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Radionuclides Determination and Hazard Indices Assessment of NORM Contaminated Soil for Oil Fields in Southern Iraq

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Abstract

Oil and gas extraction greatly impacts the environment due to the existence of natural radionuclides in crude oil. Uncontrolled dealing with the extraction can lead to radiological contamination. This research aims to determine the radioactivity concentration of naturally occurring radioactive materials (NORM) and risk assessment of contaminated soil in oil fields. Three oil fields in Al- Basra government southern of Iraq were investigated; 45 soil samples were collected from several locations, the first group of soil samples from Al Rumaila southern oil field, the second group was collected from Khudair Almay storage site, and the third group collected from a waste pit near an oil-well in Majnoon oil field. High purity germanium (HPGe) detector was used to determine the radioactivity concentrations of radionuclides of the series ²³⁸U and ²³²Th as well as ⁴⁰K. The mean concentration of ²²⁶Ra, ²²⁸Ra, and ⁴⁰K was found in the soil samples of the first group: $5.625 \times 10^4 \pm 1.5\%$, $5.157 \times 10^3 \pm 1.43\%$, and $1.744 \times 10^3 \pm 5.7\%$ Bq kg⁻¹, respectively, $1.07 \times 10^5 \pm 1.5\%$, $7.90 \times 10^3 \pm 1.43\%$ and 8.33×10^2 \pm 5.7% Bq kg⁻¹, respectively for the second group and $4.85\times10^{2} \pm 5.7$ %, $5.5\times10^{1} \pm 9$ % and $1.65 \times 10^3 \pm 15.9\%$ Bq kg⁻¹, respectively for the third group.

The hazard indices, total radium equivalent, representative gamma level index, and annual effective dose equivalent were calculated to estimate the potential radiological health risk in these oil fields. The results fluctuate as acceptance in some samples of the third group and not acceptance in most of the first and the second group soil samples.

The statistical analyses of data show a high positive correlation coefficient (r= 0.996) with significant p-value ($p \le 0.05$) and coefficient of determination ($R^2 = 0.993$) between ²²⁶Ra and ²²⁸Ra in the first and second groups soil samples, whereas there is no correlation in the third group.

Keywords: NORM; radiological assessment; correlation coefficient; R²

تحديد تركيز النويدات المشعة وتقييم مؤشرات المخاطر للمواد المشعة طبيعية المنشأ)NORM **) للتربة الملوثة في حقول النفط جنوب العراق**

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الخالصة

استخراج النفط والغاز له تأثير كبير على البيئة بسبب وجود النويدات المشعة الطبيعية في النفط الخام ، ويمكن أن يؤدي التعامل غير المسيطر عليه عند االستخراج إلى التلوث أإلشعاعي. يهدف هذا البحث إلى تحديد تركيز النشاط اإلشعاعي للمواد المشعة طبيعية المنشأ)NORM)وتقييم مخاطر للتربة الملوثة في حقول النفط. تم

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فحص ثالثة حقول نفطية في محافظة البصرة جنوب العراق ، وتم جمع 45 عينة تربة من عدة مواقع ، المجموعة األولى عينات التربة من حقل الرميلة الجنوبي النفطي، المجموعة الثانية جمعت من موقع خضر الماي ، والمجموعة الثالثة تم جمعها من حفرة النفايات بالقرب من بئر نفط في حقل مجنون النفطي.

تم استخدام كاشف الجرمانيوم عالي النقاوة) HPGe)لتحديد تركيزات النشاط اإلشعاعي للنويدات المشعة للسلسلتين الرئيسيتين ل²³⁸ و ²³²Th وكذلك ⁴⁰K، حيث كان متوسط تركيز ²²⁶Ra و ²²⁸Ra في عينات $\%$ التربة من المجموعة الأولى 5.625×1.0 $\mathrm{^{4}10\times{5.625}}$ و $1.43\mathrm{\pm^{3}10\times{5.157}}$ و 1.43×1.744 $\%$ بيكريل/كغم على التوالي و 1.0710×1.5 %و 7.90 $^{10\times1.43}\pm310\times1.43$ و 3.3 $\times10\times25$ % بيكريل/كغم على التوالي للمجموعة الثانية و4.85×10 $\sim 5.7\pm 10\times 1.65$ % و 1.65×10 $^{-3}$ 10×1.65 % بيكريل/كغم على التوالي للمجموعة الثالثة.

