Volume 10, Number 1, 2017. pp. 23-32

Jordan Journal of Physics

ARTICLE

Assessment of Natural Radioactive Concentration Levels in the Oil Drilling Wells in Erbil Governorate Blocks

Ahmed I. Samad[a], **Ali H. Ahmed**[a] and **Saman K. Ezzulddin**[b]

[a] *Department of Physics, Faculty of Science, Salahaddin University, Erbil, Iraq.*
[b] *Department of Environmental Science, Faculty of Science, Salahaddin University, Erbil, Iraq.*

Received on: 7/10/2016; Accepted on: 24/1/2017

Abstract: The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the core samples obtained from Erbil oil blocks have been measured by using NaI(TI) gamma ray spectrometry. Radiological hazard indices were evaluated from the activity concentrations of these radionuclides in order to estimate health effects of the exposure of the public to radiation at the studied area. The calculated Radium equivalent activity (Ra_{eq}) varied from (33.48) to (321.13) Bq.kg^{-1} with a mean value of (127.50) Bq.kg^{-1}. The values of annual gonadal equivalent doses (AGED) indicated 57.89% exceeding the limit of the maximum contamination level. The measured absorbed dose rate (D) values ranged from (16.17) to (140.43) nGy/ h, with a mean value of (56.95) nGy/ h in the oil field, which were 36.8 % higher than the international recommended value of 60 nGy/h [11]. Representative level index, I_γ, for all the samples varied from (0.25 to 2.23), with a mean value of (0.90). The external hazard index (H_{ex}) varied from (0.09 to 0.87) with a mean value of (0.34) and the internal hazard index (H_{in}) ranged from (0.18 to 2.17), with a mean value of (0.91). The calculated annual effective dose equivalent values were 42% higher than the recommended value of (0.29). On the other hand, the values of ELCR in all the samples were higher than the maximum contamination level of (0.29). The measured values of outdoor and indoor AEDE varied from (0.02 to 0.18) mSv/y and from (0.05 to 0.69) mSv/y, with mean values of (0.07) and (0.28) mSv/y for outdoor and indoor effective doses, respectively, which were all below the maximum contamination level of (1) mSv/y.

Keywords: Radioactivity, Hazard indices, NaI(TI) detector, Oil field.

Introduction

Human beings and their surroundings are continuously exposed to different types of ionizing radiation from naturally occurring radioactive materials (NORM), mainly Uranium U, Thorium Th, Potassium ^{40}K and radionuclides produced by human activities [1]. The natural sources of radiation are commonly distributed in all places of the earth crust. On the other hand, man-made sources are generally regional and influence just a small portion of the people [2].

In the petroleum industry, naturally occurring radioactive materials can be generated along their production. NORM may be observed in several geological structures and can be delivered to the earth surface when oil/gas wells are drilled and constructed. Later, these materials may accumulate at the surface in the form of scales and sludge as well as inside equipment used for processing and drilling wells. In addition, they may accumulate in salt wells and sediments inside tanks or basins.

It is lately understood that more than 109 tons of naturally occurring radioactive material (NORM) wastes are produced from different sources all over the world each year. The value of NORM wastes collected during the years is

Corresponding Author: S.K. Ezzulddin **Email**: saman.ezzlddin@su.edu.krd

higher than (1011 tons.m^{-3}). About (1.8 tons.m^{-3}) of NORM wastes are formed by oil and gas industry alone. There are appraisals that only one well can produce about (2.3 tons.m^{-3}) of sludge, mud and scales every year [3].

Gas and oil production is an important industry in Iraq in terms of economy. Important basins in the world contain different types of rock. The northern part of Iraq is characterized by containing more than one bed rock type, including: Kurra Chine, Najmah, Singar, Sargelu, Butmah, Balambo, Naokelekn, Qamchaqa and Sarmord [3]. These bedrocks belong to the Cretaceous-Tertiary sources [1, 2]. Gas and oil basins are distributed between Cretaceous and Tertiary bedrocks [4].

The aim of this paper is the measurement of natural radioactivity level in Erbil oil field as a part of North Iraq. This will confirm a baseline map of natural radioactivity concentration levels in Erbil oil field and determine the absorbed dose rate (D), the radium equivalent activity (R_{aeq}), the external hazard index (H_{ex}), the internal hazard index (H_{in}), the annual effective dose rate (AEDE) and Gamma radiation index (I_γ) for workers who live inside the limits of the Erbil oil field (Oryx Oil Company).

Area of Study

Area of study is located between latitudes 36° 02′ and 36° 25′ N and longitudes 43° 27′ and 43° 49′ E as shown in Fig. 1.

It is situated within the foothill zone of the zagros fold and thrust belt. Tectonically, it was generated and progressed between the Eurasian and Arabian plates as a consequence of various stages of collision. The main geological structure of this area varies spanning from Bakhtiary (Tertiary), Adaiyah, Alan Halite, Baluti (Triassic), … etc. formations, as shown in Table 1 [6]. The major river running to the area of study is the Grater Zap as shown in Fig. 2.

