Al-Noor Journal Oil and Gas Studies, Vol 2, No. 1, 2026 (33-40)
ISSN: 3078-8706 Doi https://doi.org/10.69513/jnog.v2.i1.ar6 .

Al-Noor Journal for Oil and Gas Studies

https://jnog.alnoor.edu.iq/

Radiological Assessment of Soil Contamination by NORM in Ain Zalah Oil Field, Nineveh, Iraq

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Article information

Article history:
Received 23 September 2025
Revised 30 December 2025
Accepted 25 January 2026

Keywords:
NORM,
Ain Zalah
oil field,
Soil,
Activity
concentration,
radiological haza

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Abstract

The activity concentrations and radiological hazard parameters (equivalent activity of radium Ra_eq, absorbed dose rate D, annual effective dose AEDE, external and internal hazard indices H, Gamma radiation representative level index I_γ, Alpha radioactivity level index (I_α), and excess lifetime cancer risk ELCR) of the terrestrial Naturally Occurring Radioactive Materials NORM such as (_ ^226)Ra,(_ ^232)Th and (_ ^40)K were assessed in soil samples collected from Ain Zalah oil field, Nineveh, Iraq, using HPGe detector. The analysis of NORM gives obtainable data for rules about radiation protection. The results were depicted in graphical and tabular formats and subsequently compared to the established international permissible thresholds set by UNSCEAR. Consequently, it is possible that the petroleum extraction activities in the studied area could result in some radiological risks for both the workers and the surrounding environment due to the elevated activity concentration of radionuclides.

Keywords:

DOI: https://doi.org/10.69513/jnog.v2.i1.ar6, ©Authors, 2026, College of Engineering, Alnoor University.

1-Introduction

The naturally occurring radionuclides present in soil and rocks are not distributed uniformly around the world; their concentrations depend mainly on local geological and geographical conditions. Cosmic radiation is largely influenced by radionuclides such as 40K, and by members of the 238U and 232Th decay series, which are present in the Earth's crust. In this study, the ELCR values exceeded the internationally recommended annual effective dose limits. (1,2). Natural occurring radioactive materials

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(NORM) expose people to a variety of radiation sources (3). Because of the body's exposure to gamma rays and the lung tissues' exposure to radiation from inhaled radon and its daughters, it is well known that even small amounts of these radionuclides can cause detrimental biological effects. (4) Accurate measurement of radiation from naturally occurring radioactive materials in land, air, water, food, and buildings is essential to estimate human exposure and implement protective measures (5). Since soils are the main source of radioactivity, the natural radioactivity mentioned usually arises from the presence of the 238U, 232Th and 40K. Therefore, determining the radioactive content of soils worldwide is essential for assessing health risks (6).

Estimating the level of natural radioactivity in soils is crucial to assessing the rate of terrestrial gamma dose for outdoor employment (7,8). Soil is a complex mixture of various compounds and rocks. It is an important source of gamma radiation exposure (9).

The levels of radioactivity dispersion in the soil, oil, and gas field environments have been the focus of radiation research on soil and sediment samples from different parts of the world. This information will be crucial and additional radiological data for the global radiation database.

In this study, the natural radioactivity levels of the soil at the Ain Zalah oil field in Iraq were evaluated. Additionally, radiological hazard indices such as the radium equivalent activity (Ra_eq), absorbed dose rate (D), external hazard (Hex), and internal hazard (Hin)were calculated. The annual effective dose rate (AEDE), excess lifetime cancer risk (ELCR), gamma radiation representative level index (Iγ), and alpha radioactivity level index (Iα) were also calculated. The information produced by this study will serve as a baseline for the level of natural radioactivity in the study area and will be valuable to those responsible for enforcing radiation protection regulations for the oil industry and the general populace in host communities. The results will be compared to values reported in the literature.

Materials and methods

The Ain Zalah oil field is located in the Nineveh Governorate of northern Iraq, approximately 50 kilometers south of the city of Mosul (Fig. 1) and its geographical coordinates are N 42^° 35^' 38^" E 20^° 43^' 36^" with an elevation about 420 m. It is one of the largest oil fields in Iraq, with estimated reserves of 3.6 billion barrels of oil.

