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The Natural Radioactivity and Accompanying Radiation Hazard in the Tigris River Sediments in Baghdad, Iraq

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Abstract:
The natural radioactivity in the sediments of the Tigris River was measured using gamma spectrometry using an HPGe detector. Ten sediment samples were collected, and the target radionuclides are ^{226}Ra, ^{232}Th and ^{40}K. Radioactivity in sediment was investigated in clay and silt fractions beside the hazard indices. The ^{226}Ra concentration ranged between 6.5 and 11.2 Bq/kg, ^{232}Th from 4.1 to 9.6 Bq/kg, and ^{40}K from 133.8 to 260.2 Bq/kg. The average values of the natural radionuclides of these elements are within the worldwide average or less and comparable to those of the rivers in other countries; accordingly, the radiological hazard indices such as absorbed dose rate, annual effective dose equivalent, hazard indices and gamma index are below the recommended international values, indicating no significant hazard. The study concluded a positive relationship between the radioactivity and clay and silt content.

Keywords: Radioactivity, Hazard indices, natural radionuclides, river sediment

النشاط الاشعاعي والمخاطر الاشعاعية المرافقة لها في رواسب نهر دجلة ضمن مدينة بغداد

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الخلاصة:
تم قياس النشاط الإشعاعي الطبيعي في رواسب نهر دجلة باستخدام مقياس طيف جاما المرتبطة بكاشف جرمانيوم عالي النقاوة HPGe. تم جمع عشر عينات من الرواسب وكانت النويدات المشعة المستهدفة هي Ra-226 و Th-232 و K-40. تم إجراء تحليل الرواسب لمعرفة العلاقة بين النشاط الإشعاعي في الرواسب ومحتواها من الطين والسلت بالإضافة إلى تقديرمؤشرات المخاطرالاشعاعية بسبب النشاط الإشعاعي للنويدات المشعة أعلاه في عينات الرواسب. وأظهرت النتائج أن تراكيز الراديوم Ra-226 تراوحت بين (6.5-11.2) بيكريل / كغم ، وتراوحت تراكيز - الثوريوم-232 من (4.1-9.6) بيكريل / كغم ، بينما تراوحت تراكيز K-40 بين (133.8 - 260.2) بيكريل. / كغم. وبينت النتائج ان متوسط قيم النويدات المشعة الطبيعية Ra-226 و Th-2323 و K-40 ضمن المتوسط العالمي أو أقل ومقاربة للمتوسط المحتوى الاشعاعي في رواسب الأنهار في البلدان الأخرى ، وبالتالي فان مؤشرات المخاطر الإشعاعية مثل معدل الجرعة الممتصة ، المكافئ السنوي للجرعة الفعالة ومؤشرات المخاطر ومؤشر جاما أقل

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من القيم الدولية الموصى بها ولا تشكل اي مخاطر اشعاعية غير اعتيادية. وخلصت الدراسة أيضا إلى وجود علاقة إيجابية جيدة بين النشاط الإشعاعي في الرواسب ومحتوى الرواسب من الطين والسلت.

Introduction

Most of the natural radionuclides found in river sediments are those with long half-lives, which include the uranium and thorium series and the single nuclides, the most important of which is potassium-40. Natural radionuclides are present in the sediments of rivers due to weathering processes due to the interaction between the water and the rocks on which it flows, which reflect the geological history [1] and those transported by rain and flows [2]. The concentration of these nuclides varies based on their abundance in the parent rocks, the geological characteristics of the region, radionuclide solubility, hydrochemical properties of water and sediment, and the proximity of radiation sources to the river [3.4]. The concentrations of natural radionuclides in soil and sediments are also different, as their average worldwide is about 33 Bq/kg (Becquerel per kilogram) for uranium-238, 45 Bq/kg for thorium-232, but their concentration may be as high as 480 Bq/kg in phosphate rock [5.6], and 412 Bq/kg for potassium-40 [7]. In soil, the worldwide average for radium-226 is about 32 Bq/kg [7], and in surface water, it ranges between 0.001 - 0.1 Bq/L [8]. At the same time, the use of chemical fertilizers in agriculture increases the radioactivity in soil [9].

