Research Article

THE MEASUREMENT'S OF NATURAL RADIOACTIVITY AND VARIATION IN THE URANIUM ISOTOPIC RATIOS IN SEDIMEN SAMPLES

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ARTICLE INFO	ABSTRACT
	Five samples of sediments collected from southwestern Sinai - Egypt, have been investigated. The activity concentrations of radionuclides in sediment samples were determined by γ-ray spectrometer using HPGe detector with a specially designed shield. The values of activity concentrations of uranium are high in all sediment samples, and the activity concentrations of thorium are high in all samples except one sample, where the activity concentrations of ^{40}K are low in most samples. The activity ratios between $^{226}Ra/^{238}U$ for sediment were calculated. Thorium to Uranium concentration ratios (Clark value) was also estimated. The uranium isotopes ^{234}U, ^{235}U and ^{238}U are found naturally in the environment. ^{235}U and ^{238}U are present nuclides of two independent decay series of isotopes, while ^{234}U is member of the ^{238}U decay series. When decay series occur in closed system the series tends to reach, with time, the state of secular equilibrium in which the activities of all series member are equal to the activity of its first nuclide. The $^{234}U/^{238}U$ activity ratio obtained indicated that these two uranium isotopes are in the state of secular radioactive equilibrium.

INTRODUCTION

The natural uranium consists of three isotopes; ^{238}U, ^{235}U and ^{234}U with abundances of approximately 99.27, 0.72 and 0.054 %, respectively. These isotopes comprise the natural uranium and have the same geochemical behavior. ^{238}U and ^{235}U were parent radionuclides of two natural occurring radioactive series; ^{238}U and ^{235}U finally decays to ^{206}Pb and ^{207}Pb through 14 and 11 radionuclides, respectively.[1]

Assessment of radionuclides in solid and rocks in many parts of the world has been increasing in the past two decades because of their hazard on the health of the population[2].

The understanding and awareness about radiation in outdoor and indoor environment is important for determining the population exposure to radiation.

The outdoor exposure is from the geological formation, sand, soil etc. to which human beings are exposed. This is necessary because radioactivity of the soil environment is one of the main sources of exposure to humans while the decay products of ^{40}K, ^{238}U and ^{232}Th series represent the main external source of radiation to human body. According to investigation these radionuclide become part of the soil in the following ways: 1) As part of earth's original crust (i.e primordial radionuclides),

2) produced and deposited by cosmic ray interaction (i.e Cosmogenic radionuclide) and 3) through man-made release (i.e man-made radionuclide). [3].

In this study, the natural radioactivity concentration of ^{40}K, $^{238}U(^{226}Ra),^{234}U$, ^{235}U and ^{232}Th in some samples collected in southwestern Sinai, Egypt.

($^{234}U/^{238}U$ and $^{230}Th/^{234}U$) activity ratio (AR)

The uranium and thorium contents of a black shale can be affected by a number of syndepositional and postdepositional processes not readily manifested by elemental concentrations. An integrated isotopic approach, i.e. a study of the $^{234}U/^{238}U$ and $^{230}Th/^{234}U$ AR in each fraction of the radionuclides belonging to the same radioactive series, is necessary to yield information about the differential behaviour of these radionuclides in the environment. Under typical environmental conditions, the order of mobility has been reported as being U > Ra > Th [4].

Effects of (α −Recoil)

The α -recoil process displaces the daughter nuclei in the U decay-series by ~ 200 to 350 Å In addition, it triggers off a collision cascade that creates a small structurally damaged volume. The daughter isotope, therefore, may be prone to

