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Assessment of NORM from oil refineries and fields northwest of Mosul

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Abstract

The uranium concentration and radioactivity of radon gas were measured in Al-Kasik refinery and Ain Zala field using the CR-39 detector. Soil and water samples associated with the production stages of oil, sludge and crude oil were collected. The levels of uranium concentration in soil ranged from 0.703 to 1.480ppm, in water samples from 0.681 to 0.716 ppm, in sludge samples from 0.849 to 1.014 ppm, and in crude oil from 0.785 to 0.933 ppm. As for the radioactivity of radon gas, when comparing the radon rate in the samples we obtained with the global values, it was found that it falls within the internationally permissible limit, where the radon rate in the soil was 12.81 Bq/kg and when compared with the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) which has a value of 32 Bq/kg, and in the produced water it was 8.66 Bq/kg compared to (UNSCEAR)which has a value of 50 Bq/kg. In sludge samples 11.81 Bq/kg and when compared with the International Atomic Energy Agency (IAEA) whose value is$(8 - 5 \times 10^5)$ Bq/kg, and in crude oil samples 10.56 Bq/kg and when compared with the International Federation of Oil and Gas Producer (IOGP) whose value is $(800 - 4 \times 10^5)$ Bq/kg. As for the alpha ray hazard index, the results showed that it is within the permissible limits internationally, where the results were less than 1 and therefore does not pose a threat to the health of workers and environment.

Keywords: NORM ;Oil and Gas; Radioactivity; Produced water; Sludge

تقييم العناصر المشعة المتواجدة طبيعيا والناتجة من المصافي والحقول النفطية في شمال غرب الموصل

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الخلاصة

تم قياس تركيز اليورانيوم والنشاط الاشعاعي لغاز الرادون في مصفى الكسك وحقل عين زالة باستخدام كاشف $CR - 39$. جمعت عينات التربة والمياه المصاحبة لمراحل انتاج النفط والخبث والنفط الخام. وكانت مستويات تركيز اليورانيوم في التربة تتراوح من 0.703 الى 1.480 ppm وفي عينات الماء من 0.681 الى 0.716 ppm وفي عينات الخبث من 0.849 الى 1.014 ppm وفي النفط الخام من 0.785 الى 0.933 ppm. اما النشاط الاشعاعي لغاز الرادون عند مقارنة معدل الرادون في العينات التي حصلنا عليها

مع القيم العالمية وجد انها تقع ضمن الحد المسموح به عالميا ، حيث كان معدل الرادون في التربة 12.81 Bq/kg وعند مقارنته مع لجنة الامم المتحدة العلمية المعنية باثار الاشعاع الذري (UNSCEAR) التي قيمتها 32 Bq/kg، وفي الماء المنتج فكانت 8.66 Bq/kg مقارنة مع (UNSCEAR) التي قيمتها 50 Bq/kg. وفي عينات الخبث 11.81 Bq/kg وعند مقارنتها مع الوكالة الدولية للطاقة الذرية (IAEA) التي قيمتها $(8 - 5 \times 10^5)$ Bq/kg،وفي عينات النفط الخام 10.56 Bq/kg وعند مقارنتها مع الاتحاد الدولي لمنتجي النفط والغاز (IOGP) التي قيمتها $(800 - 4 \times 10^5)$ Bq/kg. اما موشر خطورة اشعة الفا فقد اظهرت النتائج انه ضمن الحدود المسموح بها عالميا حيث كانت النتائج اقل من 1 وبالتالي لاتشكل خطورة على صحة العاملين.

الكلمات المفتاحية: نورم ، النفط والغاز ، النشاط الإشعاعي ، المياه المصاحبة لانتاج النفط ، الخبث

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1. Introduction

Human beings are exposed to natural radiation that comes from two main source is cosmic rays that come from outer space and are produced by the glow of the sun and from external galaxies, as well as the natural terrestrial radiation, and the second source is the natural radioactive elements where these elements are widely spread in the earth's crust [1].

