مجلة الدراسات التربوية والعلمية - كلية التربية - الجامعة العراقية
العدد العشرون - المجلد الخامس - علوم الفيزياء - كانون الأول 2022 م
doi.org/10.52866/esj.2021.05.20.05

Evaluation of Natural Radioactivity levels in Majnoon oil field, Basra, Iraq

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Abstract :

In this study, the concentrations of natural radioactivity nuclides and radiation hazard coefficients were measured in the Majnoon oil field. After the preparation of the samples, the radioactivity was measured using a gamma-ray system (high-purity germanium detector). The results showed that the highest value of radium concentration appeared in sediment samples with a value of (124.5 Bq/kg) and the lowest value of radium concentration in formation water samples with a value of (1.4 Bq/kg). The highest value of the concentration ^{228}Ac of the thorium series was (80.01 Bq/kg) in the sediment samples and the lowest value (1.9 Bq/kg) in the crude oil samples. While the maximum concentration ^{40}K is (106.4 Bq/kg) in sludge samples and the lowest value is (2.16 Bq/kg) in crude oil samples.

The results showed that the sediments contain the highest percentage of radioactivity values from the rest of the materials taken from inside the Majnoon oil field. It also has higher average values than what is allowed globally, so it is considered a source of radioactive pollution. The radiation risk factors for all materials taken from inside the Majnoon oil field showed that they are within the internationally permitted limits and do not cause any danger.

The results of NORM measurements in this study differed from those measured globally and locally. The reason may be the geological nature of the regions and the nature of oil.

Keywords: Majnoon oil field, the radiological hazard coefficients, NORM.

تقييم مستويات النشاط الإشعاعي الطبيعي في حقل مجنون النفطي، البصرة، العراق

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الخلاصة :

في هذه الدراسة، تم قياس تراكيز نويدات النشاط الإشعاعي الطبيعي ومعاملات الخطر الإشعاعي في حقل مجنون النفطي. تم اختيار 18 عينة مختلفة تمثل مراحل انتاج النفط الخام. بعد تحضير العينات، تم قياس النشاط الإشعاعي باستخدام منظومة اشعة كاما (كاشف الجرمانيوم عالي النقاوة). أظهرت النتائج أن أعلى قيمة لتركيز الراديوم ظهرت في عينات الرواسب بقيمة (124.5 بيكريل / كغم) وأدنى قيمة لتركيز الراديوم في عينات مياه التكوين بقيمة (1.4 بيكريل / كغم). كما ان أعلى قيمة للتركيز ^{228}Ac من سلسلة الثوريوم كانت (80.01 بيكريل / كغم) في عينات الرواسب وأقل قيمة (1.9 بيكريل / كغم) في عينات الزيت الخام. بينما أقصى تركيز ^{40}K هو (106.4 بيكريل / كغم) ظهر في عينات الحمأة وأقل قيمة (2.16 بيكريل / كغم) ظهرت في عينات الزيت الخام.

أظهرت النتائج احتواء الرواسب على أعلى نسبة من القيم الإشعاعية من باقي المواد المأخوذة من داخل حقل مجنون النفطي. كما أن لها قيمًا متوسطة أعلى من المسموح به عالميًا، لذا فهي تعتبر مصدرًا للتلوث الإشعاعي. أظهرت عوامل الخطر الإشعاعي لجميع المواد المأخوذة من داخل حقل مجنون النفطي أنها ضمن الحدود المسموح بها دوليًا ولا تسبب أي خطر.

اختلفت نتائج قياسات NORM في هذه الدراسة عن تلك المقاسة عالميًا ومحليًا. قد يكون السبب الطبيعة الجيولوجية للمناطق وطبيعة النفط.

Introduction

The oil and gas industry was classified among the industries that deal with materials containing naturally radioactive materials in its various fields, by-products, and wastes resulting from them. As a result, this industry was considered one of the potential sources of exposure facing its workers and the general public, which placed it under a regulatory framework that takes into account radiation protection standards in occupational and environmental exposure [1].

