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Assessment of Natural and Industrial Radioactivity and Radiological Hazard in Sediments of Tigris River of Dhuluiya City, Iraq

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ABSTRACT

Specific activity of Natural and industrial radionuclides in sediments samples collected from the Tigris river of Dhuliya city of Salah Uddin Governorate was measured using γ -ray spectroscopy. The γ -ray spectrometry was carried out using NaI(Tl) detector. The results indicated that specific activity of ²²⁶Ra ranged from 1.51Bq/kg to 84.68Bq/kg with average 15.48Bq/kg, ²²⁸Ac ranged from 1.47 to 49.84Bq/kg with average 8.36Bq/kg, ⁴⁰K ranged from 56.51Bq/kg to 711.34Bq/kg with average 418.47Bq/kg and ¹³⁷Cs ranged from 2.77Bq/kg to 9.78Bq/kg with average 2.88Bq/kg. The average concentrations of these radionuclides were less than the internationally accepted limit except for ⁴⁰K radionuclides. The radiological parameters of the radium equivalent activity (Raeq), absorbed gamma dose rate (D_{γ}), external and internal annual effective dose (AEDE), external and internal risks (H_{ex}, H_{in}), and gamma radiation representative level index (I_{γ}) were calculated and found to be lower than the internationally acceptable limit value.

Keywords: Natural Radioactivity, Gamma Spectroscopy, NaI(Tl) detector, Radiological Parameters

INTRODUCTION

Natural radioactivity has existed since the creation of the universe due to the long half-life of some radionuclides. Natural radionuclides emit γ , alpha, and beta radiation from the ²³⁸U, ²³²Th series and ⁴⁰K isotop everywhere in the world (Bolca *et al.*, 2007; Nada *et al.*, 2017).

The radiation background of these radionuclides is high if the environment is polluted by either human or natural activities (Chandrasekaran *et al.*, 2014).

The main external source of radiation for the human body, which is represented by gamma radiation, from these radionuclides is the largest contributor to the external dose absorbed by mankind (UNSCEAR, 2000).

The Earth's environment contains natural radioactivity and is widespread. It is found in various geological formations in sediments, rocks, soil, water, plants, and air (Singh *et al.*, 2005; Yang *et al.*, 2005).

The level of natural radioactivity of river sediments is caused by the presence of a concentration of natural radionuclides in soil and rocks (Xinwei *et al.*, 2008).

The radiation background that is produced from the ground is related to the types of rocks from which the soil is created. Therefore, natural radioactivity is mainly dependent on geographical and geological conditions (Florou and Kritidis, 1992).

High levels of radiation are in igneous rocks such as granite, shale and phosphate rocks, as they contain a high percentage of natural radionuclides, while in sedimentary rocks, lower levels (UNSCEAR, 1993).

In the first place, natural radioactivity and external exposure resulting from gamma radiation depend on geographical and geological conditions and will be at different levels in the soil of each country in the world (A. Abbady, 2006, El-Arabi *et al.*, 2000).

The measurements of the concentration and distribution of radionuclides are important because they provide useful information in monitoring environmental radioactivity in every countries of the world (AbdEl-mageed *et al.*, 2011).

The study aims to measure the radioactivity of natural and industrial radionuclides in sediment in Tigris River in Dhuluiya city and to calculate the risks caused by radiation due to the presence of radionuclides, such as: radium equivalent activity Raeq, representative level indicator I γ and the external risk index Hex. Additionally, the average absorbed dose and annual effective dose (mSv/y) in the air are calculated as a measure of human exposure to natural radiation.

MATERIAL AND METHODS

1. Study Area

Salah al-Din Governorate is an Iraqi governorate located in central Iraq north of Baghdad, the center of which is Tikrit. The province is astronomically located on a longitude 43.35 degrees east of the Greenwich Line and on a latitude of 34.27 degrees north of the equator, and its land area is 24.363 square kilometers. Where the study area including the city of Dhuluiya and the neighboring villages is located. The city of Dhuluiya is 80 km north of the capital Baghdad on the east bank of the Tigris River, its population is about 55 thousand people and it is an administrative unit that follows the district of Balad in Salahuddin governorate.