Generally, the climate of the area extends comparable to some other parts of Iraqi Kurdistan region, being semi-arid with obvious influence of the Mediterranean Sea climate on the northeastern and northern parts, which is characterized by dry and hot summer season and cold and wet winter season with rainfall of about 200mm per annum and relative humidity ranging from (46 %) to (48%) [7].

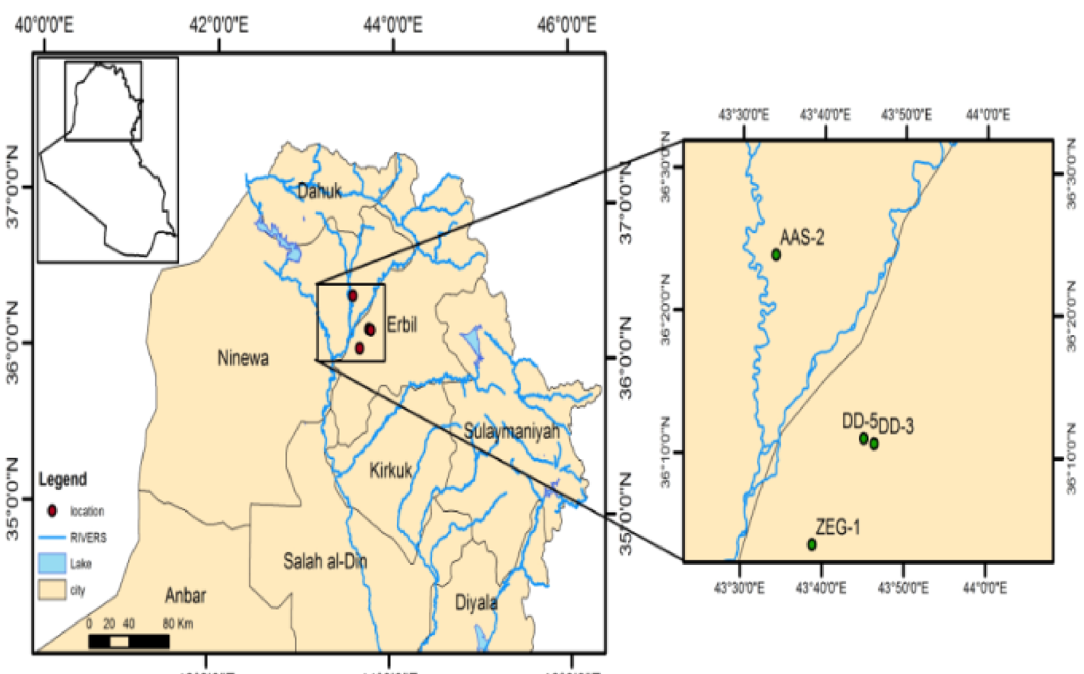

FIG. 1. Map of sample side.

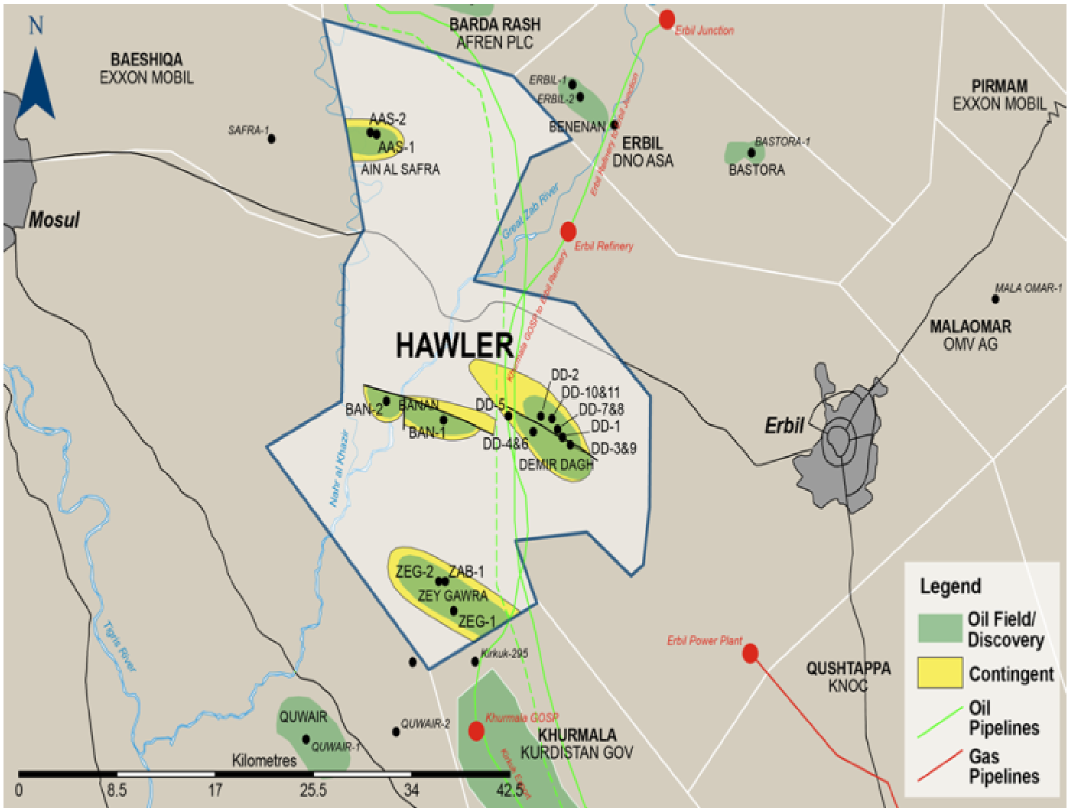

FIG. 2. Map of the oil field.