The oil and gas industry is one of the industries that use raw materials that contain NORM, which means the naturally occurring radioactive materials found in crude oil and its equipment. NORM includes long-lived radioactive elements such as uranium, thorium, and potassium and some radioactive decay products such as radium and radon. These elements are found on the earth and within the tissues of all living things. The main wastes containing NORM are widely found in crude oil and gas and their residues are from formation waters, sediments, sludge, and rocks [2].

The crude oil production process passes through several stages, resulting in solid and liquid wastes in large quantities containing natural radioactive nuclides. formation water, sediments, sludge, and rocks are among the most prominent NORM wastes [3]. Most studies were limited to measuring the NORM in sludge and rocks. The study aims to measure the natural radioactivity in the materials representing the stages of crude oil production and to know which quantities are more harmful to the environment and humans.

study area

Majnoon field is an oil field located in the east of Basra Governorate, near the Iranian-Iraqi border, 60 km from the city of Basra, and it is the third largest oil field in the world, with reserves estimated by the Iraqi Ministry of Oil at 12.6. billion barrels. It contains crude oil in a reservoir about 50 kilometers long and about 11 kilometers wide. It has an area of about 400 square kilometers. Figure 1 shows the location of the Majnoon oil field and the sampling location.

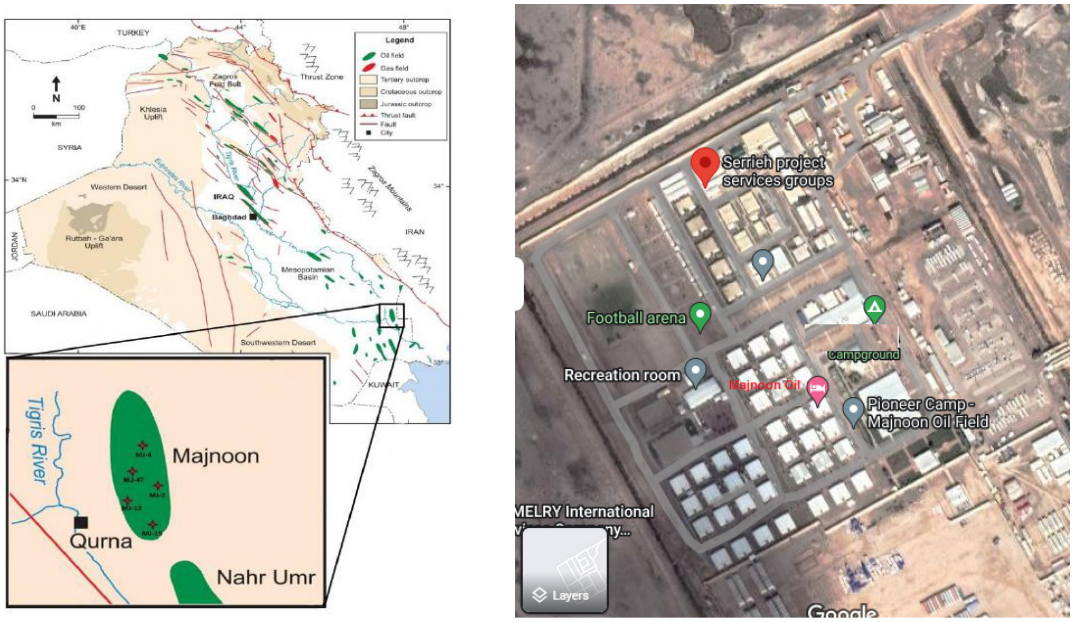

Figure 1: The location of the Majnoon oil field and the sampling location [4].

practical part

1. Samples Collection

The samples consist of several materials selected and collected from inside the Majnoon Oil Field. 18 samples were collected from different materials representing the stages of crude oil production. The samples included 5 samples of crude oil taken randomly from three different wells, 5 samples of sludge taken from oil accumulation laundries at separation stations, 5 samples of sediment collected from oil-water separation areas and 3 samples of formation water.

