation from low to high activity but it is not constant over all time. (UNSCEAR 2000, IAEA 2005). So we must study and measure the behavior of gamma radiation continuously to avoid problems which occur on human environment as a result of this tampering.

Really, the varieties of analytical techniques for radionuclides are advisable to obtain accurate and interpretation results from physical-chemical conditions so the scientists with different specializations should work together to give obvious and complete picture about the behavior of distribution and equilibrium among radionuclides. The relative activities or abundance of a parent and daughter radioactive nuclei provide basic information about the type of equilibria and enrichment depletion processes like the measured ratios of $^{238}U/^{235}U$, $^{234}U/^{235}U$, $^{234}U/^{238}U$, $^{226}Ra/^{238}U$, $^{230}Th/^{238}U$, $^{230}Th/^{234}U$, $^{226}Ra/^{230}Th$ and $^{210}Pb/^{226}Ra$. The measured activities and ratios of a daughter produced from parent nuclei depend on their decay constant. So there are three propabilities of equilibria: 1) The activity of the daughter will increase with time until it equals the activity of the parent this occurs when $\lambda_1 << \lambda_2$, at which case the parent and daughter are at secular equilibrium. 2) The parent and daughter are equal or nearly equal in the half life time this means that $\lambda_1 \sim \lambda_2$, in this case called transient equilibrium. 3) The half life time of the parent is less than that of the daughter, it means that the parent decays faster than the daughter $\lambda_1 > \lambda_2$; this case called disequilibrium or No-equilibrium. (Cooper *et al*; 2003, Pekala *et al*; 2010). In the current study, the researcher uses γ-radiometric analysis as one of physical techniques to obtain data about the distribution and equilibrium of naturally occurring radioactive materials (NORM) through relative abundance between radionuclides to

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explain the enrichment or depletion operation depend on rock structures and history formation. (Chiozzi *et al*; 2002). Finally these results can be used as data base for future study

Geologic Setting

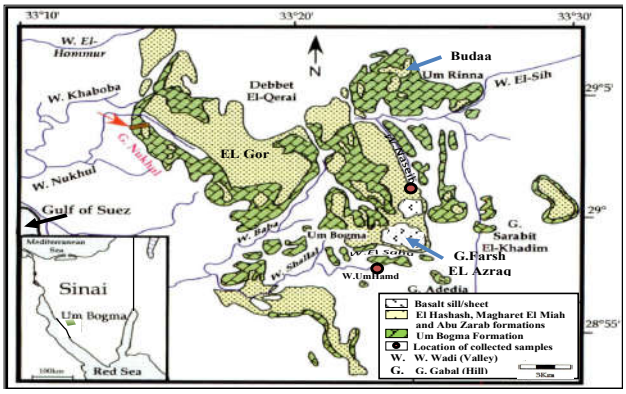

Figure 1 Geologic map of Um Bogma area, southwestern Sinai (after Kora and Jux, 1986 with modifications).

The main geologic rock unit in this study is the Um Bogma Formation (lower Carboniferous. 325 m.y). This formation is subdivided into three members as following from base to top:

Lower shaley- ore member

It is consisted of black shale with their sandy dolostone . This member was subjected to karstification (El Sharkawi *et al*., 1989) and lateritization processes (El Aassy *et al*., 2000). This rock unit is uraniferous with low to moderate uranium and low thorium contents especially in the black shale and the high carbonate contents.

Middle-Marly dolostone – siltstone member

It is also karstified and lateritized with high uranium contents than the previous rock unit and represent the main target of this study.

Upper dolostone member

It is consisted of bedded dolostone with thin shale interbeds with thickness 3-4 m. It is the lowest in the uranium contents.

