

Radioactivity Levels in Some Sediments and Water Samples from Qarun Lake by Low–Level Gamma Spectrometry

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Abstract: *The specific activities of the natural radionuclides ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K were measured in sediments and water samples collected from the Qarun lake (Middle Egypt) in order to gather information about radionuclides mobility and distribution. In the sediment and water in general, the concentration of ^{238}U was found to be higher than that of the ^{232}Th and the activities of ^{232}Th , ^{226}Ra and ^{238}U in this lake falls within the worldwide ranges while ^{40}K was observed in sediment samples, ranging from 410 to 1426 Bq kg⁻¹ be higher than the worldwide average. The results have been compared with other radioactivity measurements in different countries. Multivariate Statistical analyses were carried out between the parameters obtained from the radioactivity to know the existing relations and to study the spatial distribution of radionuclides.*

Keywords: Qarun lake; sediment; water; uranium; radium; thorium; potassium; γ -ray spectrometer

1. Introduction

Radionuclides have been released to the environment from different sources and processes. Radionuclides present in the biosphere, whether natural or artificial in origin, ultimately result in irradiation of human populations (Aheir 1995). Naturally occurring radionuclides of terrestrial origin are present in rivers and lakes sediments as well (Krmr 2009). A number of scientific efforts have been undertaken to characterize radionuclides in the marine environment, including biota, water and sediments (Dar and El Saharty 2013; Ibrahim and Ramzy 2013; El-Reefy et al. 2010; El Zakla et al. 2013; Darwish 2013). Such studies can be useful to estimate the degree of human risk associated with the ingestion of radionuclides in biota through the food chain and to establish a baseline database of radionuclides concentration, in order to monitor the possible variations in the marine environmental radioactivity due to nuclear industry and other human activities (Akram 2005 and Burger 2006). Sediment plays a role in accumulating and transporting contaminants within the geographic area and is considered the environmental host of the waste discharged by natural or artificial processes in our world. Lakes act therefore as sinks for the materials which pass through the various aquatic chemical and biological cycles including radionuclide contaminants. They were considered as sources of radionuclides to the downstream ecosystems.

Lake Qarun, is the only enclosed saline, the third largest lake in Egypt and it is the only natural contemporary lake of any size in Middle Egypt. It is therefore rich in both natural and archaeological resources. Although Lake Qarun designated as protected area back 1989, the lake has hardly been protected from various polluting elements. It suffers from a serious pollution problem due to uncontrolled solid and liquid domestic and industrial waste disposal practices. Therefore, the object of this study is to determine the activity concentrations of gamma-emitting radionuclides: ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K

in Qarun lake sediments and water, their distributions and the possible anthropogenic sources.

Geographical Aspects

The Lake is in the Fayoum Province on the fringe of the Western Desert about 80 km south of Cairo and located between longitudes of 30° 24' & 30° 49' E and latitude of 29° 24' & 29° 33' N in the lowest part of El-Fayoum depression (Fig. 1). It has an irregular shape of about 40 km in length, 5.7 km in width and 34 to 43 m below sea level with a mean depth of 4.2 m. Groundwater appears to be continuously seeping from a number of sub-surface springs at the lake bottom. A gently sloping sand-plain extends from the Lakeshore northwards and upwards to reach sea level at 7 km north of the shoreline. The bottom of the lake is muddy loam at the east, as the drainage water carries huge quantities of clay, which is discharged at the east of the lake, whereas the west is mostly sandy (Abdel-Regal 1995). The lake is an important archeological site because of the presence of marine, fluvial and continental environment, all in one area with a unique collection of fossil fauna and flora that goes back to some 40 million years. The lake's main sources of water are from agriculture drainage and domestic wastewater. Most of the drainage water reaches the lake through two main drains, Al-Batts and Al-Wady drains and two pump stations (Main pump station and Khor Alhitan pump station). Since, 1973 Al-Wady drain partially delivers most of its water from Wadi Al-Rayan Lakes to maintain an established water level of Lake Qarun.