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preferential leaching, provided low-temperature processes did not anneal its travel path. Recoiled daughter isotopes of near surface U (incorporated in the crystal lattice or adsorbed on the crystal surface), [234]Th and [230]Th, may be ejected from the crystal into the open fracture and thereby become lost. Such a sample would acquire a deficit in [234]U and [230]Th. Loss through α-recoil alone results in [[234]U]/[[238]U] and [[230]Th]/[[234]U] values < 1. Direct loss of U-daughters by α -recoil applies only to a less than 0.03 µ m wide veneer of the crystal, and thus, is not very efficient to generate disequilibrium. Leaching of the recoiled daughter isotopes from their disturbed location in the crystal or along recoil tracks may enhance the overall loss, but does not enlarge significantly the volume from which the loss occurs. Since the solubility of Th in aqueous fluids is much lower than that of U, such kind of leaching would result in lower [[234]U]/[[238]U] and enhanced [[230]Th]/[[234]U] ratios in the mineral. On the other hand, recoil implantation of U daughters through mineral surfaces into inclusions, fractures, voids, and channels eventually results in excess [234]U and [230]Th. Even here, only the outermost veneer of the crystal may experience a gain of U daughters. Thus, the critical parameter, which controls (i) whether an analytically resolvable excess of U daughters is obtained or not, and (ii) which magnitude such an excess may reach, is the surface-to volume ratio. The higher this ratio, the higher the excess may become.
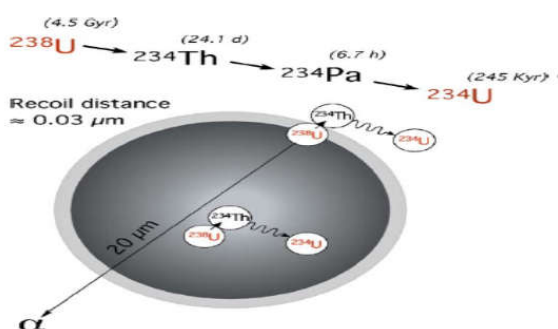

Fig 1 Schematic diagram of recoil ejection of [234]Th from a spherical grain as a result of the alpha decay of [238]U, followed by beta decay of [234]Th to [234]U [5].

Geologic setting

The study area fig. (2), is located in southwestern Sinai, between longitudes 33_20^0 and 33_25^0 E and latitudes 29_06^0 and 29_12^0 N.
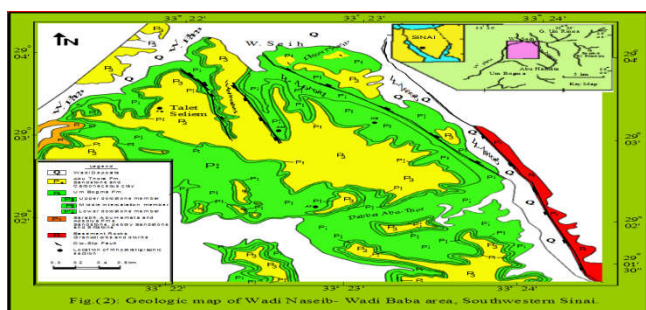

Fig 2 Geologic map of southwestern Sinai, Egypt [6-7].

MATERIALS AND METHOD

Sampling and samples preparation

For radioactivity measurements, a total of five samples each of mass 1kg. The samples were ground, homogenized and sieved to about 200 mesh. The samples were first weighted and placed in polyethylene bottles of 250 cm^3 volume. The bottles were completely sealed for more than one month to allow radioactive equilibrium to be reached. This step is necessary to ensure that radon gas is confined within the volume and that the daughters will also remain in the sample.