TABLE 1. Activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in samples collected from four sites: DD-3, DD-5, AAS-2 and ZEG-1 in Erbil oil fields.

Sample name	Sample Code	Depth/ m	C ^{226}Ra (Bq.kg^{-1})	C ^{232}Th (Bq.kg^{-1})	C^{40}K (Bq.kg^{-1})
Adaiyah	S1	2795-3785	37.21	48.99	42.65
Alan Halite	S2	2360-3635	18.67	33.67	45.76
Bakhtiary	S3	65-230	26.63	62.42	447.6
Baluti	S4	3115-4180	38.91	62.55	450.7
Butmah	S5	2990-3920	39.48	67.28	502.3
Geraus	S6	670-1150	24.02	36.5	56.11
Kolosh	S7	1090-1480	12.35	5.75	167.50
Kometan	S8	1320-1810	78.96	118.90	33.33
Kurra Chine	S9	3200-4390	39.06	63.03	316.70
lower Fars	S10	460-890	13.03	28.65	238.40
Sarmord	S11	1480-2115	30.35	44.77	238.80
Mus	S12	2710-3670	35.51	33.36	292.70
Najmah	S13	2030-2690	17.74	15.59	44.38
Naokelekn	S14	2500-3190	72.71	116.4	73.60
Pilaspi	S15	550-1040	28	49.36	3.71
Qamchaqa	S16	1400-2080	22.78	41.22	44.69
Shiranish	S17	1270-1745	11.77	7.72	247.10
Singar	S18	825-1245	40.74	42.06	99.56
Surgelu	S19	2555-3240	91.31	153.8	128.4
Range			11.77-91.31	5.75-153.80	3.71-502.30
Mean			35.75	54.32	182.84
S.D.			22.50	38.47	158.73
World Standard			35	30	400

Materials and Methods

Sampling

In order to assess the natural radioactivity levels, about 20 core samples with cylindrical shape from four different drilling wells in the Erbil oil field have been collected, including (DD-3 and DD-5) originating from Demir Dagh block, (ZEG-1) originating from Zey Gawra block and (AAS-2) originating from Ain Al Safra block south of Erbil Governorate in Iraq according to the geological formations with different depths ranging from (65) to (4180) m as shown in Fig. 3. For each oil well, 20 samples were taken from different formations as shown in Table 1.