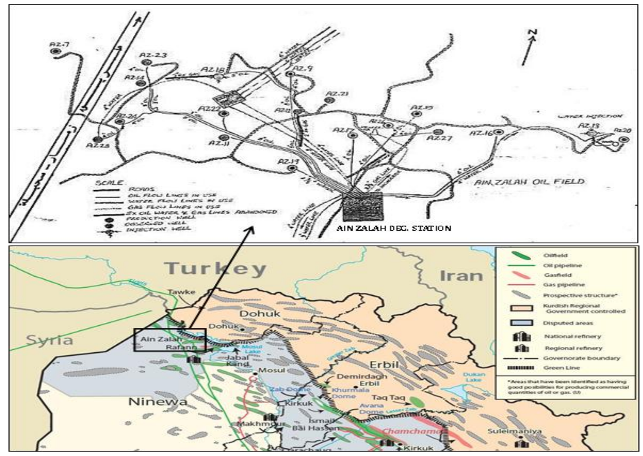

Figure 1. The location of the Ain Zalah oil field is depicted in Fig. 1, which provides a geographical representation of the field.

2.1 Sample collection and preparation techniques

Fifteen soil samples are collected from areas around oil wells, originating from the drilling process, pipelines, and storage facilities. The samples were dried for 4 h at 110 °C in an oven to reach a constant dry weight, and subsequently sieved to obtain equal particle sizes using a 1mm sieve. The dried samples are next crushed with a mortar and pestle or a ball mill into a fine powder. A plastic container (typically a Marinelli beaker for calibration) was used to pack 500 g of each soil sample, which was then sealed tightly. Finding the activity content of NORM is done using an HPGe detector. The containers were sealed and held in storage for a month to establish the secular equilibrium between the parent radionuclides and their respective daughters, specifically between 226Ra and 222Rn, and 214Pb and 214Bi. The containers were sealed and kept in storage for a month. Using gamma-ray spectrometry and a computer with a program installed for data collection and gamma-ray spectrum analysis, the samples were examined non-destructively. Using sealed point sources holding 1μ Ci each of 22Na, 60Co, and 137Cs, the measuring apparatus was calibrated. Each radionuclide's activity concentration (A) was calculated using the following equation:

$$A(Bq.Kg^{-1})=C/(\epsilon.I.W) \quad (1)$$

where C is the detected radionuclide's full-energy peak count rate (measured in counts per second), ϵ is the detection efficiency for that energy, I is the corresponding gamma-ray yield, and W is the sample's mass, measured in kilograms.

Measurements of their respective decay daughters were used to determine the activities. 214Bi at 609keV (45%) for 226Ra, 208Tl at 2614.7 keV (36%) for the 232Th-series while 40K activity direct assessment of its emission at a given concentration 1461 keV (10.7%). To calculate the background distribution spectrum, the sample and an empty container with the same geometry were both counted for the same amount of time.

2.2 Radiation hazardous indices

The total quantity of these activities has been calculated using the radium equivalent (Ra-eq) index in order to compare the activity concentration of materials containing 226Ra, 232Th, and 40K(5). The Ra-eq index has been created from the assumption that the amount of gamma rays emitted by 370 Bq/kg of 226Ra, 259 Bq/kg of 232Th, and 4810 Bq/kg of 40K are equivalent. As a result, the following is how a sample's Ra-eq can be expressed:

〖 Ra eq=C_Ra+1.43C _Th +0.077C_K (2)

where CRa, CTh, and CK are, respectively, the activity of 226Ra (238U-series), 208Tl (232Th-series), and 40K in Bq/kg.

It is well known that personnel working with materials containing radionuclides of 226Ra, 232Th, and 40K shouldn't be subjected to total absorbed dose rates (D) higher than the norm for the world 0.1314 μSv/h [1]. Therefore, Dγ in outdoor air at 1 m above the ground was calculated as follows [3].

D(μSv.h^(-1))=(0.462C_Ra+0.604C_Th+0.0417C_K)*0.001*0.7+0.0371 (3)

As 0.7 Sv. Gy-1 is the conversion coefficient from absorbed dose in air to effective dose. The number 0.0371 μSv/h is a factor included to take care of the contribution from cosmic radiation of our studied region which calculated using Excel-based Program for calculating Atmospheric Cosmic-ray Spectrum (EXPACS ver. 2.27) depending on latitude, longitude and height from sea level.