Experiment set up

High purity vertical germanium was coupled to a PC-computer with a special electronic card to make it equivalent to a multichannel analyzer. The system also contains the usual electronic components of preamplifier, amplifier and power supply. The detector has resolution (FWHM) of 1.85 keV for the 1332.5 keV γ-ray line of [60]Co. The γ-ray spectrometer energy calibration was performed using ^{60}Co, [226]Ra and [241]Am point sources. The detector was surrounded by a special heavy lead shield of 10cm thickness with inside dimension 28 cm diameter ×40.5cm height. The absolute detection efficiency of the HPGe detector was determined 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 [8-9]. 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 [8]. The uranium standard (RGU-1) is U-ore diluted with silica with 4940 Bq kg[-1] of [238]U, 228 Bq kg[-1] of [235]U, a negligible amount of [40]K (less than 0.63 Bq kg[-1]) and some traces of [232]Th (less than 4 Bq kg[-1]). The thorium standard (RGTh-1) is Th-ore diluted with silica having 3250 Bq kg[-1] of [232]Th, but containing some[238]U (78 Bq kg[-1]) and [40]K (6.3 Bq kg[-1]). The potassium calibration standard (RGK-1) is produced from high purity (99.8%) potassium sulphate with 14000 Bq kg[-1] of potassium with uranium and thorium contents lower than 0.001 and 0.01 ppm (parts per million), respectively [9].

The γ-ray transitions used to measure the concentration of the assigned nuclides in the series are follows:[238]U was determined from the gamma rays emitted by its daughter products[10][234]Th and [234m]Pa activities determined from the 63.3 and 1001 keV photo peaks, respectively, [214]Bi(609.3, 1120.3, 1238.1, 1377.7, and 1764.5 keV), [214]Pb (295.1 and 352.0 keV). To measure [230]Th activity,γ-ray emission at 67.7 keV was used. 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, 911.2 and 968.9 keV from [228]Ac and 583 keV from [208]Tl, and was measured using 1460.8 keV for potassium.

In order to determine the background contribution due to naturally occurring radionuclides in the environment around the detector, an empty polyethylene beaker of the same 250 cm^3 volume was counted with the same geometrical conditions as the sample. The measurement time for both activity and background measurement was 70.000 seconds. The background spectra were used to correct the net- gamma- ray peak areas for

the studied isotopes. The concentrations of radionuclides are calculated using the following equation:

The radioactivity concentration of the different identified radionuclides was calculated by γ-ray spectrometry with the following simple regression **[10]**:

$$ A = \frac{Net \cdot area \cdot (cps)}{I_\gamma \cdot \xi \cdot M} $$

Where:
A : Activity concentration of the gamma spectral line in Bq/kg
Net area (cps): The net detected counts per second corresponding to the energy Iγ   : Intensity of the gamma spectral
ξ: Counting system efficiency of the energy
 M: Mass of sample in kg.

RESULTS AND DISCUSSION

The activity concentration of ^{226}Ra ^{238}U, ^{234}U,^{235}U, ^{230}Th, ^{232}Thand ^{40}K (in Bq/kg) in the samples under investigation are listed in table (1).
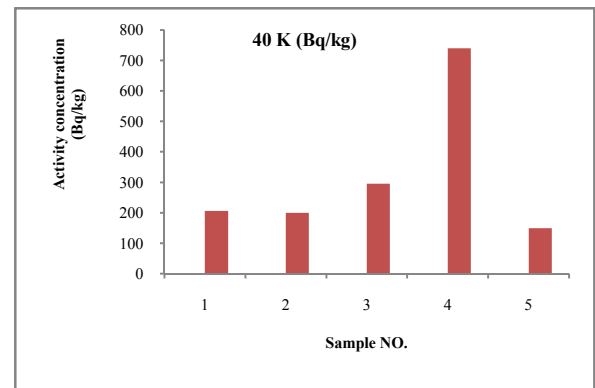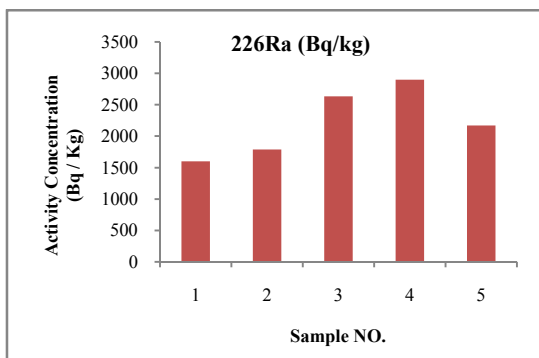
Table 1 The activity concentration of ^{226}Ra, ^{238}U, ^{234}U,^{235}U, ^{230}Th, ^{232}Thand^{40}K in Bq/kg of the sediment samples.