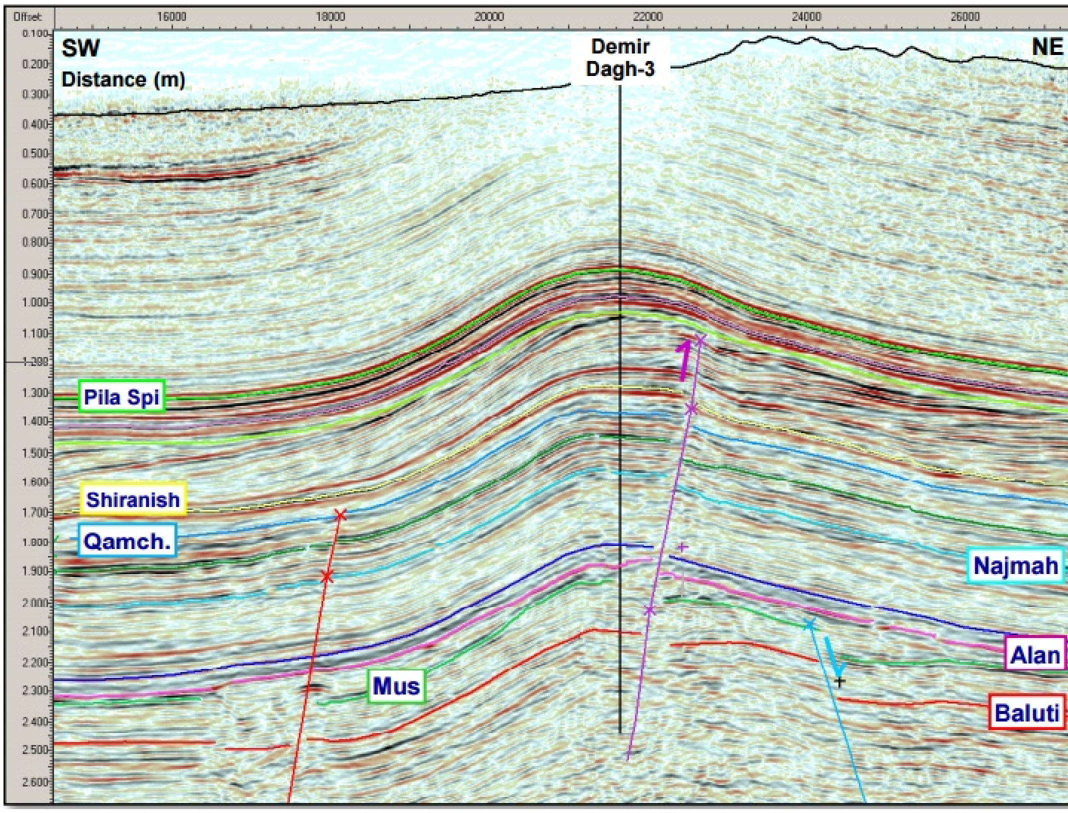

FIG. 3. Seismic section across Demir Dagh structure showing drilling oil well (DD3). (Oryx Oil Company).

The samples were crushed, dried for 24 hours in an oven at 110°C until going up to constant weight. In order to get homogenized samples, they were ground and passed through a mesh with a size of 2mm. The crushed and meshed samples were poured into a Marinelli beaker, sealed well to block radon gas leakage and stored for one month to ensure that radon and its daughters achieve a secular equilibrium before counting with NaI(TI) scintillation detector [5].

Counting and Spectra

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the samples were measured by a gamma ray spectrometer, SILENA (Model No. 3S3). This system had an NaI(TI) detector of (3×3) inch, connected to a multi-channel analyzer (512 channels) and analytical CASSY software. To reduce background radiation, the detector was surrounded by a cylindrical lead shield.

The measuring time for each sample was 21600 s. Energy calibration of the detector was performed by using ^{226}Ra standard source. The efficiency calibration was performed using ^{152}Eu, ^{137}Cs and ^{60}Co standard sources. The energy resolution of the detector is 7.4% for ^{137}Cs, at the energy of 662 keV. For the detection of ^{226}Ra, ^{232}Th and ^{40}K radionuclides, we used the line energy of 352keV, 583keV and 1461keV, respectively, as shown in Fig. 4.

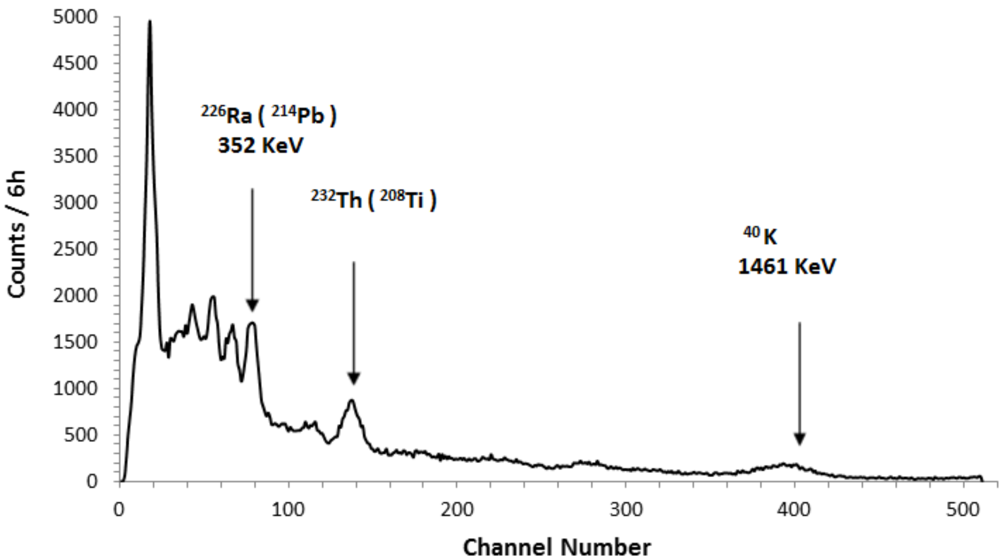

FIG. 4. The net gamma ray spectrum of Surgelu sample.

Activity Concentrations

The activity concentrations of the radionuclides in the samples were calculated using the following equation [8]:

$$A = \frac{C}{m_s.\varepsilon.p_\gamma} \quad (1)$$

where: A(Bq. kg^{-1}) is the specific activity of each radionuclide, C is the net count rate of the nuclide (counts.s^{-1}), Ɛ is the detector efficiency of the specific ɤ-ray, P_γ is the absolute transition probability of the specific ɤ-ray and m_s is the mass of the sample (kg).

Estimation of the Radiological Hazard Indices

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were used to measure the radiological hazard indices concerning the radiation exposure originating from the regional environment.