تم حساب مؤشرات المخاطر ومكافئ الراديوم الإجمالي ومؤشر المستوى التمثيلي لأشعة كاما ومكافئ الجرعة الفعالة السنوية لتقدير المخاطر اإلشعاعية المحتملة في حقول النفط هذه. حيث تراوحت النتائج كمقبولة لبعض النماذج في المجموعة الثالثة ، وغير يتم مقبولة في معظم عينات التربة للمجموعة الأولى والثانية.

تظهر التحليالت اإلحصائية للبيانات وجود معامل ارتباط موجب عالي) r = 0.996)مع قيمة p معنوية ، ومعامل تحديد (R^2 = 0.993) بين Ra°22⁶Ra و ²²⁶Ra عينات التربة للمجموعتين الأولى والثانية ، $(0.05{\ge}p)$ بينما اليوجد ارتباط في المجموعة الثالثة.

1. Introduction

 Ionizing radiation exposure comes from radionuclides of natural and artificial origin via direct contact, inhalation of dust and radon gas, and ingestion of contaminated products or water. The ingested radionuclides go into the circulation system and accumulate in specific tissues and organs, which may cause biological effects [1- 6].

 Naturally Occurring Radioactive Materials (NORM) are responsible for eighty percent of all human radiation exposure; the remaining comes from nuclear processes and cosmic rays, known as background radiation [7-9]. The primary causes of an increase in background levels of ionized radiation in the natural environment are due to some extractive activities of some industries that generate materials and wastes containing natural radionuclides at significant concentrations, and therefore they have to be considered as NORM or TENORM (Technologically Enhanced NORM) [10], such as the production of fuels (coal, oil, and gas), fertilizers and cement, mining and milling, industrial minerals, and radioisotope extraction. The oil extraction and petroleum industries are considered to contain the most radioisotopes [11-13]. When oil is extracted from the ground, it contains radionuclides from the 232 Th, 238 U series, and $40K$ that can be concentrated on the surface of equipment and pipes in the form of sludge and scale due to chemical and physical processes, as well as producing water which associated with the crude oil [14-16]. Due to the physical and chemical properties of uranium and thorium elements, they remain in the oil phase, while radium, because of its solubility in special physical conditions, appears in the liquid phase through the reservoir rocks [17-21].

The produced water may include radioactive materials such as 226 Ra and 228 Ra and their decay products from dissolved mineral salts [22]. When the produced water is brought to the surface, a decrease in temperature and pressure allows these dissolved minerals (radium salts) to precipitate as hard insoluble barite (scale) and deposit on the interior surfaces of piping and other production equipment. Therefore, the produced water may be considered the most significant source of radioactive waste production by the oil and gas industry [8],[23].

 The main environmental hazard of NORM-contaminated waste is the radioactive component of ²²⁶Ra (half-life =1620 years) from the uranium-238 series and ²²⁸Ra (half-life =5.8 years) from the thorium-232 series. None of the radium isotopes radioactive-decay products have a half-life that is as long as the parent radium nuclides; hence, the radioactivity of the decay chain will not persist significantly longer than the parents $(^{226}Ra$ and ^{228}Ra) radium isotopes [23],[24]. Radium is very important in radiological protection due to its relative presence in nature, radiotoxicity, and long half-life [25],[26]. The ²²⁶Ra $/28$ Ra activity ratio can be considered to be a fingerprint of the $^{238}U^{232}$ Th mass ratio in the geological formation of the reservoir; this ratio depends on the age of NORM waste in soil due to the difference of 226 Ra and 228 Ra half-lives [8],[27],[28].

 Some of the production companies dispose of their liquid wastes (water) into unlined pools that are drained underground creating radioactive precipitate in the soil; this is one of the origins of soil contamination. The second source of soil contamination is the periodic maintenance and cleaning of equipment (degassing stations, isolation stations, pipes, valves, etc....) that produces radioactive wastes as precipitations, scale, and sludge forms. The second source was investigated in this study.

 The current study objectives are to estimate the risk and dose of the existing contamination of oil fields in Iraq and determine the amount of activity concentration of radionuclides for contaminated soil of ground down degassing stations, selected areas, and near drilling wells, worker staff, and residents of living areas near these sites.

