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Assessment of the Natural Radioactivity Levels of Soil Samples in IT1 Oil Reservoirs in Kirkuk City, Northeast Iraq

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Abstract

In this study, Gamma-ray spectrometry with (HPGe) detector was used to measure the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples collected from IT1 oil reservoirs in Kirkuk city, northeast Iraq. The "spectral line Gp" gamma analysis software package was used to analyze the spectral data. ^{226}Ra specific activity varies from 9 ± 0.34 Bq.kg^{-1} to 17 ± 0.47 Bq.kg^{-1}. ^{232}Th specific activity varies from 6.2 ± 0.08 Bq.kg^{-1} to 18 ± 0.2 Bq.kg^{-1}. ^{40}K specific activity vary from 25 ± 0.19 Bq.kg^{-1} to 118 ± 0.41 Bq.kg^{-1}. The radiological hazard due to the radiation emitted from natural radionuclides in soil samples was also assessed. The average values of the radium equivalent (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), gamma index (I_{γ}), absorbed dose rate (D), outdoor ($AEDE_{out}$) and indoor ($AEDE_{in}$) annual effective dose equivalent were 35.391 Bq.kg^{-1}, 0.095, 0.128, 0.253, 16.118 nGy.h^{-1}, 0.0197 mSv.y^{-1}, and 0.0790 mSv.y^{-1}, respectively. All values obtained for the activity concentrations for ^{226}Ra, ^{232}Th, and ^{40}K and the radiological hazard were lower than the global values. This indicates that no harmful radiation effects are posed to the population who work and live near the study area.

Article Info.

Keywords:
HPGe detector, radiological hazard, Radium equivalent, Soil, Kirkuk City.

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1. Introduction

Naturally occurring radioactive materials (NORM) are a global type of pollutants that contaminate the environment and cause a severe negative impact on human health, soil, water, air, and living organisms [1, 2]. Natural radiation levels have increased in the last few decades due to human activities such as mining, processing of radioactive substances, handling of radioactive wastes, nuclear power used to generate energy, as well as using radiation in medicine [3, 4].

During oil and gas extraction from the ground, NORM rises to the surface through the pipes along with the crude oil and natural gas, and produces water that accumulates in scale and sludge scrapings. NORM accumulation in the oil and gas equipment can differ depending on the operational processes and geological formation, such as depth and type of the rocks in the oil wells [5]. The primary radionuclides of concern in the oil and gas stream responsible for most of the external exposure are ^{226}Ra, ^{232}Th, and ^{40}K [6]. During repeated oil and gas production operations, the NORM may be concentrated over time to reach high levels and become a health risk to those working in the oil industries and the surrounding area. It is important to know the proportions of these substances in the environment and the permissible limits for their concentrations to avoid the risks resulting from them [7].

This study aims to assess the natural radioactivity levels of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples collected from IT1 oil reservoirs in Kirkuk city, northeast Iraq and

compare the results with other studies and recommended values. The study also aims to calculate the radiological hazard due to the radiation emitted from natural radionuclides in soil samples. Radium equivalent (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), gamma index (Iγ), absorbed dose rate (D), and outdoor ($AEDE_{out}$) and indoor ($AEDE_{in}$) annual effective dose equivalents were calculated and compared with the international values.

2. Materials and Methods

2.1. Study Area and Sample Collection

The study area was located within Kirkuk city, northeast of Iraq. Kirkuk is one of the wealthiest cities with crude oil in Iraq, and its oil constitutes about 12% of Iraq's total oil reserves [8]. Kirkuk crude oil is one of the standard light oils with its abundant production and oil quality. The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K was measured in crude oil soil samples collected from IT1 oil reservoirs in North Oil Company (N.O.C) in Kirkuk city as shown in Fig. 1, and Table 1 shows the codes and location coordinates of the samples.

The soil samples were taken at a depth of 20 cm and dried in the open air, then crushed and sieved to obtain a homogeneous powder. 1kg of each sample was taken and stored for 30 days in a completely sealed Marinelli beaker to achieve radiation equilibrium before starting the measurement.

Figure 1: Sample sites in IT1 oil reservoirs.

Table 1: The collected samples' location coordinates.