2. Sample collection and Preparation

Twenty-three of sediment collected during the month of July in 2019 from the study area along the Tigris river in Al-Duluiya city, Salah Al-Din governorate. All samples were exposed to the sun for 10 days in order to to dry completely, the samples were grinded to become a fine powder and sieved with a sieve size 75 μ m mesh. One kilogram of each sample placed in a polyethylene marinelli packer is closed tightly using the paraffin tape and left for a month to obtain radiative balance between ²²⁶Ra and ²³²Th and their respective progenies (Nada *et al.*, 2017).

After that time, the samples were examined using a gamma ray spectrometer by NaI (TI) (3 "x3") to measure the activity of natural and artificial radionuclides.

3. Gamma spectrometric measurements

Gamma spectrometer used in this work consists of NaI(TI), 3"x3" detector manufactured by Canberra (USA). Multichannel analyzer (MCA) linked to the detector through the base of the preamplifier, the pulse height analyzer, to read the data.

To reduce the radiation background the detector is surrounded with a lead shield are described using the energy calibration method and then quantifying them using the relative method by comparison with standard sources.

The minimum detection activity (MDA) is defined as the smallest amount of radionuclide that can be reliably identified and coded for a specific measurement. The (MDA) for each radionuclide was the detector and the personal computer PC-MCA (4096 channel) model (Canberra, USA), the spectral data from the detector was analyzed by using computer software (GINE-2000). The time of counting was 36,000 sec for measure the natural and artificial radionuclides in sediment samples (Darko *et al.*, 2005).

Energy and efficiency calibration of gamma spectrometer were carried out using 1.0 L Marinelli beaker with mixed radionuclide standard source.

The process involves measuring the radionuclides quantitatively and qualitatively. The radionuclides calculated using equation (Nada *et al.*, 2017).

$$MDA = \frac{LD}{T \times Eff(E) \times P(E) \times M}$$
(1)

Where: T is the counting time (sec), Eff(E) the photo peak efficiency, P(E) is the gamma emission probability, M (kg) is the mass of the sample, LD is the limit of detection and calculated from the equation (UNSCEAR, 2000):

 $LD = LC + K \sigma D$

(2)

Where: LC is the level critical below which no signal can be detected, σD is the standard deviation, and K is the error probability.

4. Activity concentration of sediment Samples:

Calculating the specific activity concentration in each sample in the spectrum by using the following equation (Beck, H.L, 1972).

$$A = \frac{N}{P(E\gamma) \times Eff \times Tc \times M}$$

(3)

(4)

where: A: The specific activity concentration of radionuclides measured in (Bq/kg),

N: net area under the peak, M: mass of the soil sample (kg), Eff: The efficiency of the detectors at energy $E\gamma$, $P(E\gamma)$: is the abundance at energy $E\gamma$ and Tc: The time of measurement which was equal to (7200 s).

5. Radiological Parameters

The radium equivalent activity (Ra_{eq}) was calculated as (UNSCEAR, 2000):

$$Ra_{eq} = A_{Ra} + (\frac{10}{7}) A_{Th} + (\frac{10}{130}) A_{K}$$

Where A_U , A_{Th} and A_K are the activity concentration (Bq/kg) of ²³⁸U, ²³²Th and ⁴⁰K radionuclides. In the Raeq definition, it is assumed that 10 Bq kg⁻¹ of ²²⁶Ra, 7Bq kg⁻¹ of ²³²Th and 130 Bq kg⁻¹ of ⁴⁰K produce equal gamma-ray dose rate (Stranden, 1976).