2. Materials and methods

2.1. Area of Study

 Three oil fields in Basra governorate, which is located around 450 kilometers south of Baghdad, the capital city of Iraq: Al Rumaila southern, Majnoon, and Khudair Aalmay storage site were investigated in this study, as shown in Figure 1.

- 1- Al Rumaila oil field, a super-giant [oil field](https://en.wikipedia.org/wiki/Oil_field) located in southern [Iraq,](https://en.wikipedia.org/wiki/Iraq) about 420 km south of Baghdad, accounts for the largest oilfield in Iraq and the third largest oil field in the world.
- 2- Majnoon oil field, a super-giant [oil field](https://en.wikipedia.org/wiki/Oil_field) located in [Basra](https://en.wikipedia.org/wiki/Basra) in southern [Iraq.](https://en.wikipedia.org/wiki/Iraq) It is one of the richest oil fields in the world.
- 3- Khadhir Almay storage location of radioactive waste, which contains a huge amount of all types of waste collected from all oil companies of Basra.

Figure 1: The three oil fields in Basra governorate

2.2. Samples collecting

 Soil samples were collected (1 kg for each sample) from selected locations in Basra governorate /southern Iraq from three different oil fields: Al Rumaila southern oil field, Khudair Almay storage site, and Majnoon oil field, by a special team from the Iraqi ministry of science and technology in 2009, 2014, 2018, and 2022. The first group of soil samples (15 selected samples) were collected from different depths (30-150 cm) and near the degassing stations in the Al Rumaila oil field. The second group of soil samples (10 selected samples) were collected from the waste storage site at Khudair Amay known to be of the highest contamination level in this location. This was done with portable radiological survey devices. The third group (10 selected samples) was collected from an oil well at different depths and from a waste pit near the well in the Majnoon oil field. The fourth group (10 samples) was collected from various clean locations in each site as background samples for comparison.

2.3. Sample Preparation

The soil samples were dried at 80° for 8 hours and sieved by a mechanical 60 mish sieve (250 μm particle size). Each sample was put in a special standard plastic 500 ml container (Marinelli beaker). The samples were sealed and stored for 3-4 weeks before the measurement to establish an equilibrium of the decay daughter radionuclides with their parents and measured by gamma spectrometer [29-33], as illustrated in Figure 2. The analytical measurements were carried out in the central laboratories directorate (CLD) / Atomic Energy Commission (IAEC) in the Al-Tuwaitha site in the gamma emitter laboratory, which is granted the accreditation certificate from the Iraqi organization for accreditation system IQAS No.TL 045.

a. sample collecting b. drying of samples

c. sieving d. Soil samples in Marnillie beaker

Figure 2: Sample preparation

2.4. Measurement of Samples

 Gamma spectrometry system (ORTEC company) of 65% relative efficiency and a resolution (<1.9) keV based on measurements of (1.332) MeV gamma-ray photo peak of ${}^{60}Co$ with Gamma Vision-32 software version-6 were used in this study for the analysis of gammaray spectra. This system consists of a coaxial high-purity germanium detector with an operating positive voltage of 1500 volts. A standard multi-gamma radioactive source, type (CCPS) certification No. 9031-OL-505/13, was used for energy and efficiency calibration. This radioactive source contains 13 radioisotopes with different gamma-ray energies from 59.5 keV of 241 Am to 1836.08 keV of 88 Y to cover all radionuclides of the NORM samples. Gamma Vision software was used to acquire and treat the collected data to obtain information such as dead time, isotopes and their activity, minimum detection activity (MDA), and compound relative uncertainty for each radionuclide of each energy.

 The activity concentration (A) of radionuclides in the samples was calculated using the following equation [34]:

$$
A = \frac{N}{t \times \gamma \times \epsilon \times M}
$$

where: N is the net area of the photo peak in gamma spectrum, t is the counting time (s), γ is the emitting probability of gamma-ray $(\%)$, ϵ is the detection efficiency $(\%)$ and M (kg) is the sample mass.