The distribution of natural radionuclides in recent sediment could reflect terrestrial material input. U-progenies and anthropogenic radionuclides such as ^{137}Cs are used to recognize their distribution, possible sources and patterns along the investigated area. In Iraq, many researchers have studied the radioactivity in rivers sediments; a study conducted by Ali and Alai [10] showed that the concentrations of radioactive elements in the sediment and water of the Euphrates River western Iraq in the range of (40-213) Bq/kg for radium-226, (22-256) Bq/kg for thorium-232 and (411-45) Bq/kg for potassium - 40. Attiyah [11] studied the radioactivity of the sediments in the different phases in the drinking water refining stations along the Tigris River in Baghdad city. Ali [12] measured the activity of the radionuclides (^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs) in sediments in the upper part of the Euphrates River and springs in the region and concluded that Ra-226, Th-232, and K-40 activities were within the world average of these radionuclides in rivers.

In contrast, relatively high activities in spring sediment were found, especially ^{226}Ra. The radioactivity in river sediment in Sulaymaniyah Governorate, northern Iraq, was investigated [13]. He found that the activity of ^{226}Ra, ^{232}Th, and ^{40}K. were low compared to others in Iraq. Al-Alawy et al. [14] studied the natural radioactivity in water and sediments of the Al-Husseiniya River in Karbala, southern Iraq, and found that the average activity concentrations in sediment.

Three sites along the Tigris River were investigated for Alpha radioactivity levels in sediments in Baghdad by using CR 39 SSNTD [15]. They concluded that human activity affects the uranium concentration, which increases from north to south along the river in the city.

Mohammed and Tawfiq [16] investigated the radioactivity in selected samples from the sediment of the Tigris River. They concluded that the average activities were about 38, 21, 606, and 28 Bq/kg of ^{238}U, ^{232}Th, and ^{40}K, respectively, as well as ^{137}CsNumerous studies have been conducted regarding estimating radionuclides of terrestrial origin in river sediments using different techniques; here are some examples. In Australia, the radioactivity in the river was used as a tool for understanding the erosion and chemical weathering processes at the scale of a watershed [17]. Bastos and Appoloni [18] studied the natural radioactivity in the geologic formations in Brazil's region of the Tibagi river hydrographic basin. They concluded that the highest dose rate values were obtained from felsic rocks (rhyolite of the Castro group, 129.8 ± 3.7 nGy.h-1, and granite, 167 ± 37 nGy.h-1 while only one sample reached a Raeq (radium equivalent will be discussed later in this study)value higher than 370 Bq.kg-1.The lifetime cancer risk associated with the natural radioactivity content in river sediments was estimated [2, 19-21]. Ramasamy et al. [22] evaluated the distribution of natural radionuclide content in river sediments in India.

Radioactivity in Nigeria's surface soil and drinking water were evaluated using gross alpha and gross beta [23]. In Bangladesh, Yasmin et al. [24] investigated the natural radioactivity in soil, sand and sediments, and they found that the average activity of uranium-238 and thorium-232 in sediment is higher than that in soil and sand. Thangam et al. [25] studied and discussed the natural radioactivity of a river in Thamirabarani, Tamilnadu in India.

This study aims to evaluate the radioactivity in the sediments of the Tigris River in Baghdad city and estimate the hazard indices accompanying these sediments in addition to evaluate the potential doses in water.

Study area

The study area, represented by a part of the Tigris River, is located within the city of Baghdad, central Iraq, between the points with coordinates (44.34267° - 33.43916°) (Al-Muthana Bridge) Northern Baghdad and (44.45146°- 33.23723°) (Al-Rasheed Bridge), southern Baghdad (Figure 1)

Tigris River penetrates the city of Baghdad while it is in its maturity, forming many meanders and several small islands due to the slowing of the water velocity and the increase in the sedimentation of the river. The Tigris River flows within the flat sedimentary plain zone, the average slope of the land surface is 0.1 m / km to the south, and the height of the earth surface in the study area ranges between 22 m - 32 m above sea level. Figure 1 represents the study area within the city of Baghdad, including the sediment sampling locations.