2. Sample preparation

Plastic containers with a capacity of 1 liter were used to collect the liquid samples and left for a month to obtain the radiological balance. As for the solid samples, they were collected in plastic bags with a capacity of one kilo. Then the samples were dried after being exposed to sunlight for a whole week, then they were crushed, sieved, and then placed in a Marinilli container, thus preparing the samples for measurement.

3. Detection and measurement system

A high-purity germanium detector (HpGe) was used to measure radionuclide concentrations. Its crystal size is 209 cm^3 and its diameter is 66 cm. It operates at an operating voltage of 1332 keV for a Co-60 source. The detector is cooled to a temperature of 77 K using liquid nitrogen.

The measuring system was calibrated using Cs and Co sources for the three energies 662 keV, 1173 keV and 1332 keV. Figure 2 shows the spectrum of standard sources.

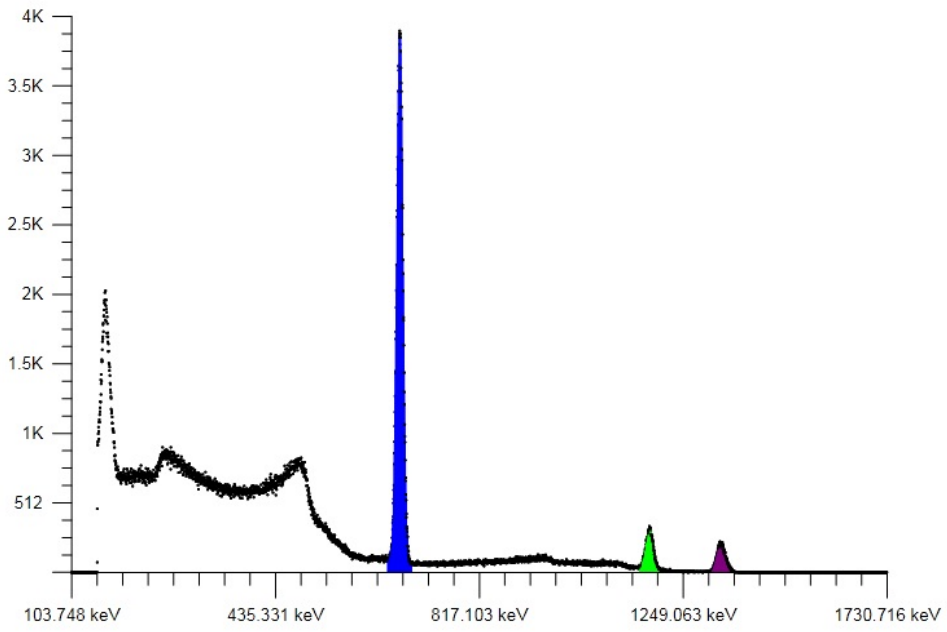

Figure 2: The spectrum of standard sources used in calibrating the system.

4. **Specific activity calculation for samples**

After the samples are placed in a Marinilli container, they are clamped around the germanium reagent. The gamma-ray spectrum is recorded for 10,000 seconds. the (Gene2000) program draws and analyzes the spectrum. From the information on the spectrum, the specific activity is calculated by the following equation (1)[5]:

$$dps = \frac{cps}{\varepsilon_0 w_0 I_\gamma} \dots\dots\dots (1)$$

Where (cps) is the net peak area of the selected energy, (ε_0) the efficiency of the detector for the selected energy, (I_γ) the relative intensity of gamma rays from special tables, (w_0) the mass of the sample [5].

Calculation of the radium equivalent radioactivity

is a popular metric for comparing the precise actions of materials containing ^{226}Ra, ^{232}Th, and ^{40}K by a single quantity that accounts for the radiation risks they pose [1]. It is calculated through the following equation (2):

$$Ra_{eq}(Bq/kg) = A_U + 1 \cdot 43 A_{Th} + 0 \cdot 077\, A_K \dots\dots\dots (2)$$

Where A_U . A_{Th} .A_K is the specific reactivity of uranium, thorium and potassium, respectively.