 The measurement methods for determining the activity concentrations of radionuclides for NORM sample are divided into indirect and direct. The indirect measurement method using progeny radionuclides is commonly applied for determining the activity concentration of parent ²²⁶Ra and ²²⁸Ra isotopes. The determination of ²²⁶Ra in environmental solids by gamma spectrometry has long been based on the detection of emissions of the radon gas progeny $(2^{22}Rn)$ nuclides, i.e. ^{214}Pb and ^{214}Bi (solid elements) after an ingrowth period of at least 20 days where sample is tightly sealed to ensure secular equilibrium between 226 Ra and its progeny. ²²⁸Ra is measured indirectly through its progeny, ²²⁸Ac in equilibrium, generally by the 911 keV [15], [34-39]. Since 238 U does not release sufficient gamma rays that can be detected directly using gamma-ray spectrometry, NORM samples from the extraction of oil and gas from a deep part of the earth are often evaluated by their daughter products in equilibrium, i.e. thorium-234 (half-life = 24.1 d) at 63.29 and 92.35 keV and $\frac{234 \text{ mPa}}{234 \text{ mPa}}$ at 1001.03 keV, the first and second daughter of ²³⁸U that were used in the present work to indicate its activity of the soil samples instead of ^{214}Pb and ^{214}Bi , which were used to determine ^{226}Ra at energy line 351.9 and 609.3 keV respectively; 214 Pb and 214 Bi were used to determine 238 U in environmental samples [20]. ²¹²Pb and ²¹²Bi were used to determine ²²⁴Ra at 238.63, and 727.33 keV, respectively, ²²⁸Ac to determine ²²⁸Ra at 911.2 keV, and used to determine ²³²Th in environmental samples [40-42] as presented in Table 1.

The direct measurement method of radioactivity concentration of ²²⁶Ra can be used at 186.2 keV energy photo-peak, while ²³⁵U activity was measured at 185.72 keV, which overlaps with the 186.2 of ²²⁶Ra keV energy line. ²³⁵U is usually present at a much lower concentration than ²²⁶Ra in environmental samples due to its abundance ratio in nature. ⁴⁰K concentrations can be measured by their gamma rays at 1460.8 keV [13],[43], as shown in Table 1.

isotope	Isotopes indicator	Energy (keV)	Gamma Emissions probability (γ) $(\%)$		
	226Ra	186.2	3.56		
^{226}Ra	^{214}Pb	351.9	35.6		
	^{214}Bi	609.3	45.49		
$^{232}Th/^{228}Ra$	228 Ac	911.2	26.2		
	^{212}Pb	238.6	43.6		
^{224}Ra	^{212}Bi	727.3	6.65		
40K	40 _K	1460.8	10.67		
238U	234 mPa	1001.03	0.84		
	234Th	92.3	2.42		
235 ^T	235 U	185.7	57.0		

Table 1: Summary of gamma spectrometry of radionuclides determination

2.5. Evaluation of Radiological Parameters

2.5.1.Radium Equivalent Index

Radium Equivalent (Ra_{eq}) index is a valuable tool of NORM for monitoring the safety standards of residential buildings, it is estimated that 370, 259, and 4810 Bq. kg⁻¹ of $(^{226}Ra$, ²³²Th, and ⁴⁰K) radionuclides, respectively, produce the same gamma dose rate. Ra_{eq} is calculated using Eq. (1) [4],[13],[33],[44];

$$
Ra_{eq} = C_{Ra-226} + 1.43C_{Th-232} + 0.077 C_{k-40} \qquad \qquad \ldots \ldots \ldots \ldots (1)
$$

where; $C_{\text{Ra-226}}$, $C_{\text{Th-232}}$ and C_{k-40} are the radioactivity concentration in Bq.kg⁻¹ of (²²⁶Ra, ²³²Th and ⁴⁰K), respectively.

2.5.2. Gamma absorbed dose rate

The absorbed dose rate (DR) of gamma-rays at 1 m up the ground is calculated from ²²⁶Ra, 232 Th, and 40 K concentrations in the soil where the contribution of other radionuclides to the environmental background is negligible [2],[6],[45],[46]. The *DR* is calculated using Eq. (2):

$$
DR (nGy/h) = 0.461A_{Ra-226} + 0.623A_{Th-232} + 0.0414A_{K-40} + \ldots \ldots \ldots \ldots \ldots \ldots \tag{2}
$$

2.5.3. Representative gamma index

 Gamma index Iγ is an indicator that can be used to examine whether the representative sample meets the dose limits criteria [45], [47]. Iy is calculated using Eq. (3):

$$
I\gamma = \frac{A_{Ra-226}}{150} + \frac{A_{Th-232}}{100} + \frac{A_{K-40}}{1500} \le 1
$$
 (3)