The External hazard index is a popular hazard metric that reflects external exposure. Here is how hex is defined. [3].

H_ex=C_Ra/370+C_Th/259+C_K/4810 ≤1 (4)

Along with the external hazard index, radon and its transient compounds are harmful to the respiratory system. The internal hazard index Hin, which is determined by the equation, measures how much radon and its offspring are exposed internally. [2] [3]

H_in=C_Ra/185+C_Th/259+C_K/4810≤1 (5)

Indicator of the representative level Iγ is a different radiation hazard index that is applied to determine the gamma radiation related to radioactive materials that are found naturally in soil. [3]

I_γ=C_Ra/150+C_Th/100+C_K/1500 ≤ 1 (6)

For the radiation risk to be insignificant, the values of the indices (Hex, Hin, and Iγ) must be less than unity. [3].

There are several indices (sometimes known as "alpha indexes" or "internal indexes") that are used to measure the excess radiation from radon inhalation that comes from building materials. The following formula was used to determine the alpha indexes:

I_α=C_Ra/(200Bq. 〖 Kg 〗 ^(-1)) (7)

where CRa stands for the samples' actual 226Ra concentration (measured in Bq kg-1). When this substance's value surpasses 200 Bq kg-1, it is likely that radon exhalation from it will result in indoor radon concentrations that are higher than 200 Bq m-3. In contrast, it is improbable that radon exhalation from building materials will result in radon levels in dwellings that are higher than 200 Bq m-3 when its value is below 100 Bq kg-1. [7].

Annual effective dose equivalent (AEDE)

The yearly effective dose equivalent (mSv y(-1)) was estimated from the absorbed dose by using the dose conversion ratio of 0.7 Sv/Gy with an occupancy factor of 0.2 for outdoor and occupancy factor of 0.8 for indoor environments. However, the average annual effective dose due to gamma-radiation from these terrestrial sources can be assessed by using the following equation [10], [33]:

AEDE_out (mSv y^(-1))=D (nGy h^(-1))×8760 (h y^(-1))×0.7 ((10^3 mSv)/(10^9 nGy))×0.2 (8)

AEDE_in (mSv y^(-1))=D (nGy h^(-1))×8760 (h y^(-1))×0.7 ((10^3 mSv)/(10^9 nGy))×0.8 (9)

where D stands for dose rate and 8760 hours make up a year. The global values for AEDE_out and AEDE_in are 0.08 mSv and 0.42 mSv, respectively. [2] [33].

Excess lifetime cancer risk (ELCR)

Exposure to ionizing radiation over a person's lifetime can result in an increased risk of getting cancer, which is referred to as excess lifetime cancer risk (ELCR). The equation used to estimate the ELCR from annual effective dose equivalent in the soil studied is as follows [1] [2]:

ELCR=AEDE×DL×RF (10)

where AEDE is the annual effective dose equivalent, DL is the duration of life (70 years), and RF is the risk factor (fatal cancer risk per sievert). Ravisankar et al. [33] defined RF as the fatal cancer risk per Sv; according to ICRP guidelines for stochastic effects in the general public, this value is typically taken as 0.057 Sv^{-1} (some sources use 0.05 Sv^{-1}). Specify which value you adopt and cite the source.

3- Result and discussion

3.1 Activity concentrations

The contour map in Fig. 2 provides a visualization of the distribution and a valuable tool for identifying areas of radium activity concentrations in the study area. The map uses contour lines to indicate areas of equal radium activity concentration, with the spacing between the lines indicating the rate of change in concentration. The map shows that radium activity concentrations are not uniformly distributed, with some areas exhibiting higher concentrations than others.

The high levels of radium activity concentration observed in the study area are attributed to a combination of factors, including local geology and soil composition. In addition, mining and industrial processes can also contribute to elevated radium levels in soil and water.