2. Sampling

Sediment and water samples were collected from 17 locations distributed throughout the studied area of Qarun Lake. Water and sediment samples were analyzed for a pre-defined set of radiological indicators which can be selected on the basis of the results reported in the previous surveys to allow the buildup of a meaningful database that can be used for comparative assessment and trend delineation. Sediment samples were collected using grab sampler and the depth of the

cores ranged from 10 to 20 cm. The average distance between the sampling sites was about 1.5 km. Each sample was about 1 kg in wet weight. The collected samples were dried at 110 °C to remove the moisture content until a constant weight was obtained, grounded into powder using mortar and pestle and sieved with a 2 mm mesh to obtain homogenized samples. The net weight of each sample was 350 gm and packing in containers of specific geometry similar to that of the calibration source for gamma activity analysis. Water samples were collected by filling pre-cleaned bottles with water in a depth of 0.25 m. All samples were acidified using concentrated HNO₃, to prevent any loss of radium-isotope around the container walls, and to avoid the growth of micro-organisms.

After packing, all samples were stored for a month to achieve secular equilibrium between radium and thorium and their progeny.

2.1 Gamma Spectrometry

In the present work, the activity levels of ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K of the collected samples were measured using gamma-ray spectrometry based on a highly pure germanium coaxial detector (HPGe) at the National Institute for Standards, Egypt. This detector has a relative efficiency of about 30% with a resolution energy of 1.9 keV FWHM for the 1332 keV gamma transition of ⁶⁰Co. The estimated data were corrected using background counts based on the measurement spectrum analysis program. The gamma-ray spectrometer was calibrated for energy using 138G Marinelli beakers, source no. 566-69-7 manufactured by Isotope Products Laboratories, containing mixed sources of ¹⁰⁹Cd, ⁵⁷Co, ^{123m}Tc, ¹¹³Sn, ¹³⁷Cs, ⁸⁸Y, ⁶⁰Co, thus allowing some peaks to occur in the measurement range. The activity concentrations of the radionuclides in the samples were determined from their gamma-ray lines emitted from their decay products. The ²³⁸U activity concentration in the examined samples was derived from the weighted mean of the activities of the photo peaks of its daughter product ²³⁴Th (63.3 keV). The gamma-ray lines 295.2 and 351.9 keV for ²¹⁴Pb and 609.3, 1120.3 and 1760.5 keV for ²¹⁴Bi were used to determine the activity concentration of ²²⁶Ra by taking their weighted mean activity concentrations. The gamma-ray lines 338.3 and 911.6 keV for ²²⁸Ac, and 583.0 keV for ²⁰⁸Tl were used to determine the activity concentration of ²³²Th (assuming secular equilibrium between ²²⁸Th and ²³²Th). ⁴⁰K was investigated by its gamma-line 1460.8 keV. The samples were counted for at least 36 000 s to reduce the statistical counting error, and the background radiation was also measured using an empty Marinelli beaker under the same measurement conditions and subtracted from the sample's spectra.

3. Results and Discussion

Sediments

The activity concentrations of ²³⁸U, ²²⁶Ra, ²³²Th, and ⁴⁰K in the sediments of Qarun lake were presented in Fig. 2. The activity level of ²³⁸U in sediments varied between

11.4 and 43.5 Bq kg⁻¹, with an average of 23.5 ± 9.7 Bq kg⁻¹; ²²⁶Ra from 6.2 to 22.4 Bq kg⁻¹, with an average of 14.1 ± 6.2 Bq kg⁻¹; ²³²Th ranged from 5.2 to 26.6 Bq kg⁻¹, averaging 15.8 ± 6.8 Bq kg⁻¹, while the level of ⁴⁰K varied from 410 to 1426 Bq kg⁻¹, and the average was 933 ± 384 Bq kg⁻¹.

Summary statistics, the ranges and means of the activity concentration for all soil samples are presented in Table 1. All of the reported activity concentrations of sediment samples are given in becquerels per kilogram dry weight and secular equilibrium in both the ²³⁸U and ²³²Th series was assumed. The associated standard deviations are also given.

The recorded average activities of ²³⁸U, ²²⁶Ra and ²³²Th at the Qarun lake sediments were lower than the global average reported by UNSCEAR (2000) of the natural radionuclide contents in soil (33, 32 and 45 Bq kg⁻¹ for ²³⁸U, ²²⁶Ra and ²³²Th, respectively), while ⁴⁰K average activity was higher than the global average (420 Bq kg⁻¹).