2.5.4. Annual effective dose

The $EQ(Sv.y⁻¹)$ is calculated using the transformation factor from the absorbed dose in the air to the effective dose of adults which equals 0.7 Sv/Gy and the indoor and outdoor fraction occupancy factor (0.8 and 0.2, respectively) suggested by UNSCEAR Report (2000, 2008), *EQ* is determined by Eq. (4) & (5) [48]:

Outdoor:
$$
EQ (mSv.y^{-1}) = (DR) (nGyh^{-1}) \times (8760) h \times (0.2) \times (0.7) SvGy^{-1} \times 10^{-6}
$$
 ... (4)
Indoor: $EQ (mSv.y^{-1}) = (DR) (nGyh^{-1}) \times (8760) h \times (0.8) \times (0.7) SvGy^{-1}$... (5)

2.5.5. External and internal radiation hazard indices

 Assuming that local soil and rocks are used to build houses, it is anticipated that radiological doses are delivered in two different ways: externally, from direct radiation from soil and stones, and internally, from exposure to radon-222 gas, a descendant of 226 Ra that poses a threat to the respiratory system. These indices are calculated using the following equations [13],[49]:

$$
H_{ex} = \frac{A_{Ra-226}}{370} + \frac{A_{Th-232}}{259} + \frac{A_{K-40}}{4810} \le 1
$$
 (6)

$$
H_{in} = \frac{A_{Ra-226}}{185} + \frac{A_{Th-232}}{259} + \frac{A_{K-40}}{4810} \qquad \le 1 \tag{7}
$$

3. Results and discussion

3.1. *Activity concentration of radionuclides*

The laboratory radioactivity measurements and analysis of soil samples are the most important in the characterization of any investigated sites. Table 2 shows the statistical summary of the activity concentration (AC) of the radionuclides in the first and second-group soil samples of Al Rumaila oil field and the Khudair Almay storage site. These two groups of soil samples were combined due to their similar characteristics. The results of 25 soil samples of NORM showed a relatively high radioactivity concentration of most radionuclides, especially ²²⁶Ra and ²²⁸Ra and their progenies. Table 3 presents the correlation between the AC of all radionuclides in soil samples of this group, Figure 3 shows the correlation between the AC of 226 Ra and 228 Ra soil samples.

 The radioactivity concentration of the third group soil samples (10 soil samples) belongs to a waste pit near an oil well in the Majnoon oil field, and from different depths from a known well, as presented in Table 4. The results showed that the AC of all-natural radionuclides of ²³⁸U, ²³²Th, and ⁴⁰K series, which included the ²³⁸U and ²³²Th nuclides, disappeared in the first and second groups of soil samples. The activity concentration of these radionuclides $(^{238}$ U and 232 Th) and their progenies were greater than the terrestrial levels because these soil samples were collected from a waste pit accumulated from drilling soil of an oil well from different levels reaching a 3500 m depth. Figure 4 shows the correlation between the AC of 226 Ra and 228 Ra of soil samples, while Table 5 presents the correlation between the AC of all radionuclides in soil samples of this group.

 Table 6 presents the AC of the fourth group of (10) soil samples from uncontaminated areas far away from the oil extraction site and gas industries, which served as the background level. The results showed AC for all-natural radionuclides of ²³⁸U and ²³²Th series and ⁴⁰K of the terrestrial primordial radionuclides and their progenies. Table 7 shows the correlation between the AC of all radionuclides in soil samples of this group; Figure 5 shows the correlation between the AC of 226 Ra and 228 Ra in soil samples, and Figure 6 presents the activity concentrations of 226 Ra and 228 Ra of all four groups of soil samples.

3.2. Evaluation of Radiological Parameters

 Table 8 shows the statistical summary of the radiological parameters for all soil samples (45 samples), where the total radium equivalent activity and the absorbed dose rate for most samples were above the acceptable levels $(370 Bq.kg^{-1}$ and $59 nGyh^{-1}$ [13], [14]. The mean of annual outdoor and indoor effective dose, and representative gamma index Iγ exceeds the acceptable levels (1mSv/y), as shown in Figure 7. The internal and external hazard values also exceeded the acceptable levels of unitary, as illustrated in Figure 8.