Samples	^{226}Ra	^{238}U	^{234}U	^{235}U	^{230}Th	^{232}Th	^{40}K
1	1599.92*	1419.64 *	1377.04*	65.48*	1436.67*	31.14*	205.9
2	1788.34	1786.55	1525.17	81.07	1615.04	61.42**	199.5
3	2630.43	2643	2716.87	128.15	2651.58	59.72	295.63
4	2892.73**	2864.09 **	2872.68**	135.0**	2815.40**	54.36	740.51**
5	2164.78	1920.83	1949.64	90.93	1943.88	47.17	149.35*

*The lower value
 **The higher value

From table (1) it is clear that the activity concentrations of the studied sediment samples are higher than the permissible level for uranium and radium (33 and 32 Bq/kg). The activity concentrations in all samples are higher than the permissible level (45 Bq/kg) for thorium except sample No.(1), and The activity concentrations in all samples are lower than the permissible level (412 Bq/kg) for potassium except sample No.(4) as fig.(3) [11].
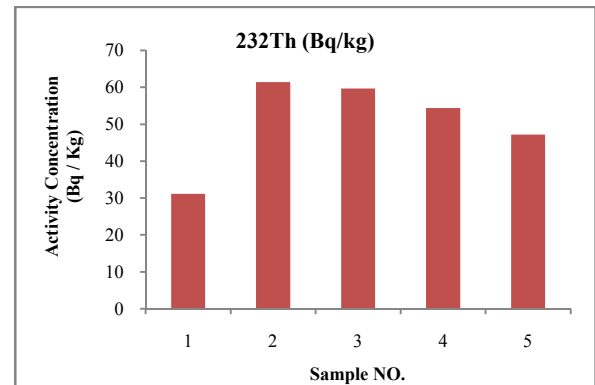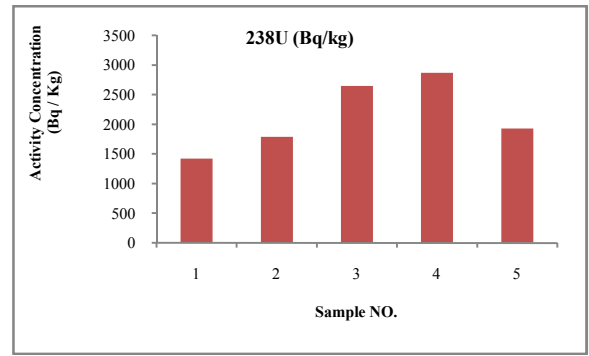
Fig 3 The activity concentrations for ^{226}Ra, ^{238}U,^{232}Th and ^{40}Kin the sediment samples

Table 2 Activity concentration of ^{238}U and ^{232}Th in (ppm) and Clark value of ^{232}Th/ ^{238}U

Samples	^{238}U(ppm)	^{232}Th(ppm)	^{232}Th/^{238}U
1	114.48	7.67	0.067
2	144.07	15.13	0.105
3	213.19	14.71	0.069
4	230.97	13.39	0.058
5	154.91	11.62	0.075

Table (2), give The radionuclides concentration of ^{238}U and ^{232}Th, for the samples which the concentrations are varying between (114.84 and 230.97 ppm) and (7.67 and 15.13 ppm) respectively. The ratios of ^{232}Th/ ^{238}U are less than Clark's value (3.5) in all samples, which indicates that these locations are enriched in uranium.