Sample codes	Longitude(°E)	Latitude(°N)
TS1	44°12'22.29"	35°25'12.28"
TS2	44°12'17.65"	35°25'20.98"
TS3	44°12'30.71"	35°25'18.81"
TS4	44°12'25.48"	35°25'22.47"
TS5	44°12'31.52"	35°25'26.61"
TS6	44°12'26.16"	35°25'28.19"

2.2. Experimental Setup

The activity concentration of radionuclides in soil samples was measured using a gamma-ray spectrometer with a high purity germanium detector (HPGe) of type PN, model BSI (Baltic Scientific Instruments), and made of a (2"x 2") crystal linked to a multichannel analyzer (4096 channel) of 20% efficiency. The detector was shielded by a

4.5 cm thick lead to reduce the level of background radiation for the system; also, copper plates of 0.8 mm thick padded the detector from the inside to minimize the X-ray fluorescence.

Detection efficiency and energy calibration have been done every day as part of quality control procedures to ensure that they were stable during the research period. The calibration was carried out by using standard sources type (CBSS2) of a capacity 1L Marinelli beaker containing a mix of different certified radionuclide sources (^{241}Am (59.3) keV, ^{60}Co (1173.1, 1332.3) keV, ^{137}Cs (661.6) keV). After calibration, each sample was measured for 3600s, and the "spectral line Gp" gamma analysis software package was used to analyze the spectral data.

The gamma-ray lines at 186.21 keV were used to determine the specific activity of ^{226}Ra. The gamma-ray lines of 238.63 keV from ^{212}Pb and 911.204 keV from ^{228}Ac were used to determine the specific activity of ^{232}Th. The specific activity of ^{40}K was measured directly by its own gamma-ray line at 1460.8 keV.

The activity concentration of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K was calculated through γ-ray spectrometry using the following formula [9, 10]:

$$A = \frac{\text{Net count}}{\varepsilon \times G\gamma \times T \times M} \quad (\mathbf{1})$$

where: ε is the absolute gamma peak efficiency of the detector, Gγ is the intensity for gamma-ray energy, T is the counting time for the measurement in seconds, and M is the weight mass of the sample in kg.

2.3. Evaluation of Radiological Hazard Indices

Radium equivalent activity (Ra_{eq}) is one of the most prevalent hazard indices in radiation protection evaluation [11, 12]. It was calculated from the following relation [13]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (\mathbf{2})$$

where; A_{Ra}, A_{Th}, and A_K are the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in Bq.kg^{-1}, respectively.

Determining the external radiation hazard index (H_{ex}) and internal radiation hazard index (H_{in}) is an important index used to investigate the risk due to gamma radiation and they determined from the following relation [14]:

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

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

Gamma radiation representative level index (Iγ) was used to estimate the level of gamma radiation hazard accompanying the natural radionuclides in the investigated samples and was determined from the following relation [14, 15]:

$$I_\gamma = \left(\frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}\right) \leq 1 \quad (\mathbf{5})$$

To determine the absorbed dose rate (D) at 1m above the ground in (nGy.h^{-1}) the following relation was used [14]:

$$D = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K} \quad (6)$$

The outdoor ($AEDE_{out}$) and indoor ($AEDE_{in}$) annual effective dose equivalents due to gamma radiation in soil samples were determined using the following equations [14, 16]:

$$AEDE_{out}(mSv.y^{-1}) = D(nGy.h^{-1}) \times 8760\ (h.y^{-1}) \times 0.7\ (Sv.Gy^{-1}) \times 0.2 \times 10^{-6} \quad (7)$$

$$AEDE_{in}(mSv.y^{-1}) = D(nGy.h^{-1}) \times 8760\ (h.y^{-1}) \times 0.7\ (Sv.Gy^{-1}) \times 0.8 \times 10^{-6} \quad (8)$$

3. Results and Discussion

3.1. The Radionuclides Activity

The specific activity concentration of the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K are presented in Table 2 and shown in Fig. 2 to Fig. 4. ^{226}Ra concentrations varied from 9 ± 0.34 Bq.kg^{-1} in sample TS1 to 17 ± 0.47 Bq.kg^{-1} in sample TS4 with an average value of 12 ± 0.28 Bq.kg^{-1}, which was less than the recommended global limits of 35 Bq.kg^{-1} [14]. ^{232}Th concentrations varied from 6.2 ± 0.08 Bq.kg^{-1} in sample TS1 to 18 ± 0.2 Bq.kg^{-1} in sample TS3 with an average value of 12.28 ±0.19 Bq.kg^{-1}, which were less than the recommended global limits of 30 Bq.kg^{-1} [14]. ^{40}K concentrations vary from 25 ± 0.19 Bq.kg^{-1} in sample TS1 to 118 ± 0.41 Bq.kg^{-1} in sample TS6 with an average value of 75.66 ± 0.10 Bq.kg^{-1}, which were less than the recommended global limits of 400 Bq.kg^{-1} [14]. Table 3 compares the average values of ^{226}Ra, ^{232}Th and ^{40}K in the soil samples obtained from other studies with those determined in the present study. It was found that the average values of ^{226}Ra, ^{232}Th and ^{40}K in the present study were lower than those obtained in other countries and world-recommended values (UNSCEAR, 2000) [14]. This might be due to the heterogeneity of the environments in which the radionuclides are deposited since it could be greatly influenced by the type of mineral or source rock and other factors.