The ²³⁸U and ²³²Th activities are not different from the typical values taken by El-Zakla et al., 2013 while ⁴⁰K activity measured by El Zakla was lower than the present study. Darwish et al. (2013) found relatively high concentrations of ⁴⁰K, nearly equal of ²³²Th concentration throughout the studied area of the lake and slightly lower concentration of ²³⁸U than the present study. Other comparisons with some different locations in Egypt and the world are listed in Table 2.

Generally, the activities in the Qarun Lake agree with those measured in Burullus Lake and Suez Canal despite the different natures of the three locations. The present concentrations of ²²⁶Ra and ²³²Th are among the lowest in activity of all the locations listed in Table 2. While the highest ranges of ²²⁶Ra activity concentration are those measured in Saudi Arabia (Khater 2013) and in Keum river in Korea (Lee et al. 2009), the highest range of ²³²Th activity concentration was reported from Coastal sediments in India (Ravisankar et al. 2014), the highest range of ²³⁸U activity concentration was reported from Gulf of Aqaba in Jordan (Ababneh et al. 2010). On the other hand, the ranges of ⁴⁰K activities in all these locations are very comparable.

The activity trends of ²³⁸U, ²²⁶Ra and ²³²Th indicate agriculture drainage and industrial wastes as the main sources of accumulations; ⁴⁰K is present to various degrees in different plant and animal tissues, including humans (UNSCEAR 2000), elevated levels of ⁴⁰K are expected in domestic effluent discharge points like wastewater treatment plants. However, ⁴⁰K may increase due to the use of fertilizers in agriculture land around this area and from domestic wastewater (Darwish et al., 2013). These patterns indicated that the measured radionuclides were mainly accumulated from agriculture and industrial wastes in the fine-grained sediments that are rich in organic matter content (Dar and El Saharty, 2013).

Skewness and kurtosis can be used to assess whether the assumption of normality is acceptable or not. The normality of a distribution is reflected in the low (close to zero) kurtosis and skewness coefficients. The values of skewness and kurtosis for all radionuclides are near to 0 and negative, respectively; therefore these radionuclides follow normal distribution (Table 1).

Water

Fig. 3 shows the activity concentrations of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K in water samples, while the descriptive statistics are summarized in Table 1.

The mean value of ^{238}U concentration was $6.4 \pm 0.6 \text{ Bq l}^{-1}$ and ranged between 5.4 and 7.49 Bq l^{-1} ; ^{226}Ra varied from 4.5 to 11.8 Bq l^{-1} with mean value $7.9 \pm 2.3 \text{ Bq l}^{-1}$; the average values of ^{232}Th is $3.2 \pm 0.43 \text{ Bq l}^{-1}$ which varied between 2.8 and 4.02 Bq l^{-1} while the values of ^{40}K ranged 22.8- 39.9 Bq l^{-1} with mean value of $31.3 \pm 5.7 \text{ Bq l}^{-1}$.

The calculated values of kurtosis and skewness of the radionuclides in water are summarized in table 1. Radionuclides ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K had also positive skewness coefficients and negative kurtosis coefficients indicating this distribution is very close to normality.

In a marine environment, radioactive materials can be attached to particulate matter in waters. Some isotopes remain dissolved and are termed conservative within water. Others are scavenged out of solution onto particulate material by biological or chemical processes, e.g., adsorption and co-precipitation. They may become deposited in sediments on the bottom of the lake. Uranium and thorium radionuclides have different behavior in the marine environment. While uranium remains dissolved in water, thorium is a particularly insoluble element in natural waters and it is usually found associated with solid matter.

The distribution of radionuclides between sediment and water can be determined using the sediment partition coefficients (K_d) (El-Reefy et al. 2010). The bottom sediment-water distribution coefficient (K_d) is widely used to describe the partitioning of radionuclides between aqueous and solid phases. In this study, the mean value of sediment partition coefficient K_d of ^{238}U , ^{226}Ra , ^{232}Th and ^{40}K were found to be 3.6, 1.84, 4.9 and 30.5 L kg^{-1} respectively. The sediment partition coefficient for ^{40}K is higher than ^{238}U , ^{226}Ra and ^{232}Th . This means that ^{40}K is more mobile and has higher solubility compared to the other radionuclides (Table 3).