Experimental Procedures

Sampling and sample preparation

Various types of rock samples were collected from different sites in vertical sequences at the studied area (Fig.1) for quantitative – qualitative analyses. Table (1) represents the detailed lithologic specification of the studied samples. The representative collected samples were thoroughly ground to fine size and divided into four parts (quartering) for distribution its component then bottled in a specific plastic bottles (marinelli beakers) with 200 ml capacity then sealed and stored for minimum 4 weeks before counting in order to allow the equilibrium between ^{226}Ra and ^{222}Rn and its decay products. ^{232}Th may be in equilibrium in most geological setting (Chiozzi *et al*; 2002, Michalis *et al*; 2003). Each sample was put in the shielded HPGe detector and measured for accumulating time nearly 24h. The environmental gamma background at the laboratory site has been determined using empty marinelli beaker under the same condition of sample detection. It is noted that small contribution of the environmental γ-ray background at the laboratory site has been subtracted from the spectra of the measured samples.

Table 1 Description of the different lithology of studied samples

Sample No.	Lithology
H.1	Calcareous shale, black, fissile, carbonaceous.
H.3	Sandy dolomite, gray to black, medium hard.
H.4	Calcareous siltstone, black, soft to medium hard.
H.5	Shale, dark brown with black patches, partly compact.
H.7	Sandy dolostone, black, medium hard to hard.
H.8	Sandy dolomite, gray, medium hard with Cu and uranium mineralizations.
H.9	Shale, black, partially fissile, Calcareous.

Measurement setting and calculations

Direct measurement of gamma rays (non-destructive method) was measured by hyper pure Germanium detector (HPGe). The spectrometry system gives us a longer wide photo-peak spectrum. It consists of co-axial and produced in a vertically n-type material. The crystal connects with a copper rod immersed in liquid nitrogen Dewar to prevent warm up to room temperature. The detection efficiency is 60% of 3"x3" NaI(Tl) , 2.3 Kev resolution and peak/Compton 1:56 at 1.33Kev gamma interaction of ^{60}Co. The detector is coupled with a multichannel analyzer (MCA). An inner shield consists of 10 cm thick lead (Pb) with a 2 mm copper sheet (Cu) to reduce background radiation. MAESTRO-32 was used as a software energy window program. The system was calibrated using a standard point source for preventing energy deviation between 46:3000 Kev. The measured activity of different identified radionuclides presented in the gamma spectra of the environmental samples was estimated according to the following expression:

$$A_E(Bq) = \left[\frac{1}{\varepsilon_E}\left(\frac{M_E}{T_E} - \frac{\bar{M}_{BG}}{T_{BG}}\right)\right] \Big/ P_E$$

Where A_E is the activity at specified radionuclide energy (E), $\varepsilon_E$is detector photo peak efficiency in counts per emitting gamma ray at specified gamma ray energy (E), M_E is the net photo peak at specified line energy E "counting area" for investigation samples, $\bar{M}_{BG}$ is the measured average background (BG) counting under the specified gamma line energy, P_E is the number of gamma photon emission per radionuclide disintegration, T_{ME} and T_{BG} are the counting time in second for samples and BG. The activities of the environmental samples were measured for ^{238}U, ^{235}U and ^{232}Th series in addition to ^{40}K non-series, which they were reported in table (2). In order to compare the specific activities of materials containing different amount of radium, thorium and also potassium an index noted as Ra equivalent concentration (Ra_{eq}) is evaluated according to (Beretka and Mathew; 1985, Shaban Harb *et al*;2012, Tufail *et al*., 2005, OECD;1979)

$$Ra_{(eq)} = F_1A_1+F_2A_2+F_3A3$$

Where A_1 , A_2 and A_3 are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in sequence in Bq/Kg. F_1, F_2 and F_3 are constant equal 1, 1.43 and 0.077 in sequence.