The external hazard index, H_{ex}, was calculated from the expression as (UNSCEAR, 2000):

$$H_{ex} = \frac{ARa}{370} + \frac{ATh}{259} + \frac{AK}{4810}$$
(5)

The internal hazard index calculated from the expression as (UNSCEAR, 2000):

$$H_{in} = \frac{ARa}{185} + \frac{ATh}{259} + \frac{AK}{4810} \le 1$$
(6)

Where A_{Ra} , A_{Th} and A_K are the specific activities of ²²⁶Ra, ²³²Th and ⁴⁰K respectively in Bq/kg.

Calculating the absorbed dose rates outdoor $(D\gamma)$ due to gamma rays in air at 1 m above the ground surface, the coefficients correspond to $0.92 \text{ nGy/h per Bq kg}^{-1}$ for ²²⁶Ra, 1.1 nGy/h per Bq kg⁻¹ for 232 Th and 0.080 nGy/h per Bq kg⁻¹ for 40 K from equation (UNSCEAR, 2000):

(7)

 $D\gamma (nGy/h) = 0.92A_{Ra} + 1.1A_{Th} + 0.080A_{K}$

To calculated the annual effective dose equivalent (mSv/y), take into account the conversion factor from absorbed dose in air to effective dose equivalent and the indoor occupancy factor, the conversion coefficient from absorbed dose in air to effective dose (0.7 Sv/Gv) received by adults and 0.8 for the indoor occupancy factor, outdoor occupancy factor (0.2) from the two equations (UNSCEAR, 2000):

$$AEDE_{in}\left(\frac{mSv}{y}\right) = D\gamma\left(\frac{nGy}{h}\right) \times 8766 \frac{h}{y} \times 0.80 \times 0.7 \frac{Sv}{Gy}$$
(8)
$$AEDE_{out}\left(\frac{mSv}{y}\right) = D\gamma\left(\frac{nGy}{h}\right) \times 8766 \frac{h}{y} \times 0.20 \times 0.7 \frac{Sv}{Gy}$$
(9)

Where $D\gamma$ (nGy/h) is absorbed dose, 8766 is the number of hours per year.

Another radiation hazard index is called representative level index I_{γ} , is defined by the following formula (UNSCEAR, 2000):

$$I_{\gamma} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_{K}}{3000}$$
(10)

where A_{Ra} , A_{Th} and A_{K} are the specific activities of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively, in Bq kg⁻¹. I \leq 1 corresponds to a dose creation of 1 mSv/y.

RESULTS AND DISCUSSION

1. Specific Activity

The activity concentrations of the 226 Ra, and 214 Pb at energies (186 and 609.31) keV as equivalent to the activity concentration of 238 U by selecting the most valuable activity, the activity concentration of ²²⁸Ac isotope at energy (911.1) keV as equivalent to the activity concentration of 232 Th, the activity concentration of 40 K at energy (1460.83) keV, and the activity concentration of ¹³⁷Cs at energy (661) keV was determined (De Corte, 2005).
Specific concentration (Bq/kg) for ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs in sediment samples are listed

in (Table 1) and Fig. (2).

The specific activity levels for 226 Ra in sediment samples recorded lowest value 8.73 Bq kg⁻¹ in sample B1 (Bishkan village water project (Al-Asala)), and the highest value 84.68 Bq kg⁻¹ in sample B18 (Dhuluiya-the first sand island) with average 14.49 Bq kg⁻¹. The average level of radioactivity of ²³⁸U nuclide in sediment samples are well below the recommended global limits 35Bq/kg (UNSCEAR, 2000).

The specific activity levels for 232 Th recorded lowest value for 228 Ac was 1.47 Bq kg⁻¹ in sample B6 (Dhuluiya-Najm Island 1), and the highest value 49.8416Bqkg⁻¹ in sample B23 (Dhuluiya village Bishkan 4) with average 8.36 Bq kg⁻¹. The average level of radioactivity of ²³²Th nuclide in sediment samples are well below the recommended global limits 30Bq/kg (UNSCEAR, 2000).