3.2 Correlations of the radionuclide contents

The correlation coefficients between each two parameters for radionuclide contents of the entire studied area were calculated and are given in Table 3. ^{238}U (^{226}Ra) and ^{232}Th shows a significant positive correlation with each other for both sediment and water indicating high similarity in accumulation sources and local conditions. High correlations have been also found between ^{40}K and ^{226}Ra concentrations in both sediments and water; ^{226}Ra and ^{232}Th , and ^{232}Th shows evidence for correlation with ^{40}K as shown in figure 2 and summary of the correlation coefficients between radionuclides as listed in Table 4.

The activity ratio between each two radionuclides can be applied to assess maintenance of the proportionality between them at the surface layer. The average $^{226}\text{Ra}/^{238}\text{U}$ activity ratios in Qarun lake sediment is low 0.59 as

compared with previous values 0.95 (0.88–0.99) in Karnaphuli, 1.10 (0.99–1.14) in the Shango river in Bangladesh (Chowdhury et al. 1999). That may be because of the amount of radium in surface waters is more variable than the amount of uranium. There are many chemical and physical circumstances in which radium is adsorbed on to or desorbed from associated sediments. The general condition is that sediment arrives at the drain's mouth with radium adsorbed to the particulate surface, and then radium is desorbed in the more saline environment. That is clear in Qarun Lake where the activity ratio is less and the salinity is high (Khater, 2006).

The $^{232}\text{Th}/^{238}\text{U}$ ratios ranged from 0.45 to 0.89 with mean value 0.67. The $^{232}\text{Th}/^{238}\text{U}$ activity ratios were 0.64–1.17 in the Suez Canal, Egypt (El-Tahawy et al. 1994), 1.97 (0.94–2.90) in Karnaphuli, 2.28 (2.05–2.50) in the Shango river in Bangladesh (Chowdhury et al. 1999), 11.46 (0.96–48.3) in sediments in India (Ravisankar et al., 2014).

All sediment samples except samples (2, 5, 8 and 9) have activity $^{232}\text{Th}/^{226}\text{Ra}$ ratios above unity, while all water samples have activity $^{232}\text{Th}/^{226}\text{Ra}$ ratios below unity. The $^{232}\text{Th}/^{226}\text{Ra}$ ratios are nearly uniform across the studied area with a mean of 1.15 ± 0.24 (0.8–1.58). This value is consistent with the reported ratios of 1.32 for China (UNSCEAR 1993) and less consistent with 0.87 for USA (UNSCEAR 1993) and 0.90 for Greece (Anagnostakis et al., 1996). The $^{238}\text{U}/^{40}\text{K}$ and $^{232}\text{Th}/^{40}\text{K}$ ratios ranged from 0.016 to 0.036 and 0.012 to 0.022, respectively, the world average for both quotients being 0.067 (UNSCEAR 1988). The range of $^{226}\text{Ra}/^{40}\text{K}$ activity ratios was (0.011–0.021) with a mean value of 0.015. Generally, the results agree with some previous measurements, e.g., 0.03–0.08 (Benamar et al. 1997), 0.04–0.1 (Lee et al. 2009), 0.024–0.032 (Godoy et al. 1998), and 0.036–0.078 (El-Tahawy et al. 1994), and are smaller than others, e.g., 0.13 (0.06–0.31) and 0.23 (0.09–0.14) (Chowdhury et al. 1999). Also, this value is to be compared with 0.06 reported for Greece (Anagnostakis et al. 1996) and with 0.11 and 0.06 reported for USA and China (UNSCEAR 1993), respectively.