6. Calculation of the absorbed dose and the effective dose

The absorbed dose (D) in the air above one meter from the ground is calculated in units (nGy/h) and the ef-

fective dose (AEDE) in units (μSv/y) for the concentrations of uranium, thorium, and potassium using the equations (3,4) [6]

$$D(nGyh^{-1}) = 0 \cdot 429A_U + 0 \cdot 666A_{Th} + 0 \cdot 042A_K \quad \text{..... (3)}$$

$$AEDE(\mu Svy^{-1}) = D(nGyh^{-1} * 8760hy^{-1} * 0 \cdot 2 * 0 \cdot 7(svGy^{-1}) * 10^{-6} \quad \text{..... (4)}$$

7. Calculation of the hazard index (H)

The rates of exposure to external and internal radiation to gamma rays emitted from natural and industrial sources are measured through the hazard index (H), which was calculated by equation (5):

$$H = \frac{(S \cdot A)^{238}U}{185} + \frac{(S \cdot A)^{232}Th}{259} \cdot + \frac{(S \cdot A)^{40}K}{4810} \leq 1 \quad \text{..... (5)}$$

where $(S \cdot A)^{238}U.(S \cdot A)^{232}Th.(S \cdot A)^{40}K$ is the specific reactivity of the isotopes of uranium, thorium, and potassium, respectively.

When the value of the hazard index(H) is greater than one, it means that there is a risk and the dose is outside the internationally permissible limits. [6]

Results and discussion

A high-purity germanium detector system was used to analyze the spectrum of gamma rays emitted from the studied samples. By determining the energy of gamma rays, the element ^{226}Ra belonging to the uranium series, and the element ^{228}Ac belonging to the thorium series were diagnosed, and the radioactive element potassium was detected.

The results of the normal radioactivity of all samples taken from inside the Majnoon oil field are shown in Table No. (1). It is noted from the table that the highest values of radium concentration appeared in the sediment samples with a value of (124.5Bq/Kg) and the lowest values of radium concentration in the formation water samples with a value of (1.4 Bq/Kg). The maximum value of the ^{228}Ac concentration of the thorium series is (80.01Bq/Kg) in sediment samples and the lowest value is (1.9 Bq/Kg) in crude oil samples. While the maximum concentration of ^{40}k is (106.4Bq/Kg) in the sludge samples and the lowest value is (2.16Bq/Kg) in the crude oil samples. Since most radium is dissolved in formation water and transferred with the associated oily water before being deposited as salts with other elements like barium and is being enhanced upon special conditions (due to changes in temperature, pressure, acidity, etc.), especially with increasing salinity, it makes sense that radium has a maximum level in sediment samples collected from the final stage of water treatment. Radium is present in large amounts in scale and sludge [7,8].

Table 1: The normal radioactivity of samples taken from inside the Majnoon oil field.

Sample NO.	material type	specific activity (Bq/kg)		
		$^{622}_{\square}Ra$	$^{228}_{\square}Ac$	$^{40}_{\square}K$
O1	Crude oil	4.5	6.5	2.16
O2	Crude oil	3.9	1.9	4.7
O3	Crude oil	6.1	4.8	5.72
O4	Crude oil	5.17	3.12	3.8
O5	Crude oil	2.6	5.09	4.9
SL6	Sludge	30.86	9.3	106.4
SL7	Sludge	40.42	15.9	96.2
SL8	Sludge	9.9	10.8	88.4
SL9	Sludge	15.98	12.8	86.2
SL10	Sludge	10.35	14.4	78.9
S11	Sediment	124.5	74.3	58.3
S12	Sediment	95.2	65.6	50.1
S13	Sediment	109.6	80.01	32.8
S14	Sediment	80.5	25.1	49.9
S15	Sediment	98.9	34.2	40.1
W16	Formation water	3.2	7.01	15.3
W17	Formation water	2.5	7.5	9.9
W18	Formation water	1.4	8.08	8.9

Table No. 2 shows a comparison between NORM measurements in oil fields in several locations in Iraq and around the world. It is noted from the table that the values of the concentrations of elements $^{622}_{\square}Ra$, $^{228}_{\square}Ac$, $^{40}_{\square}K$ and differ from what has been measured globally and locally. The reason may be due to the geological nature of the areas and the nature of the oil. When the oil contains a high concentration of natural gas, it will lead to a high concentration of radium, which belongs to the $^{238}_{\square}U$ series[9].