Figure 3 represents the radium activity concentration in Ain Zalah oil field samples, and it is clear, that the activity concentration of (_ ^226)Ra varied from 12.51 〖Bq Kg〗 ^(-1) to 120.41 〖Bq Kg〗 ^(-1) with an average 69.1 〖Bq Kg〗 ^(-1). The values of all samples were higher than the global average value of 35 〖Bq Kg〗 ^(-1), except AZ10 sample which has the lowest one with value evaluated about 12.51 〖Bq Kg〗 ^(-1). and compare them with the worldwide average, it is show that the average value of samples is higher than approximately twice of worldwide average due to the presence of NORMs, which can accumulate in the soil and other materials that can become contaminated with NORMs, including radium-226,

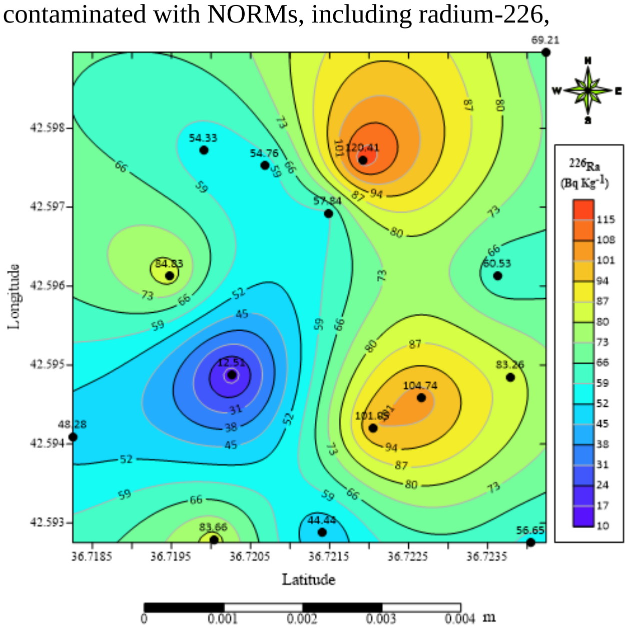

Figure 2. Contour Map of Radium-226 Activty Concenteration in Ain Zalah oil field.

The activity concentration of ^{232}Th was measured between 11.19 and 60.28 Bq Kg $^{-1}$, with an average of 29.34 Bq Kg $^{-1}$. The AZ10 sample had the lowest value, while sample AZ3 produced the highest value. Depending on the separation from the oil wells, the variations were considerable in all samples. A range of 152.46 to 475.73 Bq Kg $^{-1}$, with an average of 323.99 Bq Kg $^{-1}$, was recorded for the activity concentration of ^{40}K. The lowest value was found in AZ7, while the highest value was found in AZ9. The variances were also due to the different soil types in the area under research, in addition to the industrial oil waste. A contour map of the sample locations and concentrations for the research areas is also shown in Figures 4 and 6.

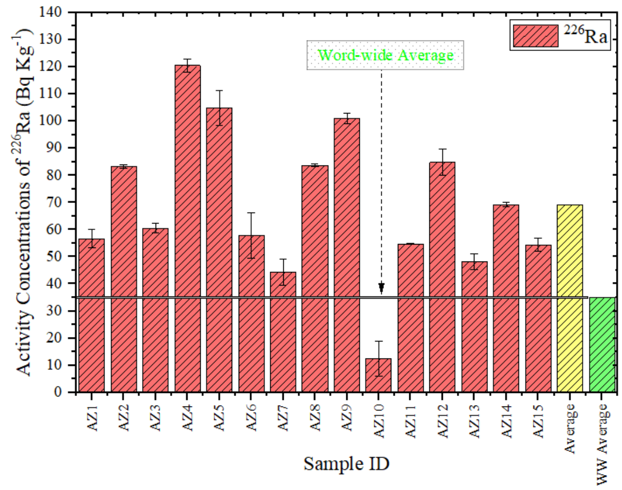

Figure 3. Column Graph of Radium-226 Activity Concenteration in Ain Zalah oil field.

In Fig. 5 the data of Thorium activity concentration samples were visually represented and compared with the worldwide average value 30 〖Bq Kg〗 ^(-1), it is show that the average value of samples is a little bit lower than the worldwide average value due to the presence of NORMs. The samples that have a high activity concentration of Thorium in respect to worldwide average were AZ2, AZ3, AZ5 and AZ8 with mentioned that the highest one was AZ3 sample.