4. Conclusion

The activity levels and distribution of natural terrestrial radionuclides of ^{238}U , ^{226}Ra , ^{232}Th , and ^{40}K were measured by gamma-ray spectrometry system for sediment and water samples collected from Qarun lake. The activity concentrations of uranium, thorium and radium in the studied sediments are found to be normal, whereas potassium is having slightly greater value. These because the use of fertilizers in large extent in agricultural lands around the lake has affected radionuclides concentration and especially potassium containing fertilizers are one of the causes of presence of high activity of ^{40}K in soil. The extracted values are, in general, comparable to the corresponding ones obtained from other lakes and other countries and they all fall within the average worldwide ranges except potassium. Hence harmful radiation effects are not posed to the public and tourists going to the beaches for recreation or to the fishermen involved in their activities in the area as a result of the natural radioactivity of lake sediments. The spatial distribution of radioactivity is studied by multivariate statistical methods. These distribution trends indicated that these radionuclides were mainly accumulated from agriculture and industrial wastes in the fine-

grained sediments rich in organic matter contents. The linear regression relations at Qarun Lake may indicate a high similarity in accumulation sources and conditions. This study can be used as a baseline for future investigations and the data obtained in this study may be useful for natural radioactivity mapping and also be used as a reference data for monitoring possible radioactivity pollutions in future.

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Table 1: Descriptive statistical characteristics of radioactive variables of sediment and water samples of Qarun lake.

	Sediment				Water			
	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K	²³⁸ U	²²⁶ Ra	²³² Th	⁴⁰ K
Mean	23.5	14.1	15.8	933	6.4	7.9	3.2	31.3
St. deviation	9.7	6.2	6.8	384	0.59	2.3	0.43	5.7
Range	32.1	16.2	21.4	1016	2.09	6.98	1.22	17.1
skewness	0.54	0.01	0.15	0.01	0.005	0.32	0.689	0.18
kurtosis	-0.62	-1.78	-1.63	-1.80	-0.56	-1.38	-1.03	-1.48
minimum	11.4	6.2	5.2	410	5.4	4.5	2.8	22.8
maximum	43.5	22.4	26.6	1426	7.49	11.48	4.02	39.9
variance	95.7	38.5	47.2	148103	0.35	5.38	0.18	32.8

Table 2: Comparison of Radiological parameters of present work with other countries

Location	²³⁸ U (Bq/kg)	²²⁶ Ra (Bq/kg)	²³² Th (Bq/kg)	⁴⁰ K (Bq/kg)	Note	References
Egypt	23.5(11.4-43.5)	14.1(6.2-22.4)	15.8(5.2-26.6)	933.3(410-1426)	Qarun lake	Present work
Egypt	17.3(12.6-19.9)		10.03(8.5-10.6)	299.7(258.8-316.8)	Burullus lake	Dar and El Saharty, (2013)
Egypt	12.65(10.5-16)		7.24(5.4-8.3)	518.75(442-582)	Mariout Lake	
Egypt	20.37(11.2-39.3)		26.05(11.4-43.3)	329.05(163-508)	Idku lake	Fahmi et al., (2010)
Egypt	24.6(5.2-105.6)		31.4(2.3-222)	428(98-1011)	Red Sea sediment	El Mamoney & Khater, (2004)
Egypt	14.3-22		18.4-24.4	222-326	Nasser lake	Khater et al., (2005)
Egypt		17.2(12.2-20.8)	17(9.9-23.4)	246(144-360)	Burulus lake	El-Reefy, (2014)
Egypt	20.99(7.06-30.15)		14.8(5.7-20.35)	244.7(68-352)	Qarun sediment	El Zakla, (2013)
Egypt	15.92(9.87-27.28)		15.22(9.35-26.67)	644.6(958.3-3306.4)	Qarun lake	Darwish, (2013)
Egypt		4.9-20.2	3.3-35.4	59-368	Suez canal	El-Tahawy et al. (1994)
Bangladesh	37.9(20-90)		65.5(51-88)	272(217-320)	Karnaphuuli river	Chowdhury et al., (1999)
	25.4(21.6-28.3)		57.5(52.4-61.7)	255(212-292)	Shango river	
Nigeria		19.23(4.6-52.1)	31.59(6.8-46.7)	84.12(43.7-202.3)	Kainji Lake	Adamu, (2013)
KSA	49(21-76)	47(10-108)	34(15-49)	751(262-1387)	Clay	Khater, (2013)
USA	37.8(11.1-74.2)	21.4(11.4-41.2)	45.3(13-185.8)	609.3(385.9-1046.9)	Reedy River	Powell, (2007)
Jordan	11.2-677	5-31	3.6-32.8	71.5-901.1	Gulf of Aqaba	Ababneh et al., (2010)
Greece	28(9-43)	27(10-37)	30(12-46)	483(218-686)	Patras-Rion	Papaefthymiou, (2011)
Ghana	11-31.8		16.8-23.1	68.3-183.9	Beach resorts	Lawluvi et al., 2011
Albania	13-26.6		13.1-38.1	266-675	Butrint Lagoon	Tsabaris et al., (2007)
Korea		55.7(26.7-174)	91.1(30.9-157)	1005(707-1559)	Keum river	Lee et al., (2009)
India	3.67(2.2-20.9)		37.23(2.1-233.9)	387.2(313.3-482.5)	Coastal sediments	Ravisankar et al., (2014)
Turkey		12.2(4-21.5)	9(1.8-27.9)	157.7(19-590.3)	Beach sand	Özmen et al., (2014)
Worldwide	33	32	45	420		UNSCEAR, (2000)