The term (NORM) used in the oil and gas industry stands for natural radioactive materials such as uranium and thorium. The radionuclides that are measured in the oil and gas industry are the result of the dissolution of two natural chains, uranium U238 and thorium Th232, where these elements have a long half-life and their presence in the soil depends on the geological structure [2].Natural radionuclides are present in oil and gas reservoirs in varying concentrations. These nuclides are released when oil and gas is extracted from the ground, to accompany oil and water to the surface of the earth, and move to production equipment such as pipelines and oil tanks, and settle inside them within the slag and crusts materials [3]. Workers in the oil and gas industry are exposed to radioactive materials during oil extraction, as well as exposure to mud, and water while drilling oil wells, in addition to that during equipment maintenance and tank cleaning. There are several studies and reports, related to NORM in the oil industry, where the International Atomic Energy Agency (IAEA) [4] has,published safety and security reports on dealing with NORM . Additionally ,the International Association of Oil and Gas Producers (OGP) [5] has, published clarifications about NORM in the oil and gas industry, and the United Nations Scientific Committee concerned with the effects of atomic radiation (UNSCEAR) [6] has published several reports on NORM and provided detailed sexplanation. In Egypt, researchers were able to study the level of NORM in the slag resulting from the oil industry and determine the level of its danger [7]. In Turkey, radium isotopes have been studied in waste and crude oil produced from the oil industry [8]. Our current study aims to measure the concentration of uranium and the radioactivity of radon gas, determine the danger of alpha that affects workers in the oil and gas industry, and compare the obtained results with the internationally permissible limit, including the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the International Atomic Energy Agency (IAEA), and the International Association of Oil and Gas Producers (OGP).

Study Area

The area of this study, as shown in Figure -1 includes the Alksak refinery, located 60 km northwest of Mosul, and shown Figure -2 the Ain Zala field, which is 120 km northwest of Mosul

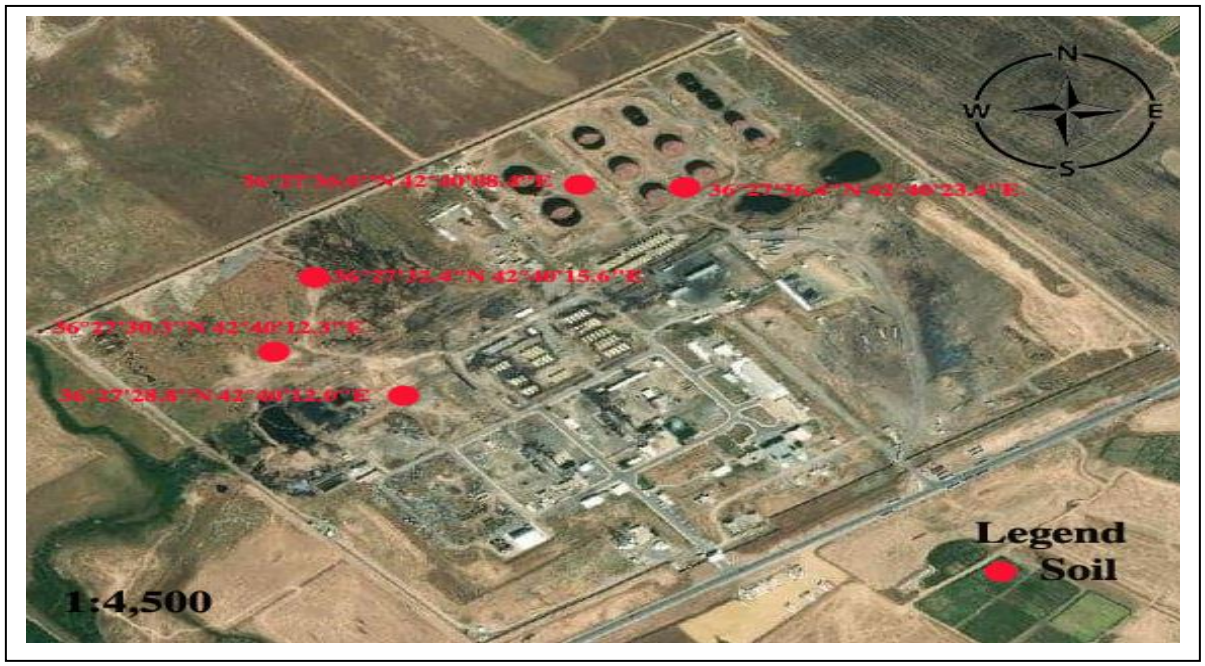

Figure - 1 A map shows the location of Alksak refinery

Figure - 2 A map shows the location of Ain Zala field

Samples Collection

two-Twenty different samples were collected from Ain Zala field and Al-Kasak refinery. The samples included soil, production water, sludge, and crude oil samples. Fifteen soil samples were collected from

two previously mentioned sites, whereas soil samples were collected from around the oil tanks and some of the site. In the Ain Zala field, samples were taken from around the oil wells. Soil samples were taken at a depth of 0-15 cm and stored in airtight plastic bags. As for slag, two samples were taken from the previously mentioned study sites, and sludge samples were taken from oil tanks and stored in plastic bags as well. Two samples of production water were also taken from a wet gas treatment unit and then placed into one liter (1 liter) plastic containers, and crude oil samples were taken from one of the tanks and stored in (1 liter) plastic containers with a label. The type of sample, the location, and the date the sample was taken is written on each sample .