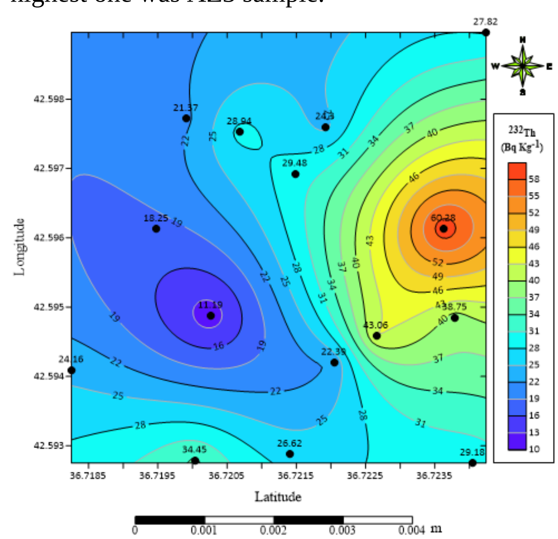

Figure 4. Contour Map of Thorium 232 Activity Concentration in Ain Zalah oil field

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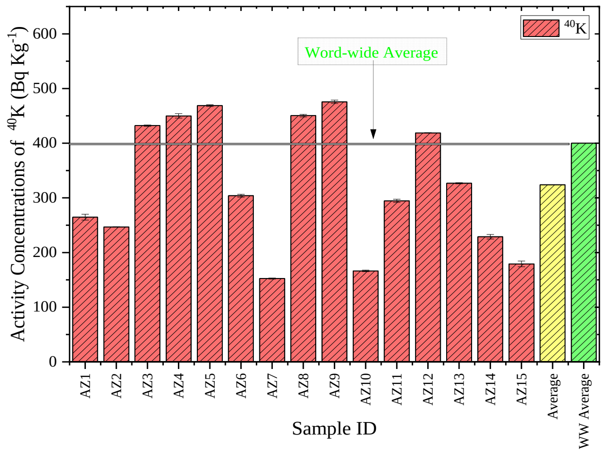

Figure5: Column Graph of Thorium-232 Activity Concentration in Ain Zalah oil field.

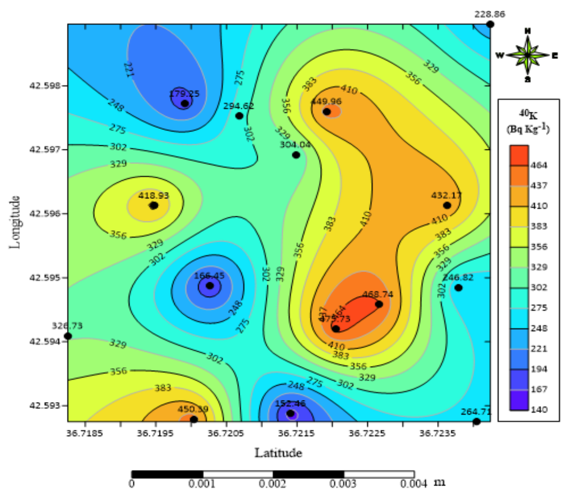

Figure 6. Counter Map of Potassium-40 Activity Concentration in Ain Zalah oil field

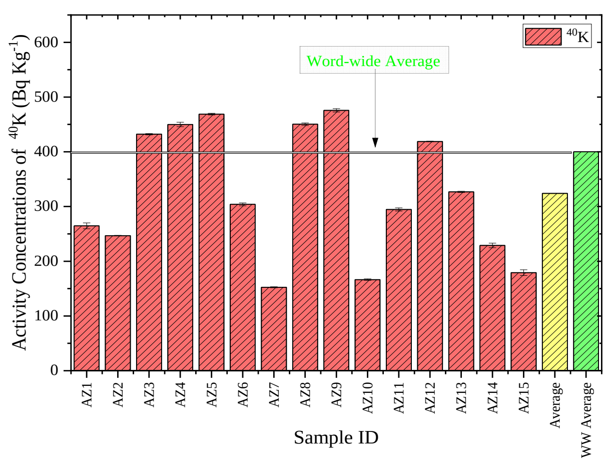

Figure 7: Column Graph of Potassium-40 Activity Concentration in Ain Zalah oil field

In the study, three soil samples (AZ3, AZ5, and AZ8) were tested for the presence of naturally occurring radioactive materials (NORMs), specifically Radium-226, Thorium-232, and Potassium-40. The results of the analysis were presented in Figures 3, 5, and 7, which show that all three samples contained detectable levels of these NORMs.