The external gamma dose rate at one meter above the surface for rock samples was estimated by using the following equation (Uosif;2007, Augustine Faanu *et al*;2010):

Dose rate (n Gy/h) = $F_K A_K + F_U A_U + F_{Th} A_{Th}$
Where F_K, F_U, F_{Th} are the dose conversion factors for ^{40}K, ^{238}U, ^{232}Th and equal 0.041, 0.462, 0.604 nSv/h/Bq Kg^{-1} in sequence. The annual effective dose ($eD_{(\gamma)}$) was estimated from the following equation:

$eD_{(\gamma)}$ (mSv/y)= $D_{(\gamma)}$ (n Gy/h)
$T_{(EXP)}$(Sv/Gy)$F_{(EXT)}$*(10^3msv/10^9 nGy)

Where $D_{(\gamma)}$ is the average gamma dose rate (mSv/y), $T_{(EXP)}$ is the exposure for hours in1 year equal 8.76* outdoor occupancy factor 0.2, $F_{(EXT)}$ is the effective dose to absorbed dose conversion factor of 0.7 Sv/Gy for environmental exposure to gamma rays, and 10^{-6} is conversion between nano and milli. (UNSCEAR;2000, Huy and Luyen; 2006, Hasan *et al*; 2010, El Aassy *et al*, 2012).

RESULTS AND INTERPRETATION

The results in table (2) show the activity concentrations of different decayed daughter nuclides for each parent series nuclide in Bq/Kg. The measured radionuclide was estimated for the gamma lines of intensity greater than the minimum detectable sensitivity. Figure (2) represents the average activity of different gamma photon energies corresponding to specified daughter nuclides in the parent nuclides, data of table(2) and fig(2) showed that:-

1. In case of ^{238}U; the highest arithmetic mean activity (^{238}U and ^{234m}Pa) photopeak are ranged between 533±17 and 14045±96 Bq/Kg. These results are much higher than of the permissible level which is 35 Bq/Kg (UNSCEAR, 2000). All distribution of studied samples was good secular equilibrium between ^{238}U and^{226}Ra except sample code H-5 which indicate migration out of ^{238}U under acidic media.
2. In case of ^{235}U; the arithmetic mean ranged between 24±2 and 647±1 Bq/Kg, this result has an agreement with the magnitude of ^{238}U which keeps NORM distribution ratio for $^{238}U/^{235}U$ this ratio ranged between 20.37 and 22.21 as shown in table (3).

Table 2 The specific activity (Bq/Kg) of radionuclides

Parent nuclide	Energy (keV)	A_E±σ						
		H.1	H.3	H.4	H.5	H.7	H.8	H.9
Uranium-series(^{238}U)								
^{234}Th	63.29	9301±48	2927±23	533±11	4079±31	6075±34	13987±50	8815±39
^{234m}Pa	1001	9484±100	2857±44	533±23	3675±96	6016±54	14045±96	8822±79
$\bar{A}_E$		9393	2892	533	3877	6046	14016	8819
^{234}U	53.2	7217±76	1585±42	375±18	2794±43	4484±52	13428±50	6389±91
^{234}U	120.9	5561±701	1862±178	394±144	2766±6	3457±508	12579±506	2480±445
$\bar{A}_E$		6389	1724	385	2780	3971	13002	4435
^{230}Th	67.7	8784±162	3164±57	611±30	13773±113	4882±99	13942±102	8822±96
^{226}Ra	186.1	9959±20	2575±8	594±4	11804±21	5934±13	13642±19	7751±14
^{214}Pb	295.1	6383±8	1428±3	421±2	7151±7	4160±6	8482±6	5660±6
^{214}Pb	352	6436±6	1471±2	425±1	7191±5	4197±4	8618±6	5697±4
^{214}Bi	609.3	6349±6	1428±3	418±2	7174±6	4140±5	8460±6	5624±5
^{210}Pb	46.5	8569±21	2220±9	541±4	6823±17	4781±13	11691±21	5697±14
Uranium-series (^{235}U)								
^{235}U	143.76	436±4	141±1	25±1	206±4	278±3	642±5	410±3
^{235}U	163.35	433±10	141±1	21±1	156±9	277±6	647±12	411±7
^{235}U	185.7	436±1	142±0.4	24±0.2	178±0.4	276±0.6	647±1	411±1
^{235}U	205.3	441±8	140±5	23±2	172±7	272±6	650±8	412±7
$\bar{A}_E$		436	142	24	178	276	647	411
Thorium-series (^{232}Th)								
^{228}Ac	338.4		21±1	40±2		38±2		
^{228}Ac	911.2	33±2	18±1	40±2	37±2	34±2	14±2	20±1
^{208}Tl	583		22±1	35±2		28±1		
^{208}Tl	2614.4	34±	20±1	38±2	35±1	36±1	14±0.4	22±1
$\bar{A}_E$		34	20	38	36	34	14	21
^{40}K	1460.8	213±6	130±3	249±3	231±6	234±5	84±5	120±4