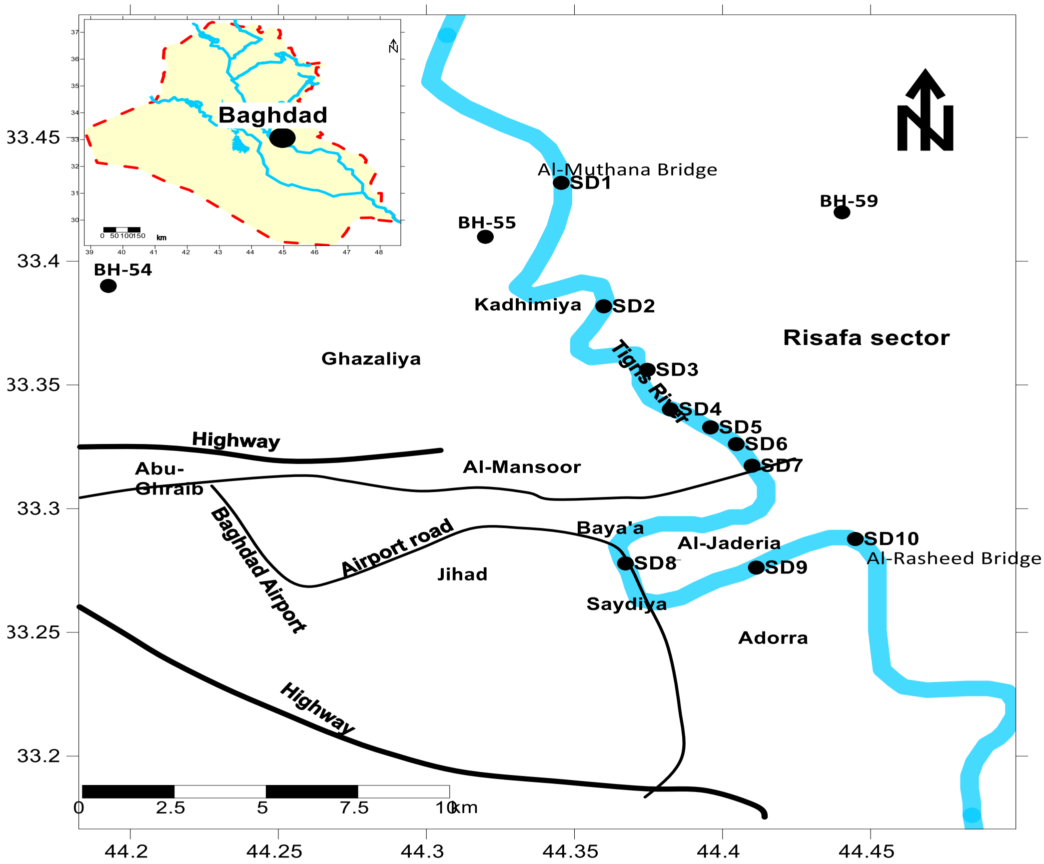

Figure 1: Sampling locations of sediments along the Tigris River near the bridges along the river. The names of the bridges are listed in Table 1.

Geology of the Study Area

The sediment in the study area represents a Quaternary sediments deposited in the Mesopotamian Zone. These sediments were deposited due to the interaction of the rivers passing in the area, especially the Tigris and Euphrates Rivers, and the marine incursions from the Arabian Gulf effect [26].

The sediments are mainly from the deposits of the Tigris River and its tributaries, in addition to aerobic sediments. The riverbeds consist of sand, silt, and clay, which is characterized by inflexions and numerous islands.

The river's width differs from place to place; therefore, it is considered one of the active rivers because of the continuing erosion and sedimentation in the islands, erosion in concave areas and sedimentation in convex regions.
The Tigris River in the city of Baghdad is part of the river that passes in the late stages of maturity, where it is characterized by the expanding of inflexion zones that reflect the deviation from the normal stream of the river and reflect the advanced stage of the life cycle of the river [27].