3.3. Statistical Analysis

 The statistical analyses were conducted using Microsoft Excel 2010. Pearson's correlation coefficient (r) was used to verify the significance of the possible relationship between variables at a 95% confidence level; when the correlation coefficient value is close to 1 and p \leq 0.05, there is a significant correlation between two variables [50], [51]. Table 2 shows the degree of association between each radionuclide AC for the Al-Rumela and Khudhr Almay oil fields. Figure 3 shows the significant correlation between the radioactivity concentration of ²²⁶Ra and ²²⁸Ra (strong positive correlation r=0.996 with a significant p-value of ≤ 0.05)

and a coefficient of determination (R^2) of 0.993. Table 4 and Figure 4 show an insignificant correlation between ²²⁶Ra and ²²⁸Ra in Majnoon oil field soil samples. Table 6 and Figure 5 shows a significant correlation between the radioactivity concentration of ^{226}Ra and ^{228}Ra (positive correlation $r = 0.72$) and a coefficient of determination (R^2) of 0.525 of the background soil samples.

Table 2 The statistical summary of the radioactivity concentration (Bq.kg⁻¹) of Al-Rumaila and Khudair Almay oil field soil samples (the compound uncertainty $\pm\%$ of the radionuclide activity)

	226 Ra	228 Ra	40 _K		^{212}Pb	^{214}Pb	212 Bi	^{214}Bi	208 Tl
Max	8.24×0.5 ± 1.5	6.16×04 ±1.43	4.77×03 ±6.9	9.75×04 ± 0.6	2.61×0.5 ± 1.2	1.34×0 $5\pm$ 3.4	2.27×0 5 ± 1.1	5.20×04 ± 2.1	
Min	2.46×02 ± 1.1	3.01×01 ± 1.6	2.74×02 ± 4.5	3.07×01 ±6	1.83×02 ± 2.5	4.80×0 1 ± 3.6	1.86×0 $2 + 2.6$	1.62×01 ± 1.5	
mean	7.67×04	6.26×03	1.38×03	9.89×03	4.57×04	1.29×0 4	4.03×0 4	7.43×03	
median	1.34×04	1.17×03	5.51×02	1.75×03	1.32×04	2.28×0 3	1.15×0 4	5.09×02	
range	8.23×0.5	6.16×04	4.50×03	9.75×04	2.61×0.5	1.33×0	2.27×0	5.20×04	
	No. of sample	25							

Table 3 Correlation coefficient between different radionuclides of Al- Rumaila and Khudair Almay oilfield

*Shows significant correlation coefficient $(P = 0.95)$.

Figure 3: The correlation between radioactivity concentration of ²²⁶Ra and ²²⁸Ra of Al-Rumaila and Khudair Almay oil field soil samples

$\frac{1}{2}$ and $\frac{1}{2}$										
	226R a	228 Ra	40 _K	^{212}Pb	214P _b	212 Bi	^{214}Bi	235 U	234 m Pa	234 Th
Max	608. $6 + 8.$	$95.4 \pm$ 16	2662.2 _± 3	149.9 _± 4.1	527.0 ± 3 .2	296.9 ± 1	611.0 ± 3.1	$14.1\pm$ 23	1035. $8 + 28$	237.9 ± 5 3
Min	312. 1 ± 3 . $\overline{4}$	$31.3+$ 20	$856.7+4$	23.8 ± 1 $\overline{4}$	$40.9 + 4$	40.4 ± 5	292.5 ±6	$BDL*$	BDL	BDL
Ave.	484. 8	55.0	1650.1	61.0	356.9	104.6	448.7	9.4	589.1	156.4
median	508. 3	50.5	1702.3	44.5	401.1	67.5	479.5	9.8	680.4	189.4
range	296. $\overline{}$	64.1	1805.5	126.1	486.2	256.5	318.5	10.3	974.2	185.4
Std	97.2	20.4	590.9	42.0	136.0	76.9	101.5	4.3	363.7	71.7

Table 4: The statistical summary of the radioactivity concentration (Bq.kg⁻¹) of Majnoon oil field soil samples (the compound uncertainty $+$ % of the radionuclide activity)

*BDL: below detection limit

Table 5: Correlation coefficient between different radionuclides of Majnoon oil field

	226 Ra	228 Ra	40 _K	^{212}Pb	^{214}Pb	^{212}Bi	^{214}Bi	235 U	234mPa	234Th
226 Ra	1									
228 Ra	-0.34	1								
40 _K	-0.09	0.69	1							
^{212}Pb	-0.24	0.93	0.65	1						
^{214}Pb	$0.61*$	0.00	0.38	0.00	1					
^{212}Bi	$0.91*$	$0.91*$	0.62	0.88	0.25	1				
^{214}Bi	0.82	-0.13	0.00	-0.35	$0.58*$	0.29	1			
235 U	0.24	$0.57*$	0.53	$0.56*$	0.31	$0.84*$	0.25	1		
234 m Pa	0.23	-0.51	-0.52	-0.37	0.41	-0.12	0.29	0.37	1	
234 Th	-0.01	0.38	0.20	0.35	-0.01	0.74	-0.07	0.95	0.34	1

*Shows significant correlation coefficient ($P = 0.95$).