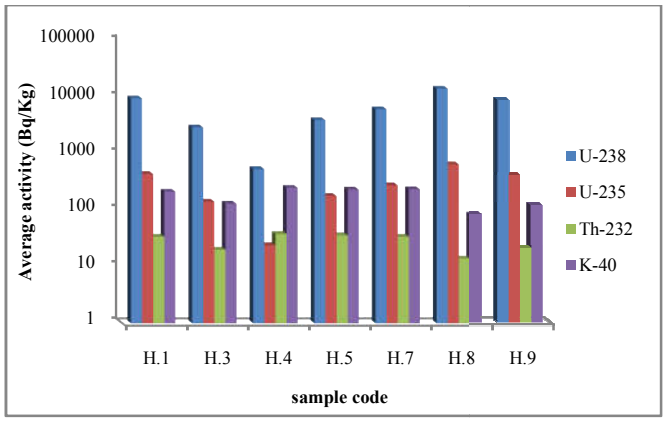

Figure 2 The average activity (Bq/Kg) for ^{238}U, ^{235}U, ^{232}Th and ^{40}K.

3. In case of ^{232}Th; the arithmetic mean activity ranged between14±1 and 38±2 Bq/Kg, which are lower than the global levels (45 Bq/Kg) reported by (UNSCEAR, 2008).
4. The activity concentration of ^{40}K ranged between 84±8 and 249±3, these values are lower than the global levels (412Bq/Kg) reported by (UNSCEAR, 2008; Abd El-mageed *et al*, 2011).

Table (3) represents the different ratios of radionuclides activity concentrations in (Bq/Kg) in natural series which illustrate the relation between parent/daughters radionuclides and these results reflect the type of equilibrium in different lithologies. The strongest correlation coefficient equals 0.9996 between the activity of ^{238}U and ^{235}U due to the two isotopes accompanied each other with a constant ratio in NORM (≈21.7).

Table 3 Environmental correlation between radionuclides through different activity ratios

Sample code	$^{238}U/^{235}U$	$^{234}U/^{235}U$	$^{234}U/^{238}U$	$^{226}Ra/^{238}U$	$^{230}Th/^{238}U$	$^{230}Th/^{234}U$	$^{226}Ra/^{230}Th$	$^{210}Pb/^{226}Ra$
H.1	21.54	14.65	0.68	1.06	0.94	1.37	1.13	0.86
H.3	20.37	12.14	0.60	0.89	1.09	1.84	0.81	0.86
H.4	22.21	16.04	0.72	1.11	1.15	1.59	0.97	0.91
H.5	21.78	15.62	0.72	3.04	3.55	4.95	0.86	0.58
H.7	21.90	14.39	0.66	0.98	0.81	1.23	1.22	0.81
H.8	21.66	20.75	0.96	0.97	0.99	1.04	0.98	0.86
H.9	21.04	10.79	0.50	0.88	1.00	1.99	0.88	0.74

The limit of the current calculation fluctuated between 20.37 and 22.21which are in normal distribution. The $^{234}U/^{235}U$ ratio ranges between 10.79 and 20.75 which is acceptable values with correlation coefficient equal 0.8374. All samples are less than unity for the $^{234}U/^{238}U$ ratio which reflects that the enrichment ^{238}U with depleted in ^{234}U, this result due to the process of α-recoil of ^{234}U. (Pekala *et al*; 2008, Pekala *et al*, 2010, Abdel Gawad and Ibrahim, 2015). The correlation coefficient equals 0.8448 between the activity of ^{238}U and ^{234}U as shown in fig (3).