Table 3: The specific activity ratios of radionuclides in surface sediment and water samples, and the sediment partition coefficient K_d

activity ratios and transfer factor	Range		Mean	
	Sediment	water	Sediment	Water
²²⁶ Ra/ ²³⁸ U	0.39-0.79	0.83-1.59	0.59±0.12	1.2±0.25
²²⁶ Ra/ ⁴⁰ K	0.011-0.021	0.19-0.29	0.015±0.002	0.25±0.03
²³⁸ U/ ⁴⁰ K	0.016-0.036	0.17-0.24	0.026±0.005	0.21±0.02
²³² Th/ ²³⁸ U	0.45-0.88	0.45-0.55	0.67±0.12	0.50±0.03
²³² Th/ ²²⁶ Ra	0.81-1.58	0.33-0.62	1.16±0.24	0.42±0.07
²³² Th/ ⁴⁰ K	0.012-0.024	0.09-0.12	0.017±0.003	0.10±0.008
$K_d(^{238}\text{U})$	1.7-5.8		3.65±1.4	

$K_d(^{226}\text{Ra})$	0.6-3.5	1.84 ± 0.8
$K_d(^{232}\text{Th})$	1.6-8.0	4.9 ± 2.1
$K_d(^{40}\text{K})$	13.4-56.0	30.5 ± 13.5

Table 4: Pearson correlation coefficients between all measured natural radionuclides in the examined sediment (water) samples.

	^{238}U	^{226}Ra	^{232}Th	^{40}K
^{238}U	1			
^{226}Ra	0.82(0.79)	1		
^{232}Th	0.85(0.80)	0.86(0.92)	1	
^{40}K	0.69(0.82)	0.85(0.95)	0.83(0.90)	1