The soil of Baghdad, which is part of the alluvial plain, is of soil-borne type of rock fragments deposited by the upper basin of the Tigris and Euphrates rivers. These fragments are transmitted by hydraulic river energy and deposited when the hydraulic forces cannot be attributed to sediment movement [28].

The different nature of the sediment is due to repeated floods on the flood plains in addition to irrigation methods, which led to variations in surface soil from one area to another with time. Irrigation channels and drainage digs in the area lead to the accumulation of irregular and heterogeneous sand and alluvial sediments. The sediment in the study area is of different mud and coarse sand and gravel sizes. Boreholes (BH-55) and BH-54 (Figure. 1) are located in the northern part of the study area showing that generally, the area covers with Quaternary sediments (Qf: flood plain deposits sand, slit and clay and Qm: marsh deposits mud with organic materials) of Pleistocene and Holocene age (Figure. 2).

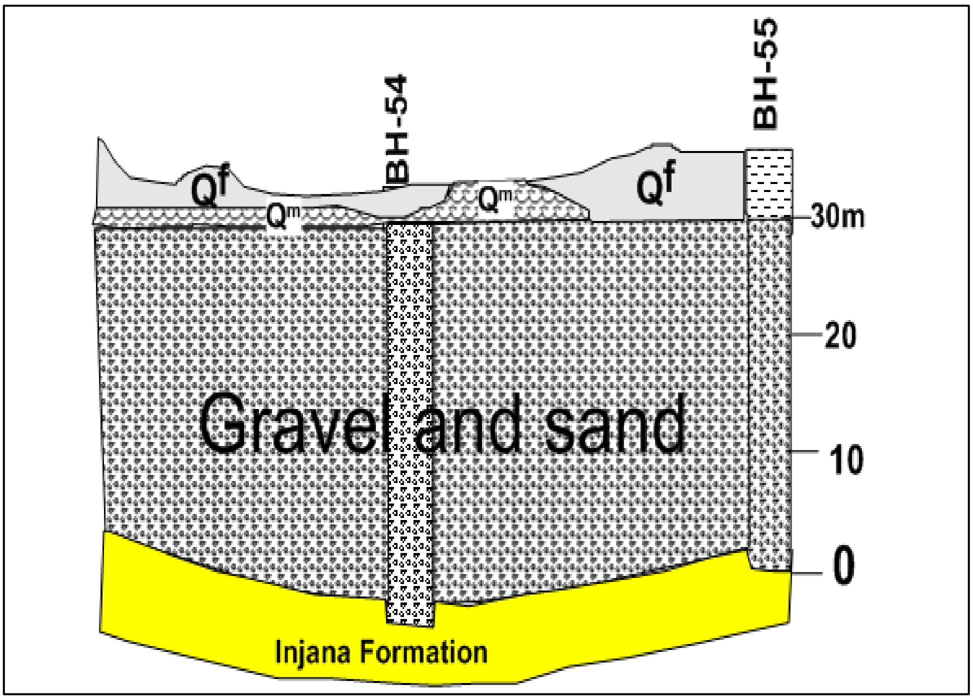

Figure 2: geological section in two boreholes in the study area [29]. The elevation scale is above sea level.

The upper part of the soil in the study area is characterized by being heterogeneous laterally and vertically due to the influence of the seasonal floods of the Tigris River, and recently, the impact of human activities is clear.

Materials and Methods

Ten samples were selected from the Tigris River sediments near the bridges and water refining stations along the river within Baghdad (Table 1 and Figure. 1).

Table 1: the sampling locations along the Tigris River in Baghdad city.

Sample ID	East (°)	North (°)	Bridge Name
SD1	44.34553	33.43167	Al-Muthana
SD2	44.33909	33.38177	Al-Aameh
SD3	44.37459	33.3561	Sarafia
SD4	44.38234	33.34011	Bab-Al-Muuadam
SD5	44.39594	33.33285	Al-Shuhada
SD6	44.40463	33.32606	Al-Ahrar
SD7	44.40996	33.31734	Al-Jumhuria
SD8	44.3993	33.29651	Al-Jadiria
SD9	44.41141	33.27617	Two-stories
SD10	44.44484	33.287731	Al-Rasheed

At least 2 kg of sediment was collected from the bottom of the river and placed in nylon bags on which all information was written (sample number, Coordinates, date, and type). The sampling locations were accurately determined using GPS (± 10 feet). The samples were sent to laboratories for preparation and radioactivity analysis.