^{226}Ra and ^{230}Th are attained to close secular equilibrium. It is clear from the $^{226}Ra/^{230}Th$ activity ratios are near to unity, ranging from 0.81 to 3.55. So that the $^{226}Ra/^{238}U$ and $^{230}Th/^{238}U$ are similar distribution for all samples except one sample H-5 equal 3.04, 3.55 for $^{226}Ra/^{238}U$ and $^{230}Th/^{238}U$ in sequence. It means that uranium is affected by physical/chemical properties of the sediments originates due to their mobility or/and migration where these ratios are sensitive to changes of geochemical process.(Krmar *et al*., 2013, Qidong *et al*., 2016, El-Reefy *et al*.,2014).

The ratio of $^{230}Th/^{234}U$ activity concentration ranges between 1.04 and 1.99. It is greater than unity but it is near to equilibrium except samples H-5 as it recorded 4.95, which they are highest than unity. It is the best evidence for disequilibrium due to preferential mobilization of uranium, the weak correlation coefficient equals 0.4722 between them as shown in fig (3). The ratio of $^{210}Pb/^{226}Ra$ fluctuated between 0.58 and 0.91 which is lower than unity. This indicates that they are out of equilibrium.

In a plot of $^{234}U/^{238}U$ versus $^{230}Th/^{238}U$ as shown in fig (4), the pathways of return to equilibrium for solid phases are shown for two cases: accumulation of U ($^{230}Th/^{238}U$ decrease) and leaching of U ($^{230}Th/^{238}U$ increase) (Thiel *et al*., 1983; Brantley *et al*., 2008). The presence of data points in the forbidden zones may be explained as a result of continuous and contrasting U mobilization processes (Chabaux *et al*., 2003). According to judging standards, activity ratios between 0.90 and 1.10 are referred as secular equilibrium within the conservative (10 %) analytical error for the samples (Min *et al*., 2005).