The radioactive element is apparently radium-22, which is found largely in the Earth's crust. It is the result of the decay of uranium 238, which is also present in the Earth's crust. The other element, thorium 232, is naturally concentrated in soil, rocks and minerals. As for potassium 40, it is found in all types of soil and rocks in small concentrations.

These limits found in the soil samples indicate that the area may be contaminated with naturally occurring radioactive materials. The sources of this pollution can be natural or artificial. Natural materials are produced naturally through the decomposition of uranium and thorium in rocks and soil, while anthropogenic sources include activities such as mining and oil and gas exploration. We have verified and examined the data obtainable from published works. And matching the concentrations of soil samples studied in the research plan with previous studies.

Valuable information about radionuclide levels and their distribution in the study area and how they may change over time can be obtained by comparing measured concentrations with those reported in the literature.

Through comparison we note that the concentrations of radionuclide activity in our soil samples were generally consistent with those reported in previous studies.

However, some variations were observed, which may be attributed to differences in the sampling techniques, analytical methods, and the natural variability of radionuclide concentrations in soil.

According to the Organization for Economic Cooperation and Development's recommendation [39], the maximum value of Ra_eq in soil needs to be less than 370 Bq Kg^{-1} to be used safely. Table 2 displays the computed radium equivalent activity for the various soil samples seen in this investigation. The radium equivalent activities in this investigation had the following lowest, maximum, and mean values <370 Bq Kg^{-1}.

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The population-weighted figures result in an outdoor absorbed dose rate from gamma radiation from the earth being equal to 57 nGy h^{-1}.

It is noted that doses out of 11 sites are greater than 57 out of a total of 15, more than the permissible limits globally, and this high dose causes health risks or disease.

The H_{ex}, H_{in}, I_{α} is less than one for all sites and is less than the minimum limits

The I_{γ} values for seven sites are greater than 1, therefore, these values are greater than the internationally permitted values. This is due to the presence of radioactive elements in the soil that emit gamma rays, namely radium and uranium, which at high doses pose health risks and disease.

It can be noted from the results presented in Table 2 that the values obtained for $AEDE_{out}$ and $AEDE_{in}$ range from 0.023-0.11mSv y^{-1}. It should be noted that a number of sites have values exceeds internationally permissible values due to excavation of clay soils and crust sediments generated during the extraction process; therefore, it is important to properly manage and dispose of waste to minimize potential risks.

Table 1. An evaluation of radioactivity satisfied from other amounts of the world.

Location	Commotion concentration (**Bq Kg^{-1}**)			Ref.
	^{226}Ra	^{232}Th	^{40}K	
Iraq (Kirkuk Oil Field-Baba Gurgur)	26.02	13.75	288.89	[10]
Iraq (Kirkuk Oil Field-Shurau)	24.38	18.29	356.5	
Iraq (Kirkuk Oil Field-Terkiz)	34.80	18.80	289.20	
Iraq (Kirkuk Oil Field-Baba Gurgur dome)	57.8	25.4	479.9	[11]
Iraq (Badra Oil Field)	24.7	13.6	538.9	[12]
Iraq (South Rumaila Oil Field)	8.4 –312.8	9.4–140.8	66.4–800.8	[13]
Iraq (Basra Oil Field)	33.59	20.65	511.6	[14]
Qatar (Dukhan Oil Field)	20.05	16.43	216.99	[15]
Louisiana (USA)	43 - 95	50 - 190	43.729	[17]
Spain	46	49	650	[18]
China	42.7∓15	46.3∓12	578∓164	[19]
Cyprus	7.1∓0.6	5.0∓0.7	105∓95	[20]
Kenya	28.7∓3.6	73.3∓9.1	255.7∓38.5	[21]
Republic of Ireland	60	26	350	[22]
Egypt	930	11.6	21.1	[23]
Australia	11.42	19.26	641.08	[24]
Korea	22.6	26.4	523	[25]
Sudan	28.31	20.12	280.29	[26]
Malaysia	31	36	369	[27]
Palestine	41	19	113	[28]
Pakistan	31	44	575	[29]
Nigeria	25	77	710	[30]
Turkey	157.7	9	12.2	[31]
Saudi Arabia	11.3	6.7	153.8	[32]
World (average)	35	30	400	[2]
The current study	69.1	29.34	323.99	

.2 Radiological effects

Table 2. Radiological hazard indices of the examined soil samples show potential health concerns associated with exposure to environmental radioactivity.