Figure 4: The correlation between radioactivity concentration of ²²⁶Ra and ²²⁸Ra of Majnoon oil field soil samples

	238 U		232 Th/ ²²⁸ Ra	226 Ra		224 Ra	235 U	40 _K
	^{214}Pb	^{214}Bi	228 Ac		^{212}Pb	212 Bi		
Max	$80.9 + 7$	88.90 ± 8	$26.30+9$	86.03 ± 8	29.50	41.90	$0.88 + 8$	$427.0 + 5$
Min	$29.30 \pm$ 13	31.50 ± 11	13.90 ± 12	$15.8 + 9.$	10.4 ± 6.3	21.8 ± 5.7	BDL	224.0 ± 8
Ave.	40.50	47.90	17.80	30.70	14.40	29.90	0.83	361.00
median	33.30	43.00	16.70	21.00	12.60	26.70	0.82	384.00
range	51.60	57.30	12.40	70.20	19.10	20.10	0.08	204.00

Table 7: Correlation coefficient between different radionuclides of background soil samples

Figure 5: The correlation between radioactivity concentration of ²²⁶Ra and ²²⁸Ra of background soil samples

Figure 6: Radioactivity concentration of ²²⁶Ra and ²²⁸Ra of four soil samples groups

Figure 8**:** The external and internal hazard indices in different in three different oilfields

4. Conclusion

 According to the recommendations of the International Atomic Energy Agency (IAEA), the activity concentration results (AC) to the first and second group soil samples of NORM showed a relatively high level of 226 Ra and 228 Ra and their progeny, which they are from the 238 U and 232 Th series radionuclides. The results also showed that radium isotopes leached and transferred with extracted oil and produced water because its solubility in water in high depth by a physical condition (increasing in pressure and temperature) which will be in the liquid phase, therefore ²³²Th and ²³⁸U radioactivity concentrations did not appear or were neglected in these analyzed samples. There is a strong correlation coefficient between 226 Ra and 228 Ra radionuclides in the first and second groups $(+0.996)$, and the ²²⁶Ra / ²²⁸Ra ratio is 12.3 of the mean values.

The AC results of the third group showed all-natural radionuclides of 238 U and 232 Th series, which disappeared in the first and second groups. AC of these radionuclides and their progenies were noticed to be greater than the terrestrial levels. There was no correlation between ²²⁶Ra and ²²⁸Ra in the third group (-0.34), which belongs to a waste pit near an oil well containing drilling soil from various depths of 3500m maximum depth. AC of ²³⁸U and 232 Th series varied in each depth depending on the geological formation of the earth levels. In the first and second group samples, all radiation hazard indices, such as radium equivalent activity, absorbed dose, and effective dose rate, were higher than the permissible limits,

whereas they varied in the third group. So, it may be said that the soil samples cannot be used as safe building materials.

 The internal and external hazard indices should be less than or equal to unity because most of the places under investigation in the study areas have ambient dose rates higher than the global average of 59 nGy/h. The external hazard index is derived from the formulation of the radium equivalent $(R_{a_{eq}})$ because the upper limit is the highest allowable value. Assuming that its highest permitted value corresponds to the upper limit of radium equivalent (Ra_{eq}) 370 Bq/kg^{-1} , the external hazard index is obtained from the expression of radium equivalent, therefore, the external dose rate did not exceed 1.5 mSv.y⁻¹. Gamma index Iy values must remain below the 1.0 mSv.y^{-1} safe limits. Certain locations in the research region have radiation dose levels higher than those recommended by the ICRP, UNSCEAR, and IAEA for the general population but within for the workers. Hence, to reduce the radiation dose values as low as possible, the workers should use the proper personal protective equipment and limit their exposure time.

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