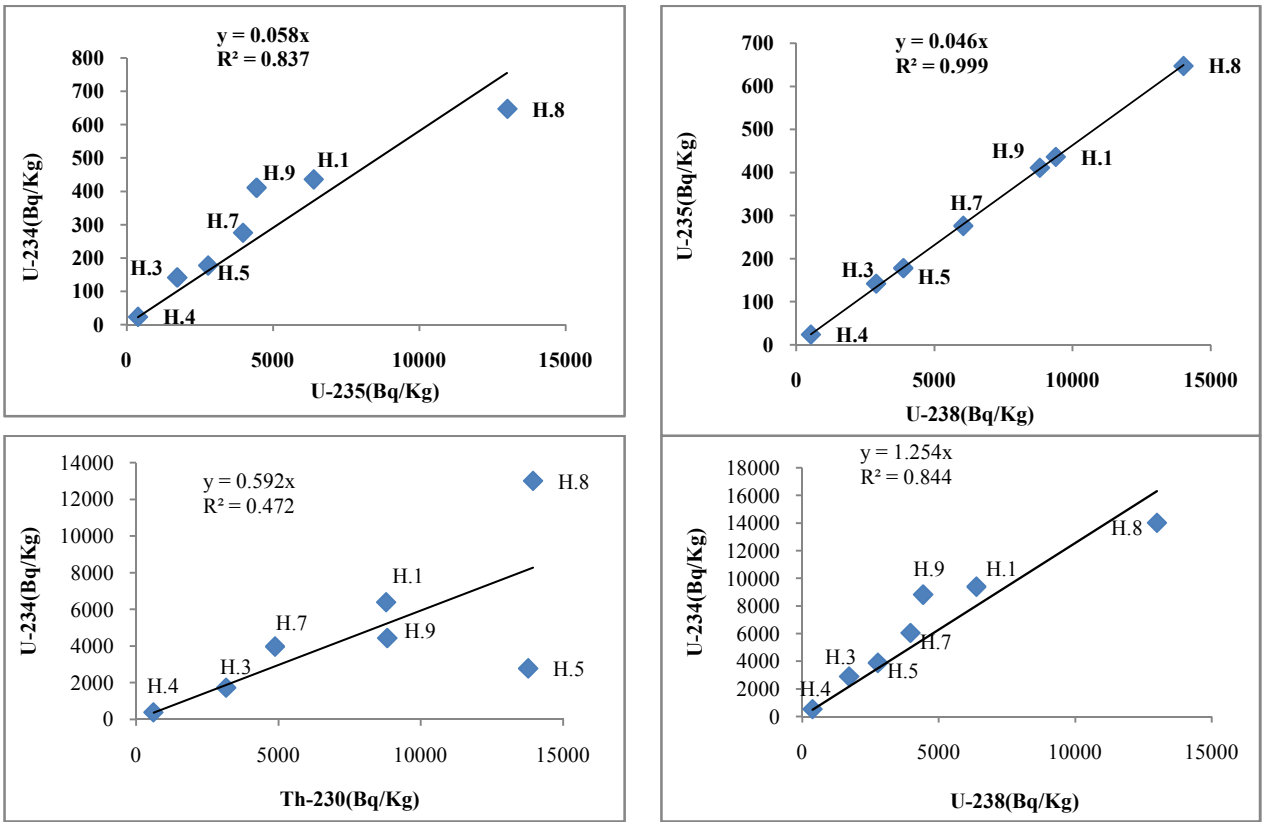

Figure 3 The relations between different radionuclides

It is supported by the Thiel diagram (Fig. 4), in which the samples that plot into the boxed-in area are considered near or at secular radioactive equilibrium. All cases of U leaching are characterized by ($^{234}U/^{238}U$) AR < 1 and all cases of U accumulation are distinguished by the relation ($^{234}U/^{238}U$) AR > 1 (Thiel *et al*., 1983).

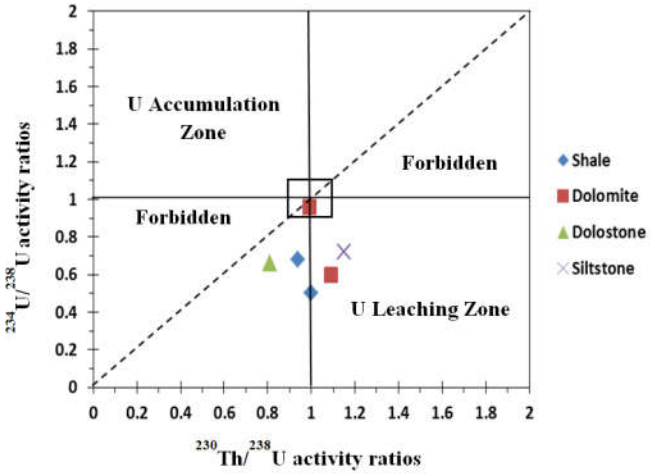

Figure 4 Thiel diagram showing the evolution of $^{234}U/^{238}U$ versus $^{230}Th/^{238}U$ activity ratios.

The Thiel diagram in Fig (4), points to the fact that one sample (H8) lies on the verge of the secular equilibrium condition. All remaining samples lie on the verge of the uranium leaching region for an initial uranium accumulation phase may have been followed by relatively recent uranium leaching.

fixed in sediments with continuous recharge (El Galy *et al*., 2008) except one sample (H.5).

Table 4 The equivalent concentration (ppm) for radioelements.

Sample code	U (ppm)	Th (ppm)	K (%)	Th/U	K/Th
H.1	757	8	0.68	0.01	0.09
H.3	233	5	0.42	0.02	0.08
H.4	43	9	0.8	0.21	0.09
H.5	313	9	0.74	0.03	0.08
H.7	488	8	0.75	0.02	0.09
H.8	1130	3	0.27	0.0027	0.09
H.9	711	5	0.38	0.01	0.08

According to report of (OECD, 1979; UNSCEAR, 2000&2008) radium equivalent activity should be less than 370Bq/Kg for healthy use. The current results (as it shown in table 5) show that Ra_{eq} varies between 667.44 and 13371, it is equivalent two times to five times exceed than healthy use.