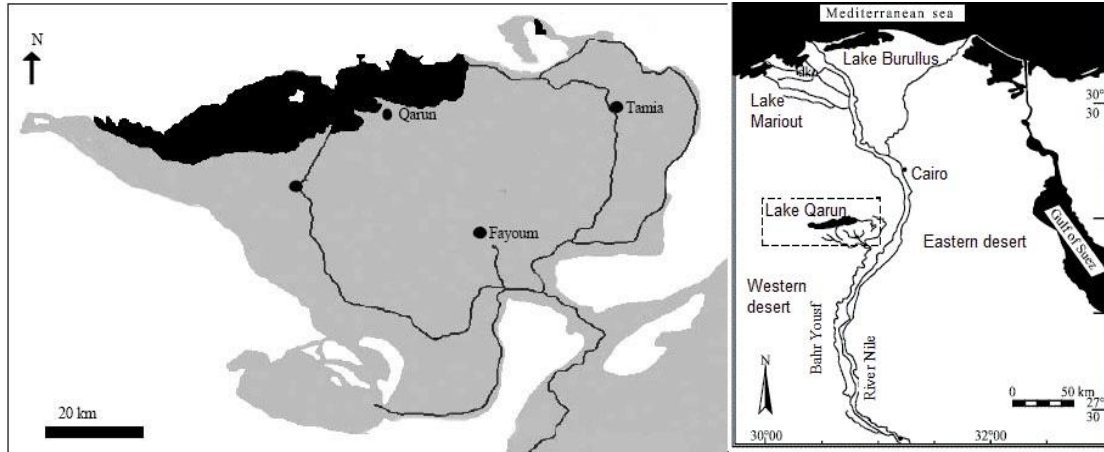


Figure 1: Location map of the studied Qarun lake

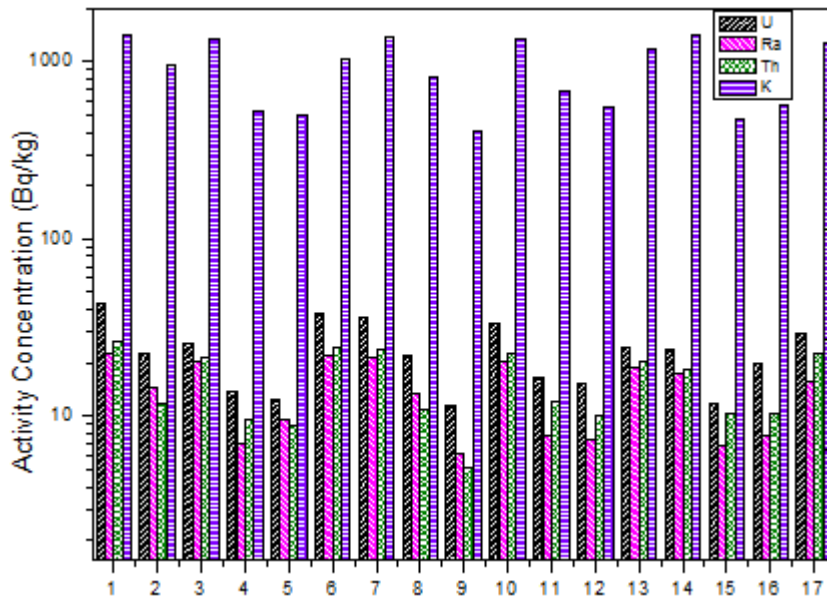


Figure 2: The activity concentrations of ^{238}U , ^{226}Ra , ^{232}Th , and ^{40}K in the sediment samples

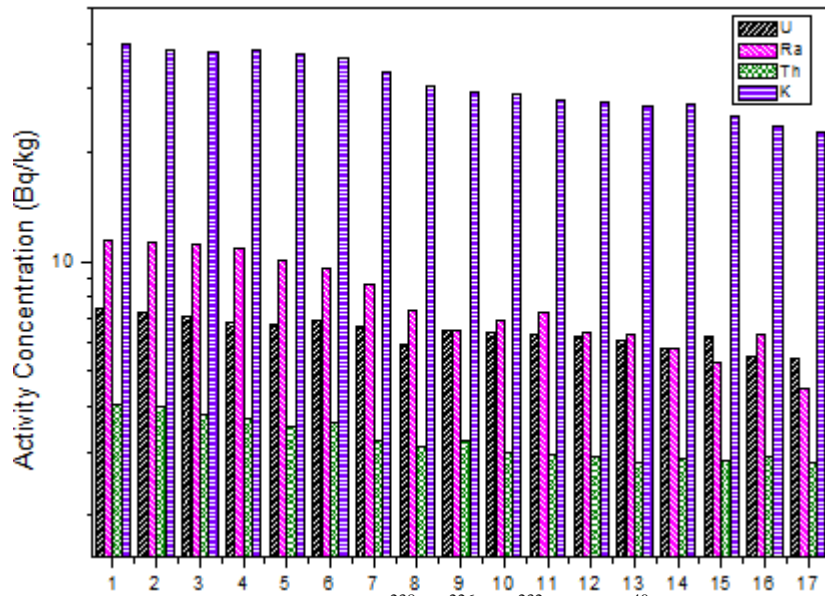


Figure 3: The activity concentrations of ^{238}U , ^{226}Ra , ^{232}Th , and ^{40}K in the water samples