The samples were dried in an electric oven with a fixed temperature of (50 ºC) to avoid the loss of radionuclides from the soil until they completely dry. Hand mixing of the sediments before sub-sampling [31] removes impurities and gravel. Then, 1 kg of each sample was taken after the samples were sieved using a 1 mm sieve to prepare them for the radioactivity measurements. All samples are stored for 25 days in a completely sealed Marinelli beaker to achieve equilibrium between radon and its daughters [9]. After measuring the radioactivity, these selected samples were subjected to volumetric analysis to determine their sand, silt and clay content using the separation method using special sieves. The diameter of the sieve used to separate the silt and clay was (0.037) mm.

Radioactivity measurement

Radioactivity in the sediment samples is measured using a gamma-ray spectrometry system at the Center Radiation Production (CRP) Ministry of Environment - Iraq. The system used a high-purity germanium (HPGe) detector with an efficiency of 40%, and the computer code Genie - 2000 is used for measuring and analysis. The specific activity for gamma-ray lines of ^{214}Bi at 609.3 and 1764.5 KeV are used to determine the particular action of ^{226}Ra. For ^{232}Th, ^{208}Tl with energy lines at 583.2 and 2614.5 KeV while at 338.4 and 911.2 KeV from ^{228}Ac are used. The activity of ^{40}K is measured at gamma line 1460.8KeV. Every sample was counted for 10800 sec. Germanium detectors have good energy resolution, making them suitable for many applications where complex gamma-ray spectra are studied with energy ranging from 50KeV to 3000KeV. To make the efficiency and energy calibration a standard source of a multi-energy made by the American Canberra Company was used. A Marinelli geometrical shape was used to measure the samples' activity.

Results and discussion

Table 2 shows the radioactivity of ^{226}Ra, ^{232}Th, and ^{40}K in the samples. The percentage of accuracy analysis for all samples is below or slightly above 5%. The results in Table 2 show no high coefficient variation, meaning there is no high variation in the radioactivity of the sediments from site to site along the river because sediments in river bottoms of the same source have no significant variation in their chemical and mineralogical properties. The table also shows the average values and the standard deviations.

Table 2: radioactivity in sediment samples in the Tigris River

Sample ID	Activity (Bq/kg)		
	^{238}U (^{226}Ra)	^{232}Th	^{40}K
SD1	9.7 ± 0.1	8.7 ± 0.1	260.2 ± 10
SD2	9.1 ± 0.1	9.3 ± 0.2	252.4 ± 9
SD3	8.2 ± 0.2	8.7 ± 0.1	195.5 ± 11
SD4	8.5 ± 0.1	8.3 ± 0.2	233.1 ± 12
SD5	8.9 ± 0.2	9.6 ± 0.4	244.5 ± 14
SD6	6.7 ± 0.1	6.4 ± 0.1	189.7 ± 7
SD7	8.9 ± 0.1	5.9 ± 0.1	196.5 ± 10
SD8	6.3 ± 0.1	4.1 ± 0.1	135.2 ± 5
SD9	6.5 ± 0.1	6.8 ± 0.1	140.2 ± 5
SD10	8.7 ± 0.1	7.5 ± 0.2	205 ± 8
Range	6.3-9.7	5.9-9.6	135.2-260.2
average	8.2	7.5	205.2
St. Dev.	1.2	1.7	43
Coef. Var.	14.8	22.9	21.2

Table 2 indicated that there were concentrations of Uranium-238 (^{226}Ra) that ranged between (6.5-11.2) Bq/kg, and Thorium-232 concentrations ranged from (4.1-9.6) Bq/kg, while the concentrations of Potassium-40 ranged between (133.8-260.2) Bq/kg and cesium-137 concentrations between (2.5-6.3) Bq/kg.