Table 2: Comparison of NORM measurements in oil fields in several locations in Iraq and around the world.

Country	References	material type	specific activity (Bq/kg)		
			^{622}Ra	^{228}Ac	^{40}K
Oman	[10]	Crude oil	6380	2920	-
		Sludge	547	271	118
		Sediment	367	13	286
		Formation water	-	-	-
Romania	[11]	Crude oil	0.021-0.041	0.0002-0.007	0.2-0.83
		Sludge	3.7-59.2	0.05-0.12	710-1100
		Sediment	60-330	8-87	53-960
		Formation water	-	-	-
Austria	[12]	Crude oil	0.1-40	0.03-2	-
		Sludge	5-8 × 105	2-10	-
		Sediment	-	-	-
		Formation water	-	-	-
Iraq (Oil Field in North Region of Iraq)	[9]	Crude oil	38.27	38.62	22.37
		Sludge	75.73	74.40	6.19
		Sediment	492.2	629.7	368.0
		Formation water	-	-	-
Iraq (East Baghdad Oil)	[1]	Crude oil	2.3-5.8	-	2.8-36.0
		Sludge	1.8-38.1	-	12.1-242.1
		Sediment	252.4	-	120.0
		Formation water	1.2		52.8
Iraq (South Rumaila Oil Field)	[13]	Crude oil	33.6	-	197.0
		Sludge	312.8	-	502.7
		Sediment	-	-	-
		Formation water	20.3	-	66.4
Iraq (Majnoon oil field	(Present work)	Crude oil	2.6-6.1	1.9-5.09	2.16-5.72
		Sludge	9.9-40.42	9.3-15.9	78.9-106.4
		Sediment	80.5-124.5	25.1-80.01	32.8-58.3
		Formation water	1.4-3.2	8.08-7.01	8.9-15.3

The radium equivalent was calculated in all samples taken from inside the Majnoon oil field using equation (2). The results are shown in Table 3 and Figure 3. It is noted that the highest value is (235.238 Bq/Kg) appeared in sample S11 (a sample of sediment) and the lowest value is (6.979 Bq/ Kg) in sample O2 (a sample of crude oil). It is noted that all values were within the internationally permissible limits (370Bq/Kg) [14].

The risk factors resulting from exposure to NORM in all samples taken from inside the Majnoon oil field are shown in Table 3. It is noted from Table 3 and Figure 4 that the highest value of the dose absorbed in the air is (105.343 nGy/h) in sample S11 (a sample of sediment) because it contains high percentages of uranium and thorium among all the materials examined in the study, and the lowest value of (3.136 nGy/h) in Sample O2 (a sample of crude oil). When comparing these values with the values determined by UNSCEAR (59 nGy/h)[15], it is noted that all sediment samples were higher than the internationally permissible values.

The annual effective dose is calculated using equation (4) and shown in Table 3. It is noted from the table that the annual effective dose values follow the pattern

$$Sediment > Sludge > Formation\ water > Crude\ oil$$

These values appear acceptable because they are less than the universally permissible values (0.45 mSv/y)[15].

All values obtained in this study are acceptable in terms of radiological safety because they were less than one. According to what was calculated using equation (4) shown in Table 3.

Table 3: Radium Equivalent, Air Absorbed Dose, Effective Dose, and Hazard Index values for samples taken from inside the Majnoon Oil Field.