Radium Equivalent Index

Radium equivalent (Ra_{eq}) index in Bq.kg^{-1} is a suitable index to match the specific activities of samples consisting of different concentrations of ^{226}Ra, ^{232}Th and ^{40}K. It was defined with the assumption that 10 Bq/ kg of ^{226}Ra, 7 Bq/kg of ^{232}Th and 130 Bq/kg of ^{40}K produce the same gamma dose rate. It was measured by using the following relation [9]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (2)$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

Annual Gonadal Equivalent Dose (AGED)

Annual Gonadal Equivalent Dose (AGED) is a measure of the hazard to sensitive cells, like bone marrow, surface cells and gonads due to exposure to a certain level of radiation for particular activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K. AGED is measured by using the following equation [10]:

$$AGED(Sv/y) = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K \quad (3)$$

Absorbed Dose Rate (D)

The dose rate due to the concentrations of the radionuclides in the samples was used to measure the radiological risk originating from external exposure to radiation arising from naturally occurring radionuclides in the human environment. It was calculated by using the following equation [11]:

$$D(nGy/h) = 0.462C_{Ra} + 0.604C_{Th} + 0.0417C_K \quad (4)$$

External Hazard Index (H_{ex})

The external hazard index, H_{ex}, was widely used to estimate the indoor radiation dose rate due to external exposure to gamma radiation arising from natural occurring radioactive substances in the oil field by using the following equation [12]:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \quad (5)$$

Internal Hazard Index (H_{in})

The internal hazard index, (H_{in}), was commonly used to assess the internal radiation exposure of humans due to ^{222}Rn and its progenies. It is given by using the following expression [13]:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_K/4810 \qquad (6)$$

Gamma Radiation Representative Level Index (I_γ)

Representative level index is used to assess gamma radiation related with the natural radionuclides in the samples. The value of (I_γ) must be smaller than unity with respect to the insignificant radiation risk. It was measured by using the following equation [14]:

$$(I_\gamma) = C_{Ra}/150 + C_{Th}/100 + C_K/1500 \qquad (7)$$

Annual Effective Dose Equivalent (AEDE)

Annual effective dose equivalent is the gamma dose rate received by people (adults) living in the surveyed houses in their environment. Outdoor and indoor AEDE can be calculated by using the following relations [15]:

$$AEDE_{Out}(mSv/y) = D(nGy/h) \times 8760(h/y) \times 0.7(Sv/Gy) \times 0.2\ x10^{-6} \qquad (8)$$

$$AEDE_{In}(mSv/y) = D(nGy/h) \times 8760(h/y) \times 0.7(Sv/Gy) \times 0.8\ x10^{-6} \qquad (9)$$

The conversion coefficient is 0.7 Sv/Gy from absorbed dose in air to effective dose received by adults, 0.2 for the outdoor occupancy factor and 0.8 for the indoor occupancy factor.

Excess Lifetime Cancer Risk (ELCR)

ELCR is a measure of the potential carcinogenic effects that are characterized by estimating the probability of cancer incidence in individuals for a specific lifetime from projected intakes and exposures. It was calculated by using the following equation:

$$ELCR = AEDE \times DL \times RF \qquad (10)$$

where DL is the average life time (70 years) and RF is the risk factor (Sv^{-1}). ICRP 103 suggested the value of RF to be 0.06 for the public exposure (ICRP, 2007).

Results and Discussion

In this study, three naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in Erbil oil blocks core samples have been studied by using NaI(TI) gamma spectroscopy system. Table 1 shows the results of activity concentrations of Radium-226 ranging from (11.77) $Bq.kg^{-1}$ in Shiranish to (91.31) $Bq.kg^{-1}$ in Surgelu formation with a mean value of (35.75) $Bq.kg^{-1}$, while Th-232 ranged from 5.75 $Bq.kg^{-1}$ in Kolosh to (153.80) $Bq.kg^{-1}$ in Surgelu formation with a mean value of (54.32) $Bq.kg^{-1}$ and ^{40}K ranged from (3.71) $Bq.kg^{-1}$in Pilaspi to (502.30) $Bq.kg^{-1}$in Butmah Formation with a mean value of 182.84B $q.kg^{-1}$. The activity concentration values of ^{226}Ra indicate an excess of 47% over the world standard of (35 $Bq.kg^{-1}$) as shown in Fig. 5. The value of activity concentration of ^{232}Th was higher by 78.9% than the world standard of (30 $Bq.kg^{-1}$) as shown in Fig.(5). Also, ^{40}K indicated 15.7% excess as compared to the world standard value of (400 $Bq.kg^{-1}$). But, the mean values for all samples were lower than the maximum contamination levels as shown in Fig. 5.