Geological formations provide the sediments with the natural radionuclides, ^{238}U series, ^{235}U series, ^{232}Th series and ^{40}K. Potassium is a major element widely distributed in crustal rocks [32]. ^{40}K represents about 0.0119% of total natural potassium [33]. Because potassium is widespread in various minerals and clays, the interaction of river water with river sediment will lead to the release of potassium, whether it is dissolved in the water or re-deposited within the river sediments when the geochemical environmental conditions change. Thus, it has higher concentrations in the sediment samples of the Tigris River [34].

In all sampling sites along the Tigris River, the mean activity concentration of naturally occurring radionuclides is ^{232}Th< ^{226}Ra< ^{40}K. The increasing trend of ^{40}K is due to clay sediments [35].

The average concentration of ^{226}Ra, ^{232}Th, and ^{40}K in measured samples are below the world average values for ^{226}Ra, ^{232}Th, and ^{40}K in the soil, are 32 Bq/kg, 45 Bq/kg and 410 Bq/kg, respectively [7], and these values comparable with the results obtained by other researchers [1, 35]. In the current study, the activity of ^{137}Cs in the Tigris River sediments is found to range between BDL and 0.79 Bq/kg. These values are too low compared to that of concentration in the Euphrates River [12].

The natural radioactivity of the sediments of the Tigris River reflects the lithological composition of the rocks and soils on which the river flows. It is known that the Tigris River, especially in its central parts, flows within rocks with a relatively low level of radiation represented in the sediments of the Quaternary era. Hence, its sediments had a relatively low radioactivity if compared with the radioactivity of the sediments of the Euphrates River [10, 12].

The average concentration of uranium-238 (^{226}Ra) in the sediment samples was about (8.2) Bq/kg, while the average concentration of thorium-232 was (7.5) Bq/kg, and potassium-40 was about (205.2) Bq/kg. These values are close to or less than the rates of these radionuclides in most surface soils and rocks in some regions of Iraq [36, 37] and less than the concentrations of these radionuclides in the sediments of the Euphrates River [10, 12]. It is close to or lower than the average concentrations of radioactive elements in river sediments in some countries [1, 38] and lower than the level of natural radioactive elements in rocks of Earth's crust, with an average of about (40, 40, and 400) Bq/kg for radium-226, thorium-232, and potassium-40, respectively [39].

Table 3 shows the silt and clay percentage in the sediment samples. To evaluate the relationship between the volumetric properties of the sediment and its radioactivity content, a correlation between the activity of ^{226}Ra, ^{232}Th, and ^{40}K and silt and clay percentages of the sediment samples was done, as shown in Figur4

Table 3: Volumetric analysis of the sediments

ID	Silt and clay%	Ra-226	Th-232
SD1	7.5	9.7	8.7
SD2	7	9.1	9.3
SD3	6.5	8.2	8.7
SD4	6.9	8.5	8.3
SD5	7.2	8.9	9.6
SD6	5.1	6.7	6.4
SD7	6	8.9	5.9
SD8	4.2	6.3	4.1
SD9	4.7	6.5	6.8
SD10	6.4	8.7	7.5