Sample NO.	material type	Raeq(Bq/kg)	AD(nGy/h)	AEDE(mSv/y)	Hex
O1	Crude oil	13.961	6.350	0.008	0.050
O2	Crude oil	6.979	3.136	0.004	0.029
O3	Crude oil	13.404	6.054	0.007	0.053
O4	Crude oil	9.924	4.455	0.005	0.041
O5	Crude oil	10.256	4.711	0.006	0.035
SL6	Sludge	52.352	23.902	0.029	0.225
SL7	Sludge	70.564	31.970	0.039	0.300
SL8	Sludge	32.151	15.153	0.019	0.114
SL9	Sludge	40.921	19.001	0.023	0.154
SL10	Sludge	37.017	17.344	0.021	0.128
S11	Sediment	235.238	105.343	0.129	0.972
S12	Sediment	192.866	86.635	0.106	0.778
S13	Sediment	226.540	101.683	0.125	0.908
S14	Sediment	120.235	53.347	0.065	0.542
S15	Sediment	150.894	66.890	0.082	0.675
W16	Formation water	14.402	6.684	0.008	0.048
W17	Formation water	13.987	6.483	0.008	0.045
W18	Formation water	13.640	6.356	0.008	0.041

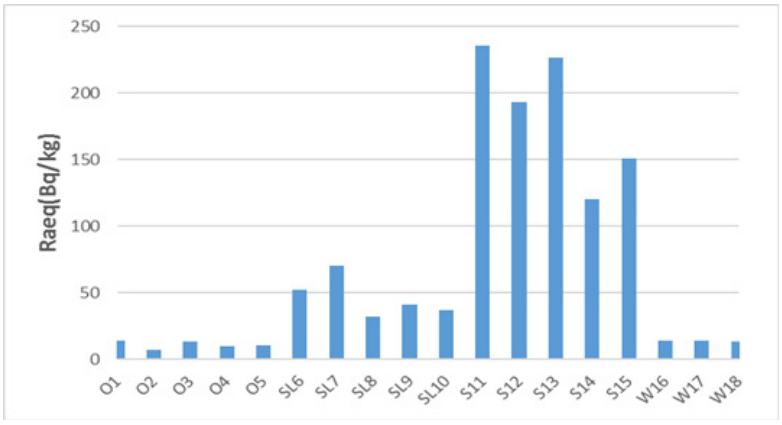

Figure 3 :
Radium equivalent values in all samples taken from inside the Majnoon oil field .

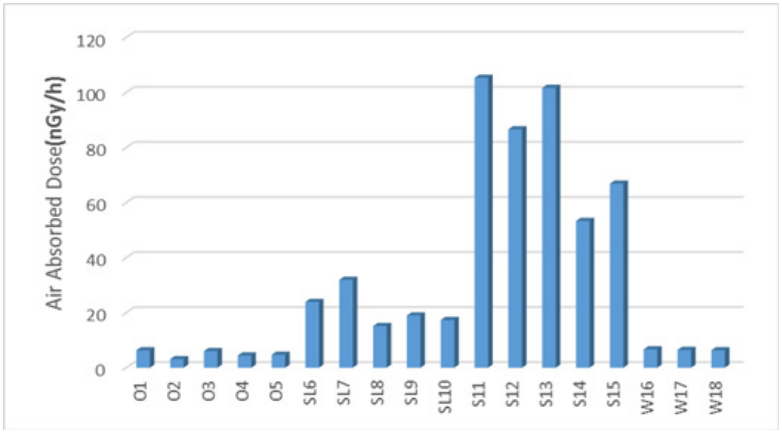

Figure 4 : The values of the dose absorbed into the air in all samples taken from inside the Majnoon oil field.

Conclusion

According to NORM measurements of samples taken from inside the Majnoon oil field, the study area is considered to have natural radioactivity and there is no radiological danger.

The sediments, which represent the last stages of the oil-water separation stage, contained the highest radioactivity than the rest of the materials taken from inside the Majnoon oil field. Also, its average values are higher than what is permitted globally, so it is considered a source of radioactive contamination and an increase in the amount of absorbed dose in the study area. As for the other samples, they all contain low concentrations of radioactivity, so they do not cause any danger.

The results of NORM measurements in this study differed from those measured globally and locally. The reason may be the geological nature of the areas and the nature of the oil. When the oil contains a high concentration of natural gas, it will lead to a high concentration of radium, which belongs to the ^{238}U series.

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