3.2. The Radiological Hazard

The radiological hazard results are shown in Table 4. The radium equivalent activity (Ra_{eq}) value ranged from 19.791 Bq.kg^{-1} to 44.439 Bq.kg^{-1}, with an average value of 35.392 Bq.kg^{-1}, which was less than the permissible limits of 370 Bq.kg^{-1} [14]. The external hazard index (H_{ex}) value ranged from 0.053 to 0.120, with an average value of 0.096. The internal hazard index (H_{in}) value ranged from 0.078 to 0.152, with an average value of 0.128. The values of the H_{ex} and H_{in} indices must be less than one for the radiation hazard to be negligible; all values that were obtained in this study were less than one, indicating that there is no radiologic risk. The gamma index (Iγ) value ranged from 0.139 to 0.318, with an average value of 0.253. The calculated values for all samples were lower than the international values (Iγ < 1). The absorbed dose rate (D) value ranged from 8.945 nGy.h^{-1} to 20.044 nGy.h^{-1} with an average value of 16.118 nGy.h^{-1}. The obtained values of the absorbed dose rate in this study were less than the reference value of 55 nGy.h^{-1} [14]. The calculated outdoor AEDE value ranged from 0.0110 mSv.y^{-1} to 0.0246 mSv.y^{-1} with an average value of 0.0198 mSv.y^{-1}. The calculated indoor AEDE value ranged from 0.038 mSv.y^{-1} to 0.080 mSv.y^{-1} with an average value of 0.068 mSv.y^{-1}. The outdoor and indoor AEDE values in the studied

samples were less than the recommended value of 0.42 $mSv.y^{-1}$ and 0.08 $mSv.y^{-1}$, respectively [14].

Table 2: The specific activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in the soil samples.

Sample codes	Specific activity of ^{226}Ra ($Bq.kg^{-1}$)	Specific activity of ^{232}Th ($Bq.kg^{-1}$)	specific activity of ^{40}K ($Bq.kg^{-1}$)
TS1	9±0.34	6.2±0.08	25±0.19
TS2	12±0.41	15.2±0.19	82±0.34
TS3	12±0.41	18±0.20	87±0.34
TS4	17±0.47	8.8±0.13	42±0.24
TS5	12±0.39	12.1±0.15	100±0.37
TS6	10±0.36	13.4±0.14	118±0.41
Min	9±0.34	6.2±0.08	25±0.19
Max	17±0.47	18±0.20	118±0.41
Ave	12±0.28	12.28±0.19	75.66±0.10

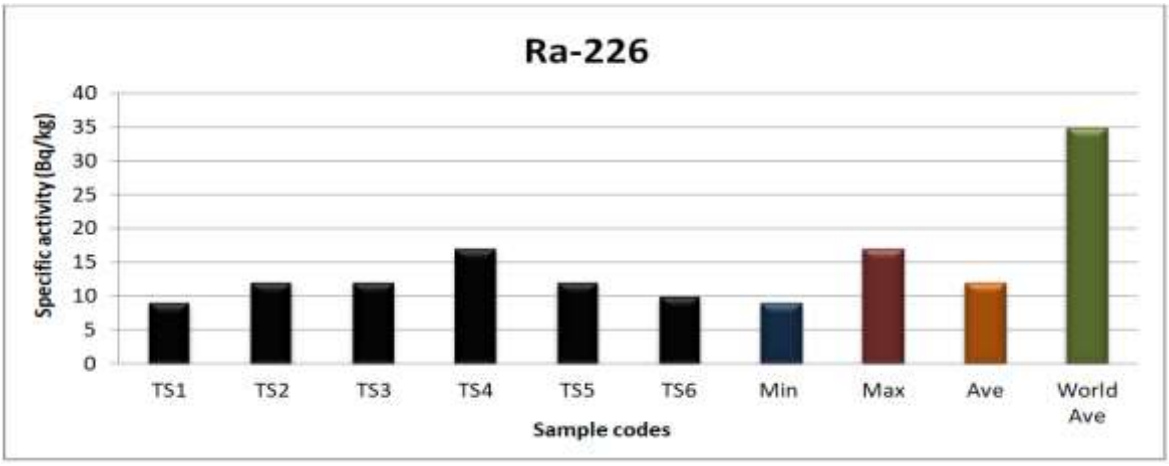

Figure 2: Comparison of the specific activity of ^{226}Ra in soil samples with the worldwide average.