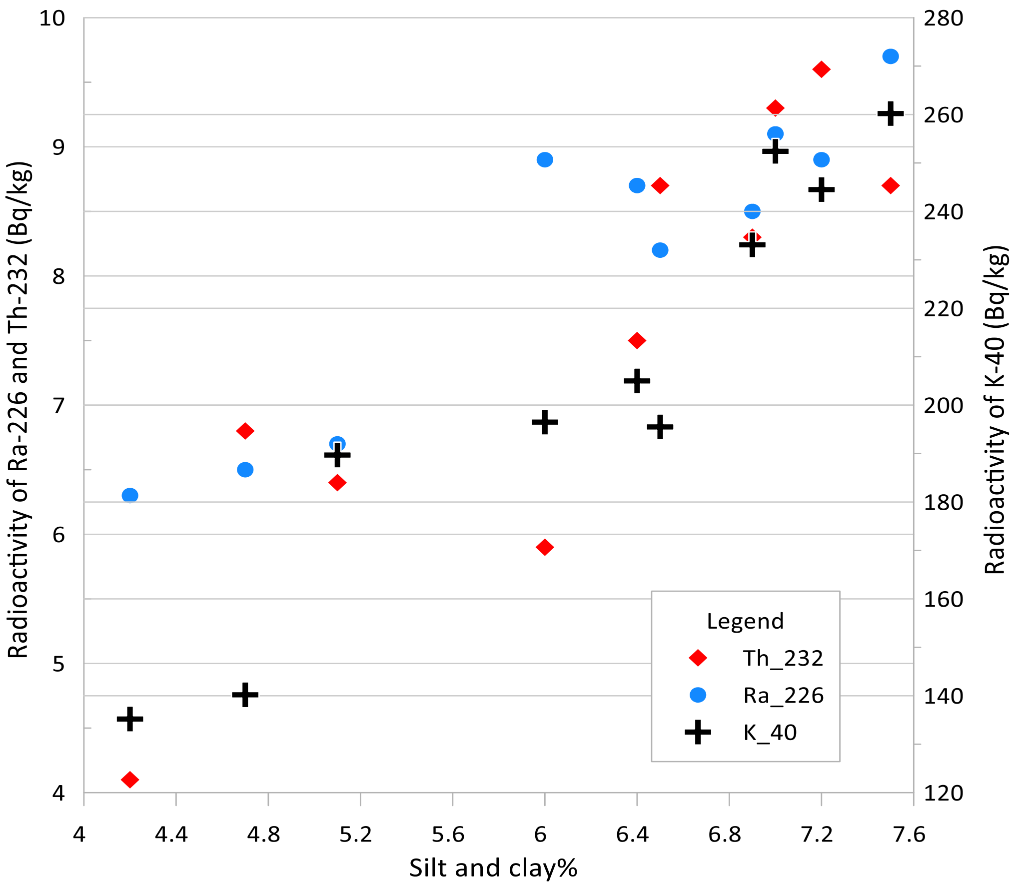

Figure 4: good positive relationship between silt and clay content and radioactivity of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in the selected sediment samples of the Tigris River in Baghdad city.

Assessment of radiation hazard

Air- Absorbed dose rate

The external terrestrial gamma-ray absorbed dose rate is calculated (Table 4) by converting the natural radionuclides concentration, ^{226}Ra, ^{232}Th, and ^{40}K in the sediments using the conversion factor given by [40] as in Eq. (1):

$$D\ (nGy/h) = 0.462CU + 0.604CTh + 0.0417\ CK \quad (1)$$

Where D is the absorbed dose rate in nGy/h and CU, CTh, and CK are the measured concentrations of 238U, 232Th and 40K in the river sediments in Bq/kg, respectively.
The absorbed dose rates range from 11 nGy/h to 20.6 nGy/h. All the values are comparable to those in other river sediments worldwide [1, 38].

Gamma index

In order to examine whether the safety requirements for using the sediments as construction materials are being fulfilled. The gamma index proposed by the European Commission (41) was calculated using Eq. (2):

$$I_{\gamma} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_{k}}{3000} \quad (2)$$

A_{Ra}, A_{Th} and A_{K} are the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in (Bqkg-1).

Therefore, controls should be based on a dose range of 0.3–1 mSv/y, as recommended [42]. The index should range between the values 0.5-6 depending on the dose criterion and the way and amount of the material used in a building [41]. The values of Iγ in all sediment samples are within 0.09-0.16 (Table 4), which gives an annual effective dose < 0.3 (35), so no hazard exists.

Internal and external hazard indices

The internal (H_in) and external hazard (H_ex) indices were calculated using Eq. (3) and Eq. (4) [43-45]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{k}}{4810} \leq 1 \quad (3)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{k}}{4810} \leq 1 \quad (4)$$

where A_{Ra}, A_{Th} and A_{K} are the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively, in (Bqkg-1).
Both indices are recommended to be less than the unity [43-45]. All the values in the current study (Table 4) are less than 1, so no hazard can be expected.