The specific activity levels for ⁴⁰K recorded lowest value 56.51Bq kg⁻¹ in sample B12 (Dhuluiya on the one hand Yathrib 2), and the highest value 711.34 Bq kg⁻¹ in sample B7 (Dhuluiya, Sandy Island 2) with average 418.47Bq kg⁻¹. The average level of radioactivity of ⁴⁰K nuclide in sediment samples is higher than the recommended global limits 400Bq/kg (UNSCEAR, 2000).

The specific activity levels for artificial nuclide ¹³⁷Cs recorded lowest value 0.84 Bq.kg⁻¹ in sample B16 (Dhuluiya-the third island) and the highest value 9.78Bq/kg in sample B2 (Dhuluiyavillage of Bishkan-Nahrawan) with average 2.88Bq kg⁻¹. The average level of radioactivity of ¹³⁷Cs nuclide in sediment samples are well below the recommended global limits 14.8Bq/kg (UNSCEAR, 2000).

Fig. (1), shows the average specific activities of 226 Ra, 228 Ac and 137 Cs (Bq/kg) in sediment samples of Tigris river in Al-Duluiya city which are below the recommended global levels except for 40 K radionuclides.

The specific activities of ²²⁶Ra, ²²⁸Ac and ⁴⁰K in sediment samples from the studied areas was compared with other countries and summary results were given in (Table 2). The comparison shows that the values of natural radioactivity are extremely low in accordance with others. It is found that the mean value of ²²⁶Ra and Ac in the present study were lower than reported for of Oman, Jordan, Iran, Nigeria, Turkey, India, Eygpt, Thi-Qar and Wassit governorates. But for ⁴⁰K is higher than reported for Oman, Jordan, Syria, Nigeria and India and lower than reported for Iran and Turkey, Eygpt, Thi-Qar and Wassit governorates.

The variations of the specific activity of the various country of the world, depend on the geological and geographical conditions of the area (UNSCEAR, 2000).

Table 1: Specific activates of ²²⁶Ra, ²³²Th, ⁴⁰K and ¹³⁷Cs radionuclides in sediments samples of Tigris River of Dhuluiya City

Samples	Specific Activity (Bq/kg)					
Code	²²⁶ Ra	$\frac{228}{\text{Ac}} = \frac{40}{\text{K}}$		¹³⁷ Cs		
B1	8.73	16.58	247.45	2.77		
B2	1.51	10.43	436.07	9.78		
B3	8.07	2.11	208.44	MDA		
B4	31.99	3.15	538.45	MDA		
B5	21.4	12.39 597.66		MDA		
B6	MDA	1.47	575.41	4.73		
B7	20.12	5.16	5.16 711.34			
B8	3.49	9.80	9.80 646.00			
B9	17.95	MDA	375.01	9.59		
B10	9.67	3.34	506.43	2.70		
B11	MDA	MDA	366.85	MDA		
B12	MDA	MDA	56.51	MDA		
B13	MDA	MDA	341.93	MDA		
B14	MDA	13.44	237.56	MDA		
B15	MDA	8.82	444.27	MDA		
B16	MDA	2.32	253.65	0.84		
B17	24.57	MDA	444.82	MDA		
B18	84.68	MDA	600.17	MDA		
B19	50.02	MDA	604.76	9.39		
B20	38.24	6.90	343.90	3.82		
B21	25.99	46.64	125.57	3.77		
B22	MDA	MDA	520.79	8.91		
B23	9.62	49.84 441.84		6.25		
Ave	15.48	8.36	418.47	2.88		
W.Ave	35	30	400	14.8		

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Fig. 1: Average Specific Activity of ²²⁶Ra, ²²⁸Ac,⁴⁰K and ¹³⁷Cs (Bq/kg) in sediment samples for various locations in the study area.