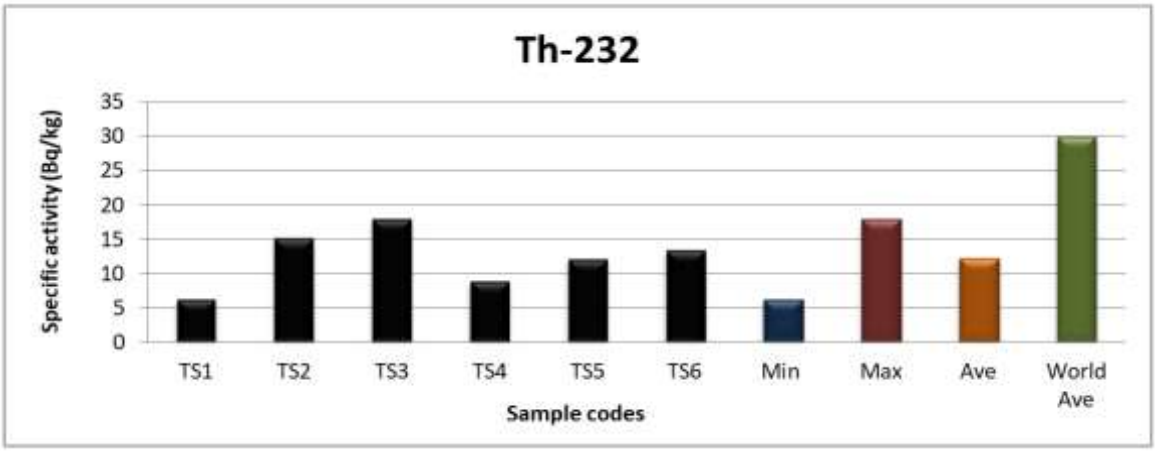

Figure 3: Comparison of the specific activity of ^{232}Th in soil samples with the worldwide average.

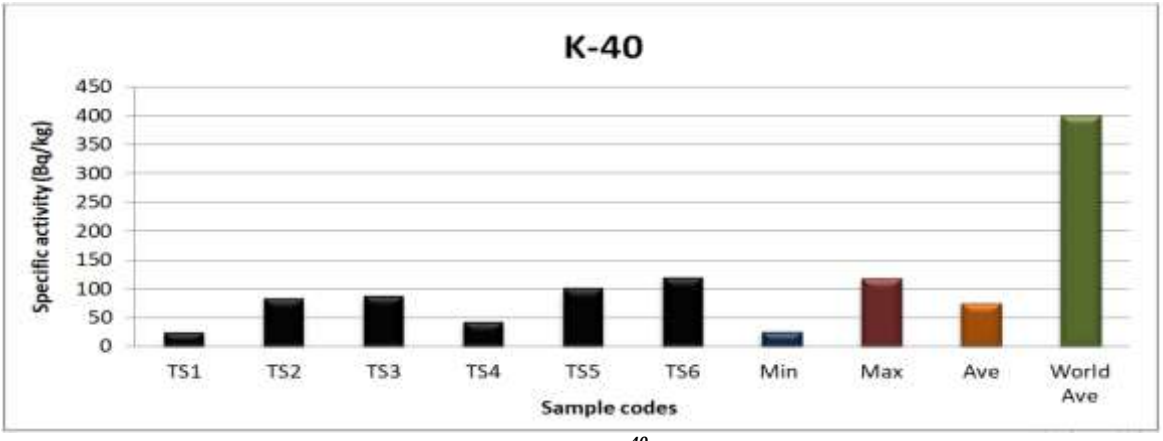

Figure 4: Comparison of the specific activity of ^{40}K in soil samples with the worldwide average.

Table 3: Comparison of ^{226}Ra, ^{232}Th, and ^{40}K average concentrations values of samples from various regions of the world.

Country	Specific activity (Bq.kg^{-1})			Reference
	^{226}Ra	^{232}Th	^{40}K	
Iran	36.68	16.12	276.4	[17]
Nigeria	41.0	29.7	412.5	[18]
Iraq	33.588	20.647	511.604	[19]
Qatar	20.05	16.43	216.69	[20]
Albania	20	-	326	[21]
Kuwait	19.41	14.85	330.31	[22]
Iraq	61.9	28.5	362.4	[23]
Iraq	12	12.28	75.66	Present Study
World average	35	30	400	[14]

Table 4: The radiological hazard in the soil samples.