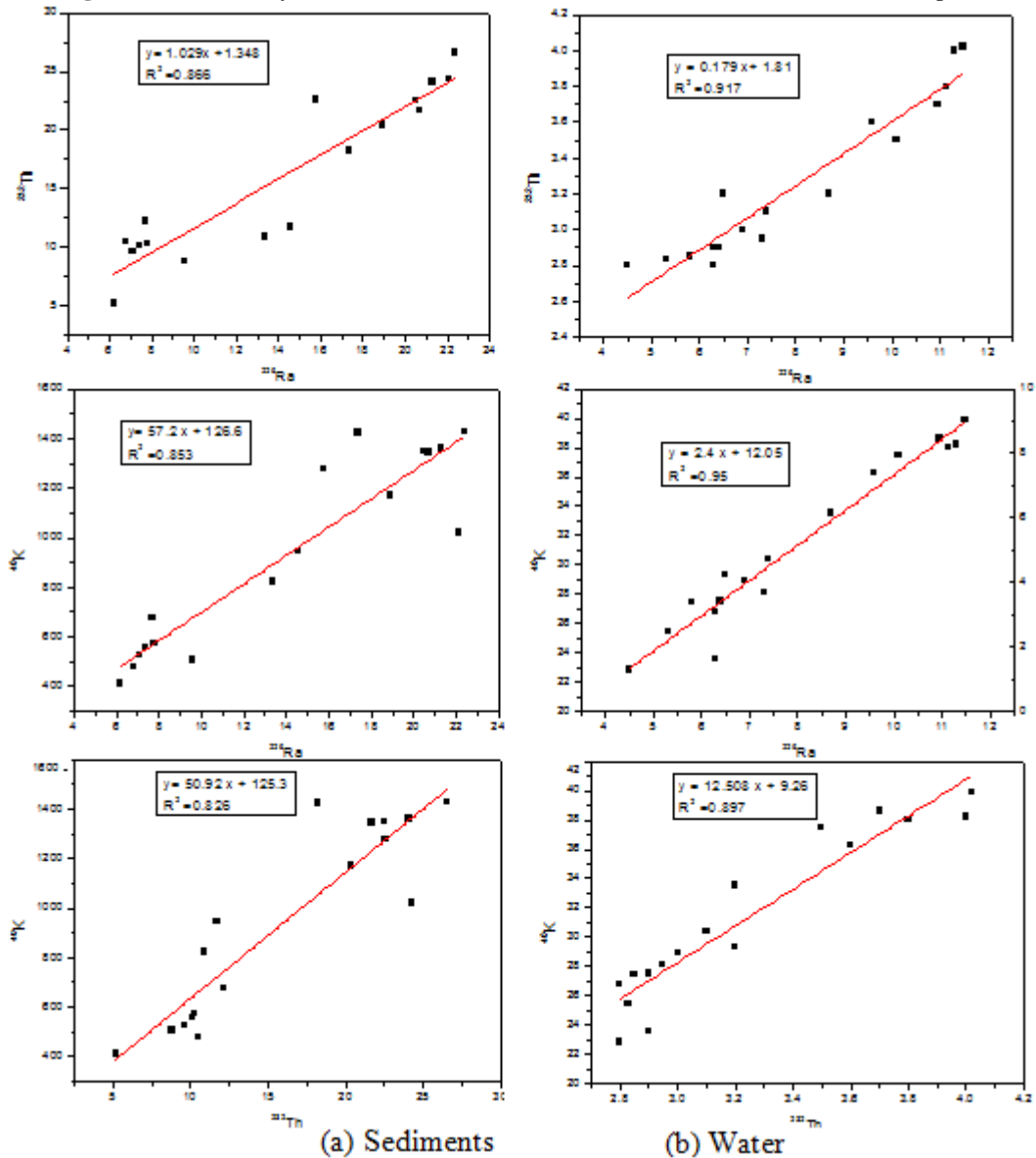


Figure 4: Correlation between activity (Bq/kg) of (^{226}Ra , ^{232}Th), (^{226}Ra , ^{40}K) and (^{232}Th , ^{40}K) in (a) sediments and (b) waters