Table (3) shows the activity ratios for sediment samples. It is seen that: The ^{238}U/^{235}U activity ratio for all samples range between 20.63 and 22.037 which reflect little deviation from the normal ratio (21.7) and the alteration process affects this ratio as shown in fig.(4).
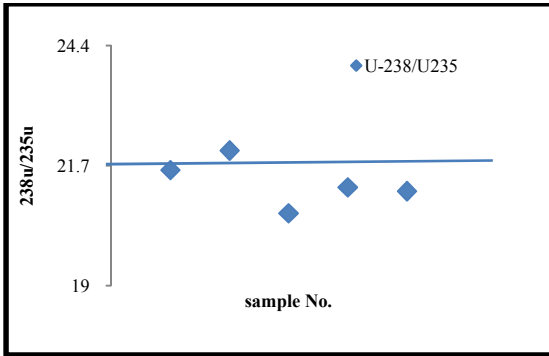
Fig 4 Variations of the 238U/235U activity ratio in samples, the certified value is (21.7)

Table 3 Activity ratios of sediment samples

Samples	$^{238}U/^{235}U$	$^{234}U/^{238}U$	$^{230}Th/^{238}U$	$^{230}Th/^{234}U$	$^{226}Ra/^{238}U$	$^{234}U/^{235}U$	$^{226}Ra/^{230}Th$
1	21.60	0.96	1.012	1.043	1.127	21.03	1.114
2	22.037	0.854	0.904	1.059	1.001	18.81	1.107
3	20.63	1.028	1.003	0.976	0.995	21.201	1.992
4	21.215	1.003	0.983	0.980	1.01	21.279	1.027
5	21.124	1.015	1.012	0.997	1.127	21.441	1.114
Avg.	21.321	0.971	0.982	1.011	1.052	20.752	1.070

Uranium concentrations in the samples were observed in the $^{234}U/^{238}U$ isotopic activity ratio to be varied from 0.854 to 1.028. The sediment samples in the study area exhibited secular equilibrium between ^{234}U and ^{238}U in all samples, $^{230}Th/^{238}U$ and $^{230}Th/^{234}U$ ratios for all samples are close to unity, as in fig. (5).
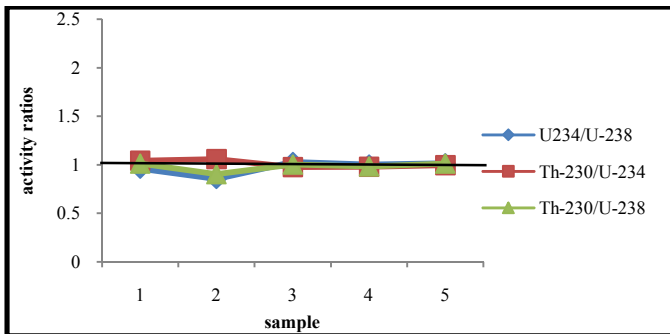

Fig 5 Variations of the $^{234}U/^{238}U$, $^{230}Th/^{234}U$ and $^{230}Th/^{238}U$ activity ratios in samples, the certified value in equilibrium is (unity)

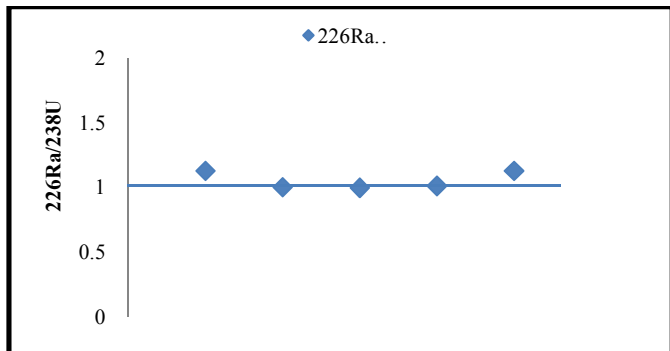The range values of $^{226}Ra/^{238}U$ ratio are between 0.995 and 1.127are near unity, as in fig. (6).