Table 4: Radiation Hazard indices resulting from using the Tigris River sediment in Baghdad city

Sample ID	D(nGy/h)	Gamma index	Hex	Hin
SD1	20.6	0.16	0.11	0.14
SD2	20.3	0.16	0.11	0.14
SD3	17.2	0.14	0.10	0.12
SD4	18.7	0.15	0.10	0.13
SD5	20.1	0.16	0.11	0.14
SD6	14.9	0.12	0.08	0.10
SD7	15.9	0.12	0.09	0.11
SD8	11.0	0.09	0.06	0.08
SD9	13.0	0.10	0.07	0.09
SD10	17.1	0.13	0.10	0.12

When the concentrations of a radionuclide in the sediment are known, the distribution coefficient (Kd) from the sediment to the water can be estimated, which is equal to the radioactivity of the radionuclide in the sediment (Bq/kg) to its radioactivity in the water (Bq/l). The distribution coefficient is used to calculate the radiation dose from consuming filtered water. The radioactivity of radium-226 in the waters of the Tigris River was estimated within the study area using the distribution coefficient (Kd) from sediment to water approved in the publications of the International Atomic Energy Agency [46] and [47].

Where the distribution coefficient of radium-226 was estimated as (7.4 x 103); and then estimated the radiation doses resulting from the consumption of that water in units (milli Sv/y) using the dose conversion factor as 2.8 × 10 -1 (Sv / kBq) [48]. The result is shown in Table 5.

Radium-226, one of the daughters of uranium-238, is the most dangerous successor in the decay, being the most mobile in the aquatic environment and being the direct parent of radon, the most hazardous gas in the uranium series, the most significant contributor to human radiation doses (6, 47), so the concentrations of radium-226 in the waters of the Tigris River were estimated within the study area, and it was found that the concentrations did not exceed (0.0013) Bq/l (Table -5). It is less than those in the Euphrates River (0.008 Bq/l) within the Shanafiya –Samawa area, southern Iraq (48). These concentrations give an average radiation dose of about 3.1 x 10-7 mSv/year, and the highest radiation dose resulting from water consumption was 3.7 x 10-7 mSv/year, which is a deficient value compared to the annual dose limits allowed for the population (1 mSv/year) resulting from all dose sources except radiation doses resulting from the radiation background [9, 48].

Table 5 the calculated activity ^{226}Ra in the Tigris River and the potential estimated radiation dose due to consumption the filtered water.

Sample ID	^{226}Ra in sediment (Bq/kg)	Calculated ^{226}Ra in the river water (Bq/l)	Estimated radiation dose (mSv/y)
SD1	9.7	0.0013	3.7E-07
SD2	9.1	0.0012	3.4E-07
SD3	8.2	0.0011	3.1E-07
SD4	8.5	0.0011	3.2E-07
SD5	8.9	0.0012	3.4E-07
SD6	6.7	0.0009	2.5E-07
SD7	8.9	0.0012	3.4E-07
SD8	6.3	0.0009	2.4E-07
SD9	6.5	0.0009	2.5E-07
SD10	8.7	0.0012	3.3E-07

Conclusion

Sediment is an indicator and accumulator of radionuclides that are insoluble and adsorbed on insoluble material of the aquatic system. The most significant contributor to the collective effective doses received by world populations comes from terrestrial radiation. So, it is necessary to evaluate the radioactivity in the sediments and estimate the radiological hazards. The radioactivity in the sediment of the Tigris River was investigated, the average values of the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K were found to be within the worldwide average or less and comparable to those of the rivers in other countries. No significant anomaly demands rather concentrated study, and accordingly, the radiological hazard indices such as absorbed dose rate, annual effective dose equivalent, hazard indices and gamma index are below the recommended international values and have no risk. However, it is a significant environmental issue to continue monitoring the radioactivity of the river, especially for the rest of the river. The natural radioactivity of the river depends mainly on the geological of the sediment. There is a high positive relationship between silt and clay content and the radioactivity of the sediments in the Tigris River.

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