Sample ID	Ra_{eq} ($Bq\ Kg^{-1}$)	D_{out} ($nGy\ h^{-1}$)	H_{ex}	H_{in}	I_{γ}	I_{α}	$AEDE_{out}$ ($mSv\ y^{-1}$)	$AEDE_{in}$ ($mSv\ y^{-1}$)	ELCR ($\times 10^{-3}$)
AZ1	118.76	54.83	0.32	0.47	0.845	0.28	0.067	0.26	0.268
AZ2	157.67	72.16	0.42	0.65	1.107	0.41	0.088	0.35	0.353
AZ3	180.0	82.39	0.48	0.64	1.294	0.30	0.101	0.40	0.403
AZ4	189.80	89.06	0.51	0.83	1.345	0.60	0.109	0.43	0.435
AZ5	202.40	93.94	0.54	0.82	1.441	0.52	0.115	0.46	0.459
AZ6	123.40	57.20	0.33	0.48	0.883	0.28	0.070	0.28	0.279
AZ7	94.24	42.96	0.25	0.37	0.664	0.22	0.052	0.21	0.210
AZ8	167.60	78.23	0.452	0.67	1.202	0.41	0.095	0.38	0.382
AZ9	169.69	80.04	0.458	0.73	1.214	0.50	0.098	0.39	0.391
AZ10	41.32	19.47	0.11	0.14	0.306	0.06	0.023	0.09	0.095
AZ11	118.82	55.06	0.32	0.46	0.850	0.27	0.067	0.27	0.269
AZ12	143.18	67.68	0.38	0.61	1.027	0.42	0.083	0.33	0.331
AZ13	107.98	50.52	0.29	0.42	0.781	0.24	0.061	0.24	0.247
AZ14	126.61	58.32	0.34	0.52	0.892	0.34	0.071	0.28	0.285
AZ15	98.69	45.48	0.26	0.41	0.695	0.27	0.055	0.22	0.222
Range Min	41.32	19.47	0.11	0.306	0.306	0.06	0.023	0.09	0.095
Range Max	202.40	93.94	0.54	0.83	1.441	0.60	0.11	0.46	0.459
Average	136.01	63.16	0.36	0.55	0.970	0.34	0.077	0.309	0.309
World Average [11]	370	57	≤ 1	≤ 1	≤ 1	≤ 1	0.08	0.42	0.29

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التقييم الإشعاعي لتلوث التربة بواسطة NORM في حقل عين زالة النفطي، نينوى، العراق

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تم تحديد تركيز النشاط ومعايير الخطر الإشعاعي (النشاط المكافئ للراديوم، Raeq ، ومعدل الجرعة الممتصة، D، والجرعة الفعالة السنوية، AEDE، الخارجية والداخلية. مؤشرات المخاطر الداخلية، ومؤشر مستوى تمثيل إشعاع جاما، Iγ، ومؤشر مستوى النشاط الإشعاعي ألفا Iα، وخطر الإصابة بالسرطان الزائد مدى الحياة، ELCR) الخاص بالمعيار الأرضي للمواد المشعة الطبيعية، مثل Ra226 تم تقييم Th232 و K40 في عينات التربة التي تم جمعها من حقل عين زالة النفطي في نينوى، العراق، باستخدام كاشف HPGe. يوفر تحليل NORM بيانات يمكن الوصول إليها لأنظمة الحماية من الإشعاع. وقد عُرضت النتائج بيانياً وبشكل مجدول ، ثم تمت مقارنتها مع الحدود المقبولة دولياً التي حددتها لجنة الأمم المتحدة العلمية المعنية بآثار الإشعاع الذري (UNSCEAR). وبالتالي، هناك احتمال أن تؤدي عمليات استخراج النفط في المنطقة التي تم فحصها إلى إثارة بعض المخاوف الإشعاعية.