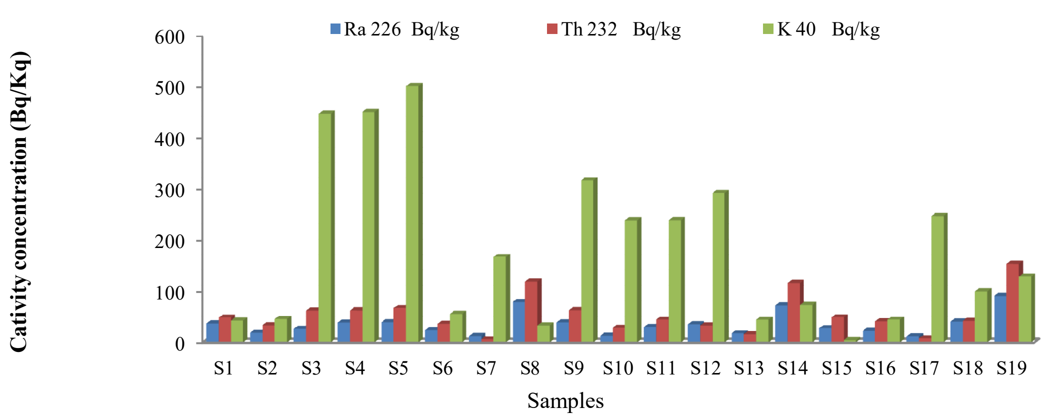

FIG. 5. Concentrations of natural radionuclides (226Ra, 232Th and 40K) in ($Bq\ kg^{-1}$) for each sample.

Table 2 presents the results of the radiological hazard indices. The value of radium equivalent due to the presence of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the samples ranged from (33.48) to (321.13) $Bq.kg^{-1}$ with an overall mean value of (127.50) $Bq.kg^{-1}$. The values of Ra_{eq} were less than the recommended maximum level of 370 $Bq.kg^{-1}$ in the soil. Thus, the soil in that area is suitable for agricultural purposes [17]. The second column in Table 2 represents the average annual gonadal equivalent doses (AGED), exceeding by 57.89% the maximum contamination level limit which is 300 mSv/y in all the samples as shown in Fig. 7. These results are in good agreement with the values reported in [14].

TABLE 2. The calculated hazard indices at Erbil oil field.

S.No.	Ra_{eq}($Bq.kg^{-1}$)	AGED (mSv/y)	I_γ	D (nGy/h)	Hazard index		AEDE(mSv/y)		ELCR $\times 10^{-3}$	
					H_{ex}	H_{in}	$AEDE_{out}$	$AEDE_{in}$	$ELCR_{out}$	$ELCR_{in}$
S1	110.55	333.14	0.77	48.56	0.29	0.38	0.06	0.24	0.24	0.95
S2	70.34	212.80	0.49	30.87	0.19	0.29	0.04	0.15	0.15	0.60
S3	150.36	483.75	1.10	68.67	0.40	1.89	0.08	0.34	0.34	1.34
S4	163.06	523.21	1.19	74.55	0.44	1.96	0.09	0.37	0.37	1.46
S5	174.37	560.95	1.27	79.82	0.47	2.17	0.1	0.39	0.39	1.56
S6	80.53	244.41	0.56	35.48	0.22	0.35	0.04	0.17	0.17	0.69
S7	33.48	114.82	0.25	16.17	0.09	0.72	0.02	0.08	0.08	0.32
S8	251.55	751.45	1.74	109.69	0.68	0.58	0.14	0.54	0.54	2.15
S9	153.58	483.60	1.10	69.32	0.42	1.45	0.09	0.34	0.34	0.60
S10	72.36	234.88	0.53	33.27	0.20	0.1	0.04	0.16	0.16	1.34
S11	112.76	355.90	0.81	51.02	0.30	1.1	0.06	0.25	0.25	1.46
S12	105.75	341.08	0.77	48.76	0.29	1.33	0.06	0.24	0.24	1.56
S13	43.45	133.92	0.30	19.46	0.11	0.27	0.02	0.1	0.1	0.7
S14	244.83	734.34	1.70	106.97	0.66	0.70	0.13	0.53	0.52	0.32
S15	98.87	294	0.68	42.90	0.27	0.18	0.05	0.21	0.21	2.15
S16	85.17	256.72	0.59	37.29	0.23	0.30	0.05	0.18	0.18	0.73
S17	41.85	146.26	0.32	20.41	0.11	1.02	0.03	0.10	0.10	0.40
S18	108.55	332.96	0.76	48.38	0.29	0.61	0.06	0.24	0.24	0.95
S19	321.13	965.35	2.23	140.44	0.87	1.02	0.18	0.69	0.69	2.75
Mean	127.50	394.92	0.90	56.95	0.34	0.91	0.07	0.29	0.28	1.12
World Standard	370	300.00	1.00	60.00	1.00	1.00	1.00	1.00	0.29	0.29
S.D.	77.10	230.78	0.53	33.59	0.22	0.61	0.04	0.16	0.16	0.66
Max.	321.13	965.35	2.23	140.43	0.87	2.17	0.18	0.69	0.69	2.75
Min.	33.48	114.82	0.25	16.16	0.090	0.18	0.02	0.08	0.08	0.32

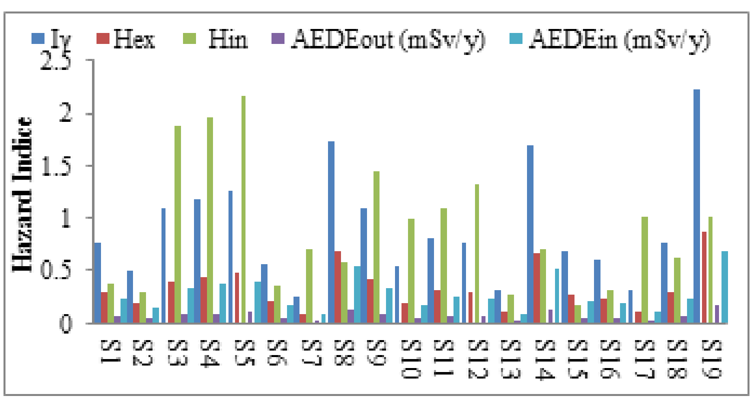

FIG. 6.The Radiological hazard indices: H_{ex}, H_{in}, I_γ, $AEDE_{out}$ (mSv/y) and $AEDE_{in}$ (mSv/y) for each sample.