The external gamma dose rate at one meter above the surface for rocky samples range between 279.41 and 6466.96 nGy/h as it is shown in table (5).

These results are higher than the public average 57 nGy/h. the magnitude of annual effective dose varies from 0.34 to 7.93mSv/y, it seems that all rocks are higher than unity except one sample (H-4) which recorded 0.34 mSv/y. In general all samples of the current study are more than the permissible limits and they are considered to be used with cautions.

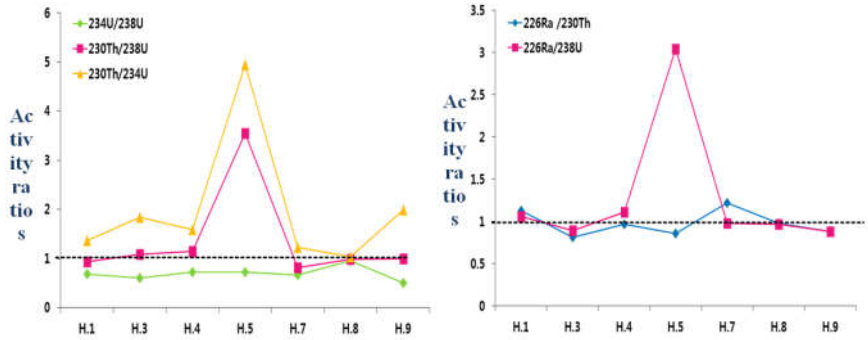

Figure 5 Diagram showing the activity ratios of radionuclides versus samples code.

Fig (5) Diagram showing the activity ratios of radionuclides versus samples code. It is clear that H.5 sample recorded high values.

Table (4) shows that conversion from the activity concentration (Bq/Kg) and the equivalent concentration by (ppm), 1ppm for U= 12.4 Bq/Kg (Chiozzi *et al*, 2002), 1ppm for Th= 4.04 Bq/Kg and 1% for K= 313Bq/Kg.

According to the results of table (4), the average concentration of uranium for sedimentary rocks is high in content and low in content of thorium and percentage potassium comparable with U. All of these rocks are classified to middle –member of Um Bogma Formation as pervious illustrated. Th/U (ppm) ratio indicates depletion or enrichment of radioisotopes, which it ranges from 0.01 to 0.21 except one sample (H-8) is 0.0027. These results are very low from natural value which indicates relative U enrichment as U was migrated from its source and

Table 5 Radium equivalent, the dose rate and annual effective dose

Sample code	Ra(eq) (Bq/Kg)	Dose (n Gy/h)	annual eD (mSv/y)
H.1	10023.96	4368.84	5.36
H.3	2613.57	1353.51	1.66
H.4	667.44	279.41	0.34
H.5	11873.20	1822.39	2.23
H.7	6000.57	2831.70	3.47
H.8	13371.46	6466.96	7.93
H.9	7790.23	4091.98	5.02

CONCLUSION

HPGe detector was used to study gamma rays (non-destructive method) in the current study. The results were obtained to discuss the variation of environmental formation on the study area (Um Bogma). It is happened due to geochemical history in addition to alteration and weathering processes. It is clear that enrichment or depletion operations through distribution and

redistribution of radionuclides have been carried out. The type of equilibrium, mobilization and migration of U and Th was illustrated through the route of this study. The average activity concentrations of ^{238}U, ^{235}U, ^{232}Th, ^{226}Ra and ^{40}K for sedimentary rocks under investigation are more exceed the magnitude of safe criteria and exposure risks which are recommended in public papers. The results of the current study encouraged the researchers to study these samples by other techniques like geochemical and theoretical techniques to assist in more interpretations about the study area in future papers.

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