Table 2: Comparison of Natural Radioactivity Specific Activities (Bq·kg ⁻¹) for Previous Stud for Different Countries of the World and Present Study							
Fable 2: Comparison of Natural Radioactivity Specific Activities (Bq·kg ⁻¹) for Previous States for Different Countries of the World and Present Study Country Mean activity concentration (Packg ⁻¹)							

Country	Mean activity concentration $(Bq \cdot kg^{-1})$				
	²²⁶ Ra	²³² Ac	⁴⁰ K	Reference	
Oman	29.7	15.9	225	(Goddard, 2001)	
Jordan	49	27	291	(Al-Hamarneh, et al.,	
				2009)	
Syria	23	20	270	(UNSCEAR, 2000)	
Iran	28	22	640	(UNSCEAR, 2000)	
Nigeria	7.8	29.4	229	(Kolo et al., 2012)	
Turkey	37	40	667	[20]	
India	41	29	400	(UNSCEAR, 2000)	
Eygpt, Aswan	14.86	13.78	175.4	(Atef El-Taher et al.,	
governorate				2018)	
Eygpt, Qena	14.44	15.02	197.57	(Atef El-Taher et al.,	
governorate				2018)	
Iraq, Thi-Qar	29.2	22.7	304.6	(Laith Najam et al.,	
Governorate				2016)	
Iraq, Wassit	19.420	18.487	204.266	(Laith Najam et al.,	
governorate				2017)	
Salah-Al-Din	15.48	8.36	418	Present Study	
Governorate,					
Dhuluiya City					

2. Assessment of Radiological Parameters

The radiological parameter or radiological hazards indices were calculated as shown in (Table 3). The radium equivalent Ra_{eq} (Bq/kg) which recorded the lowest value 4.35 in B11 location and highest value of 130.89 in B18 location with average of 56.38 Bq/kg. The average of radium equivalent of sediments of the Tigris River for Dhuluiya city of Salah Uddin Governorate is less than the internationally accepted limit value 370 Bq /kg as shown in Fig. (2). The absorbed dose rate in air D γ (nGy/h) recorded lowest value 2.31 in B12 location and the highest value of 64.15 in B18 location with average 27.76 nGy/h.

The risk index value I_{γ} (Bq /kg) recorded lowest value 0.04 in B12 and the highest value 0.96 in B18 with average 0.42 Bq/kg.

The internal hazard index H_{in} , (Bq.kg⁻¹) recorded lowest value 0.01 in B11 location and the highest value of 0.58 in B17 location with average 0.20 Bq kg⁻¹. The external hazard index for

sediment samples H_{ex} (Bq.kg⁻¹) recorded lowest value 0.01 in B12 location and the highest value 0.35 in B18 location with average 0.16 Bq kg⁻¹.

The annual effective dose equivalent indoor $AEDE_{in}$ (mSv/y), recorded lowest value 0.11 in B11 location and the highest value 0.59 in B15 location with average 0.16 mSv/y. The annual effective dose equivalent outdoor $AEDE_{out}$ (mSv/y), recorded lowest value 0.01 in B17 location and the highest value 0.08 in B19 location with average 0.04 mSv/y. The average values of the radiological hazard effects rate for gamma ray in Tigris river in Al-Duluiya city sediment samples, Salah Al-Din governorate were all lower than the value of the internationally accepted limit (UNSCEAR, 2000).