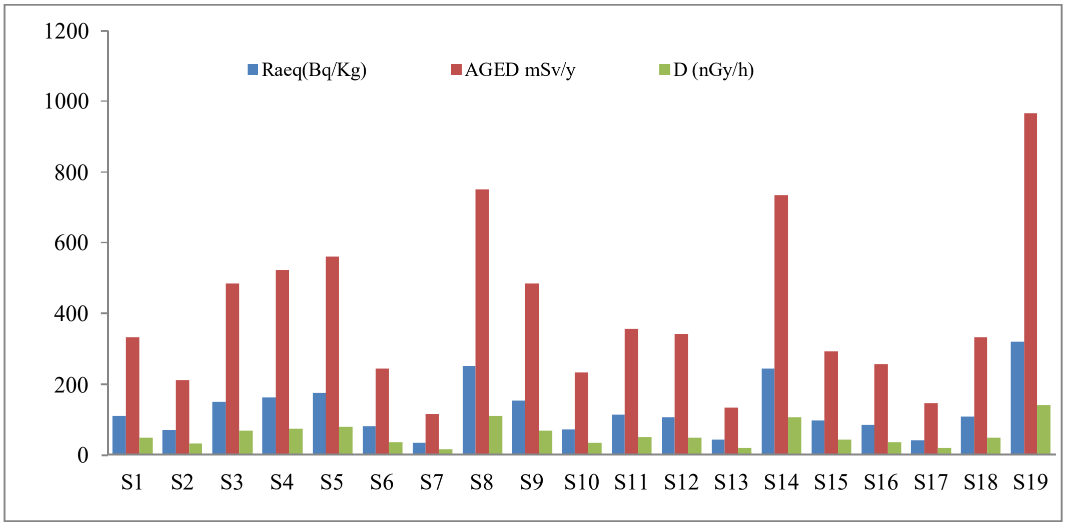

FIG. 7. Radium equivalent (Ra_{eq}), annual gonadal equivalent dose (AGED) and absorbed dose rate (D) for each sample.

The measured absorbed dose rate (D) ranged from (16.16 to 140.44) nGy/ h, with a mean value of (56.95) nGy/ h in the oil field, indicating that 36.8 % of the studied samples were with rates higher than the international recommended value of (60) nGy/h [11], while the mean value was less than that recommended level. The obtained external hazard index (H_{ex}) values ranged from (0.09) to (0.87), with a mean value of (0.34) in Erbil oil field. The measured mean value was lower than 1. The value of internal hazard index (H_{in}) varied from (0.18) to (2.7) with a mean value of (0.91). The results indicated that 42% of the studied samples were with values higher than the recommended value of (1) [11], but the mean value over all samples was lower than (1), as shown in Fig. 6.

The estimated values of I_γ for all the samples presented in Table 2 varied from (0.25) to (2.23) with a mean value of (0.90). The calculated values indicated that 31% of the studied samples were with values higher than the international value (I_γ > 1), but the mean value over all samples is lower than (1). The measured values of outdoor and indoor AEDE varied from (0.02) to (0.18) mSv/y and from (0.08) to (0.69) mSv/y, with mean values of (0.07) and (0.28) mSv/y for outdoor and indoor effective dose, respectively. The calculated values of $ELCR_{out}$ indicated that 42% of the studied samples were with values higher than the recommended value of (0.29). On the other hand, the values of $ELCR_{in}$ in all samples are higher than the maximum contamination level (0.29), as shown in Fig. 8.