Sample codes	Ra_{eq} (Bq.kg^{-1})	H_{ex}	H_{in}	I_γ	D (nGy.h^{-1})	$AEDE_{outdoor}$ (mSv.y^{-1})	$AEDE_{Indoor}$ (mSv.y^{-1})
TS1	19.791	0.053	0.078	0.139	8.945	0.011	0.0439
TS2	40.05	0.108	0.141	0.287	18.144	0.0223	0.089
TS3	44.439	0.12	0.152	0.318	20.044	0.0246	0.0983
TS4	32.818	0.089	0.135	0.229	14.921	0.0183	0.0732
TS5	37.003	0.1	0.132	0.268	17.022	0.0209	0.0835
TS6	38.248	0.103	0.13	0.279	17.634	0.0216	0.0865
Min	19.791	0.053	0.078	0.139	8.945	0.011	0.0439
Max	44.439	0.12	0.152	0.318	20.044	0.0246	0.0983
Ave	35.391	0.095	0.128	0.253	16.118	0.0197	0.0790
World average	370	≤ 1	≤ 1	≤ 1	55	0.42	0.08

4. Conclusions

The gamma-ray spectrometry with (HPGe) detector was used to determine the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples collected from IT1 oil reservoirs in North Oil Company (N.O.C) in Kirkuk city. The average activity concentration for ^{226}Ra, ^{232}Th, and ^{40}K obtained was lower than the international recommendations. Therefore, these activity concentrations are acceptable for human health or environmental pollution.

The average values of the obtained radiological effects of radium equivalent (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), gamma index ($I\gamma$),

absorbed dose rate (D), and outdoor ($AEDE_{out}$), and indoor ($AEDE_{in}$) annual effective dose equivalents were within the limits of the international recommended values. Thus, the population in this area is not at any health risk due to the exposure to radiation emitted by ^{226}Ra, ^{232}Th, and ^{40}K in the soils.

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Conflict of interest

Authors declare that they have no conflict of interest.

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تقييم مستويات النشاط الإشعاعي الطبيعي لعينات التربة في خزانات نفط IT1 في مدينة كركوك شمال شرق العراق

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الخلاصة

في هذه الدراسة تم استخدام مطياف أشعة جاما مع كاشف (HPGe) لقياس تراكيز النشاط الإشعاعي لـ ^{226}Ra, ^{232}Th, ^{40}K في عينات التربة التي تم جمعها من خزانات النفط الخام IT1 في مدينة كركوك شمال شرق العراق. واستخدمت حزمة برمجيات تحليل جاما (Spectral line Gp) لتحليل البيانات الطيفية. النشاط الاشعاعي النوعي لـ ^{226}Ra تراوحت من 9 ± 0.34 Bq.kg^{-1} الى 17 ± 0.47 Bq.kg^{-1}. النشاط الاشعاعي النوعي لـ ^{232}Th تراوحت من 6.2 ± 0.08 Bq.kg^{-1} الى 18 ± 0.2 Bq.kg^{-1}. النشاط الاشعاعي النوعي لـ ^{40}K تراوحت من 25 ± 0.19 Bq.kg^{-1} الى 118 ± 0.41 Bq.kg^{-1}. كما تم تقييم الخطر الإشعاعي الناجم عن الإشعاع المنبعث من النويدات المشعة الطبيعية في عينات التربة. كانت معدل قيم الفعالية المكافئة للراديوم (Ra_{eq})، معامل الخطورة الخارجي(H_{ex})، معامل الخطورة الداخلي(H_{in})، معدل جرعة كاما(I_γ)، معدل معامل الامتصاص(D)، الجرعة السنوية الفعالة الخارجية ($AEDE_{out}$)، والجرعة السنوية الفعالة الداخلية ($AEDE_{in}$) 35.391Bq.kg^{-1}, 0.09, 0.128, 0.253, 16.118 nGy.h^{-1}, 0.0197mSv.y^{-1}, 0.0790 mSv.y^{-1} على التوالي. جميع القيم التي تم الحصول عليها لتراكيز النشاط الإشعاعي لـ ^{226}Ra, ^{40}K,^{232}Th والخطر الإشعاعي كانت أقل من القيم العالمية، وهذا يشير إلى عدم وجود آثار إشعاعية ضارة على السكان الذين يعملون ويعيشون بالقرب من منطقة الدراسة.

الكلمات المفتاحيه: كاشف HPGe ، الخطر الإشعاعي ، مكافئ الراديوم ، التربة ، مدينة كركوك.