Fig 6 Variations of the $^{226}Ra/^{238}U$ Activity ratio in samples

In fig. (7) the value of ratio $^{234}U/^{235}U$ is varied between 18.81 and 21.441 which mean uranium leaching out from sample due to alteration processes.
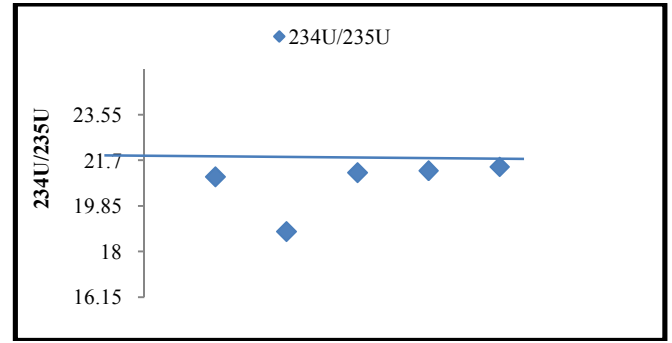

Fig 7 Variations of $^{234}U/^{235}U$ activity ratio in granite samples. The certified value of $^{234}U/^{235}U$ Activity ratio is 21.7

In a plot of $^{234}U/^{238}U$ versus $^{230}Th/^{238}U$ fig.(8), the pathways of return to equilibrium f0r dashed phases are sh0wn for two cases: accumulation of U ($^{230}Th/^{238}U$ decrease) and leaching of U ($^{230}Th/^{238}U$ increase) [12, 13]. The presence of data p0ints in the forbidden z0nes may be explained as a result of continu0us and contrasting U mobilization pr0cesses [14]. Depending 0n the relative rates of U gains and l0sses and on the intensity 0f the U fracti0nation, it is p0ssible to explain the data p0ints present in the forbidden z0ne [15]. Acc0rding to judging standards, activity ratios between 0.90 and 1.10 are referred as secular equilibrium within the c0nservative (10 %) analytical error f0r the samples [16]

For samples, the Thiel diagram fig.(8) in which almost all samples that are pl0tted into the b0xed-in area are c0nsidered near secular radioactive equilibrium (at limit 0.90 to 1.10) for $^{234}U/^{238}U$ and $^{230}Th/^{238}U$. The sample (2) fall to the U-leaching region in the Thiel diagram.
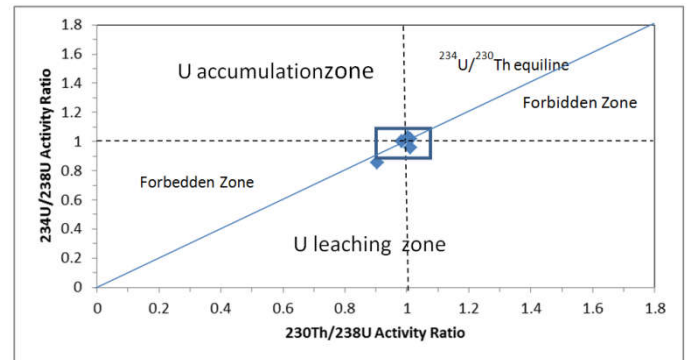

Fig 8 Schematic diagram showing the evolution of $^{234}U/^{238}U$ versus $^{230}Th/^{238}U$ activity ratios for samples

CONCLUSION

The areas under investigation, Sinai with high uranium contents and low thorium relative to uranium, the values of potassium in all samples are low except one sample. All samples are less than Clark's value of $^{232}Th/^{238}U$ which indicates that these areas are enriched in uranium. The mean isotopic activity ratio $^{238}U/^{235}U$ was found to be around 21.7 in all samples, the $^{234}U/^{238}U$ isotopic activity ratio varied from 0.854 to 1.028 in samples under investigated exhibited secular equilibrium between ^{234}U and ^{238}U. In a plot of $^{234}U/^{238}U$ versus $^{230}Th/^{238}U$(the Thiel diagram) , all samples that are plotted into the boxed-in area are considered near secular radioactive equilibrium (at limit 0.90 to 1.10) except one sample fall in the U-leaching region.

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