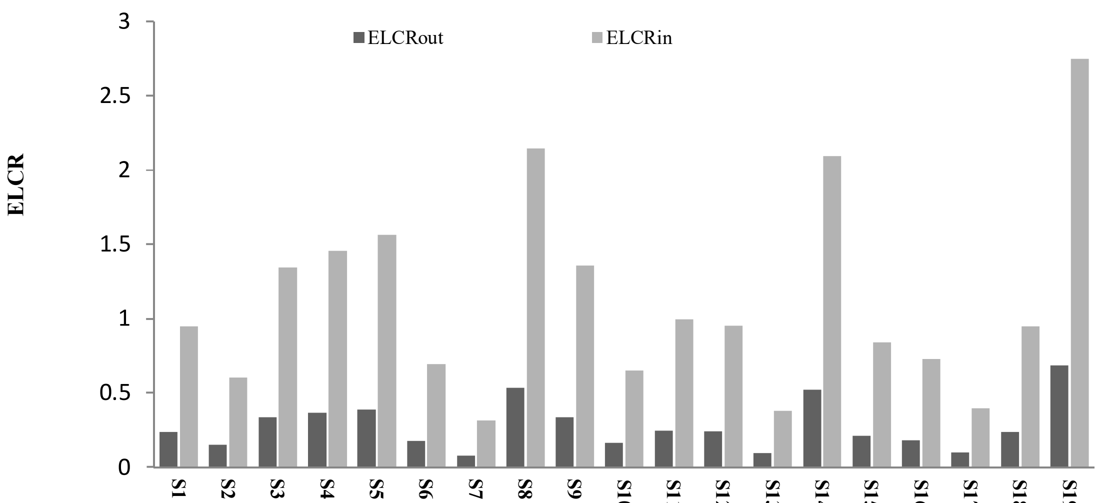

FIG. 8. Excess lifetime cancer risk for each sample.

Conclusions

The activity concentrations of ^{226}Ra and ^{232}Th radionuclides show highest values in the Surgelu sample due to its shale structure which contains high organic materials that accumulate heavy elements. Potassium-40 concentration was highest in the Butmah sample, which may refer to the high content of KCl compound in its rock structure. The evaluated radiological parameters and hazard indices show acceptable average values, except for the $ELCR_{in}$ values which exceed the permissible values for all samples. Although the measured hazard indices in the studied samples are lower than the permissible limits, they were still higher than the normal values. Hence, the oil and gas activities will influence negatively the radiological situation of the environment. It can be concluded that there are significant radiological hazards to the people in that area.

TABLE 3. Comparison of natural radioactivity levels in the samples to other countries.

Country	^{40}K(Bq. kg^{-1})	^{226}Ra(Bq. kg^{-1})	^{132}Th(Bq. kg^{-1})
Niger [8]	128.6	75.60	21.30
Yemen [18]	697. 9	59.39	71.32
Cameroon [19]	186.96	34.52	16.67
Egypt [11]	320	17	18
USA [11]	370	35	40
Malaysia [11]	331	67	82
Spain [11]	470	32	33
Present work	158.73	22.50	38.47

References

[1] United Nations Scientific Committee on the Effects of Atomic Radiation, (1988), Report to the UN General Assembly, with Annexes, United Nations.

[2] Mettler, F.A., Sinclair, W.K., Anspaugh, L., Selby, P.B., Edington, C., Webster, E.W., Harley, J.H., Wyckoff, H.O. and Ricks, R.C., Health Phys., 58 (1990) 241.

[3] Paschoa, A.S., "Radiological Impact Due to Oil and Gas Extraction", (Business Briefing: Exploration and Production, 2003), pp. 1-4.

[4] Isinkaye, M.O. and Emelue, H.U., Journal of Radiation Research and Applied Sciences, 8 (2015) 459.

[5] International Atomic Energy Agency (IAEA), A Guidebook, Technical Report Series No. 295, IAEA, Vienna (1989).

[6] Pitman, J.K., Steinshour, D. and Lewan, M.D., GeoArabia, Gulf Petrolink, Bahrain, 9(4) (2004) 41.

[7] Iraqi General Organization for Metrological Information, 2000. Climate Atlas of Iraq for the Years (1981-2000).

[8] Jibiri, N.N. and Ajao, A.O., Journal of Environmental Radioactivity, 78 (2005) 105.

[9] Beretka, J. and Mathew, P., Health Physics, 48 (1985) 87.

[10] Avwiri, G.O, Agbalagba, E.O. and Chad-Umoreh, Y.E, Journal of Environmental Radioactivity, 109 (2012) 64.

[11] UNSCEAR Report, United Nations Scientific Committee on the Effects of Atomic Radiation (Report to the General Assembly, with Annexes, 2000).

[12] Hamzah, Z., Saat, A., Mashuri, N.H. and Redzuan, S.D., The Malaysian Journal of Analytical Sciences, 12(2) (2008) 419.

[13] Cottens, E. and Radon, J. (Eds.), Proceedings of the Symposium on SRBII, Royal Society of Engineers and Industrials of Belgium, Brussels, (1990).

[14] Ali, H., Laith, A. and Abbas, M., Journal of Radiation Research and Applied Sciences, 9 (3) (2016) 259.

[15] UNSCEAR, (2010) Report to the UN General Assembly, with Scientific Annexes, Vol. 1 (United Nations, New York) 1-219.

[16] Anekwe, U. and Avwiri, G., American Journal of Scientific and Industrial Research, 7(3) (2016) 50.

[17] Shaban, H., Abd El-Hadi, K., Abd El-Bast, A., Imran, I. and Abdallah I., Journal of Medical Physics, 37(1) (2012) 1.

[18] Harb, S., El-Kamel, A.H., Zahran, A.M., Abbady, A. and Ahmed, F.A., International Journal of Recent Research in Physics and Chemical Sciences, 1(1) (2014) 1.

[19] Pascal, T., Thomas, B. and Robert, M., J. Environmental Protection, 3 (2012) 1571.