Sample Code	Ra _{eq} (Bq/kg)	D (nGy/h)	Hazard Indices (Bq/kg)		AEDE (mSv/y)		Ιγ (Ba/kg)
	1 8/		H _{ex}	H _{in}	AEDE _{in}	AEDEout	T 8/
B1	51.49	24.64	0.14	0.16	0.12	0.03	0.25
B2	49.74	25.24	0.13	0.14	0.12	0.031	0.20
B3	27.13	13.73	0.07	0.3	0.19	0.02	0.21
B4	77.96	39.19	0.210	0.29	0.21	0.05	0.41
B5	85.14	42.50	0.23	0.13	0.12	0.05	0.67
B6	46.4	24.91	0.13	0.27	0.19	0.03	0.4
B7	82.27	39.35	0.22	0.19	0.17	0.05	0.66
B8	53.22	34.64	0.18	0.17	0.12	0.04	0.43
B9	46.82	23.93	0.13	0.17	0.14	0.03	0.37
B10	42.40	27.66	0.14	0.08	0.08	0.03	0.44
B11	4.35	15.3	0.08	0.01	0.01	0.02	0.24
B12	26.33	2.36	0.01	0.07	0.07	0.03	0.04
B13	53.45	14.26	0.07	0.10	0.09	0.02	0.23
B14	37.51	18.25	0.10	0.13	0.03	0.02	0.29
B15	46.82	24.0	0.13	0.06	0.59	0.07	0.38
B16	22.85	12.92	0.06	0.23	0.15	0.07	0.19
B17	58.82	29.90	0.16	0.58	0.03	0.01	0.46
B18	130.89	64.15	0.35	0.4	0.24	0.04	0.96
B19	28.37	48.33	0.26	0.30	0.11	0.08	0.74
B20	74.59	21.95	0.20	0.35	0.23	0.06	0.55
B21	102.36	46.21	0.28	0.11	0.11	0.03	0.72
B22	40.10	21.72	0.11	0.34	0.64	0.06	0.35
B23	107.76	23.35	0.31	0.1	0.07	0.03	0.36
Max	130.89	64.15	0.353	0.58	0.59	0.08	0.96
Min	4.35	2.36	0.011	0.01	0.01	0.01	0.04
Ave	56.38	27.76	0.16	0.20	0.16	0.04	0.41
W.Ave	370	84	1	1	1	1	1

Table 3: Radiological Hazards Indices in Sediments of Tigris River of Dhuluiya City



Fig. 2: Radium Equivalent Activities (Bq/kg) for various Locations in the Study area

CONCLUSION

In the present study, the specific activity of radionuclides, ²²⁶Ra, ²²⁸Ac, ¹³⁷Cs are less than the world average values except for ⁴⁰K. And the specific activates variations in the sediments samples of Tigris river at various locations in Dhuluiya city of Salah Uddin Governorate depend on the geographical and geological of the area.

Radiological hazards indices in sediments of Tigris River of Dhuluiya city is less than the world average values. None of the site considered is a radioactive hazard of sediment that can be safely used in agriculture and construction.

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تقييم النشاط الإشعاعي الطبيعي والصناعي ومؤشرات الخطورة الإشعاعية في رواسب نهر دجلة بمدينة الضلوعية، العراق الملخص

تم قياس النشاط النوعي للنويدات المشعة الطبيعية والصناعية في عينات الرواسب التي تم جمعها من نهر دجلة بمدينة الضلعية بمحافظة صلاح الدين باستخدام التحليل الطيفي الشعاعي، كاشف (NaI TI). أوضحت النتائج أن الفعالية النوعية ل ²²⁶Ra تراوحت بين 1.51 بيكريل/كغم إلى 84.68 بيكريل/كغم بمعدل 15.48 بيكريل/كغم، ²²⁸Ac تراوحت من 1.47 إلى 49.84 بيكريل/كغم بمعدل 8.36 بيكريل/كغم، ⁴⁰K تراوحت من 15.51 بيكريل/كغم، 1.32 بيكريل/ كغم بمعدل 49.84 بيكريل/كغم و ¹³⁷Cs تراوحت بين 2.77 بيكريل/كغم إلى 56.51 بيكريل/كغم بمعدل 49.84 بيكريل/كغم، معدل معدل 49.84 بيكريل/كغم و ¹³⁷Cs مو د كان متوسط متوكيزات هذه النويدات المشعة أقل من الحد المقبول دوليًا باستثناء النويدات المشعة ⁴⁰K. تم حساب مؤشرات الخطورة الإشعاعية، مكافئ للراديوم (Ra_{eq})، معدل جرعة كاما الممتصة (Dγ)، الجرعة الفعالة الخارجية والداخلية السنوية (AEDE)، المخاطر الخارجية والداخلية (H_{in} ، H_{ex})، وركت

الكلمات الدالة: النشاط الإشعاعي الطبيعي، مطيافية أشعة جاما، كاشف ايوديد الصوديوم المنشط بالثاليوم (NaI(Tl)، مؤشرات الخطورة الإشعاعية.