Sample Preparation

Soil and sludge samples were exposed to a heat source for several hours in order to remove moisture from them. Impurities and gravel were removed from samples using a sieve of 2 mm diameter. Samples were grinded and a fine powder was obtained. Samples were weighed using a sensitive balance ,as for the production water and Crude Oil samples, they remained as they are. After that, all samples were placed inside plastic cups which were sealed to prevent the occurrence of interaction between the air inside the samples and their external surroundings, where the samples were left for 30 days to achieve the ideal balance between radium and radon, reaching 98% of the ideal equilibrium state. This time is calculated through the equilibrium relationship of radioactivity [9] [10]:

$A_{\mathrm{Rn}} = A_{Ra}(1 - e^{-\lambda Rn\, t})$

Where A_{Rn} effective means radon and $A_{Ra}$and represents the effectiveness of radium and λ_{Rn} represents the radon decay constant 0.1814 d^{-1} and t is the time required to reach equilibrium.

Measurements

The concentration of uranium and specific radioactivity of radon gas was measured for all samples using the solid state nuclear trace detector CR-39, where the samples were placed in irradiation chambers consisting of two plastic cups with identical size and shape, placing a filter paper between them and closing the mugs with adhesive tape tightly. The CR-39 detectors were left inside the chambers for 30 days [11] [12]. After that, the chemical 6.25 N NaOH erosive solution was prepared. At the end of the experiment, the reagents were removed and placed inside the chemical erosive solution and placed inside the water bath at a temperature of 70 $°C$ for 3 hours. The radiation background reached 180 $Tr.cm^{-2}$ and was subtracted from the intensity of the effects of alpha particles emitted from the samples under study. Microscopic observation was carried out using an optical microscope with a magnification of 400X . Figure 3 shows the irradiation system that was used.

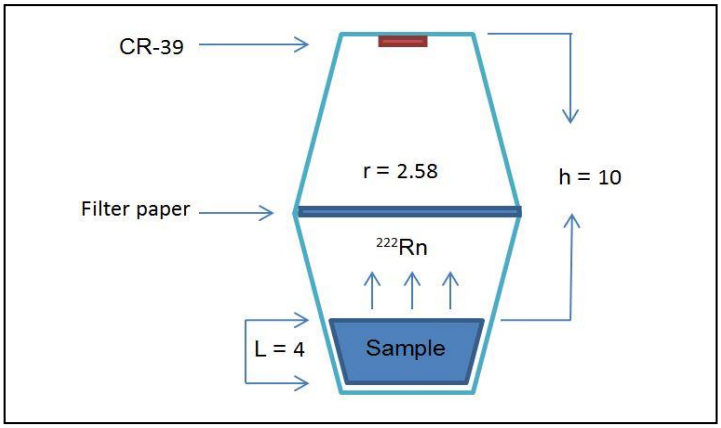

Figure 3 Irradiation system

The following equations numbered (1) to (7) were used for calculation :

1- Measure the concentration of radon gas through the following relationship [13]

$P = KC_aT$ ……. 1

Where P ($Tr.cm^{-2}$) represents tracks density, K ($Tr.\frac{cm^{-2}}{h^{-1}Bq.m^{-3}}$) represents the diffusion constant, T represents the irradiation time in hours, and C_a represents the radioactivity of radon in the air space in units ($Bq.m^{-3}$).

2- Determination of the propagation constant k [14]:

$K = \frac{1}{4}r\left(2Cos\theta - \frac{r}{R\alpha}\right)$ ……. 2

Where r represents the radius of the cup and is equal to $2.58\ cm$ and $R\alpha$ represents the range of alpha particles in the air produced by radon and is equal to $4.16\ cm$ and represents the θ angle of $35°$

3- Determining of radon concentration in samples (C_s) :

The radon concentration in the samples was calculated by the following relationship:

$C_S = h\,\lambda_{Rn}\,T\frac{C_a}{L}$ ……. 3

Where C_s represents the radon activity within the samples in units ($Bq.m^{-3}$), h represents the height of detector from the sample surface in cm units, T represents the irradiation time in days , λ_{Rn} represents the radon decay constant and L represents the thickness of the sample in cm .

4- The radioactivity of radon gas produced from samples (Bq) unit :

$A_{Rn} = C_SV$ ……. 4

$V = L\pi r^2$ ……. 5

Where A_{Rn} the radioactivity of radon gas in the samples(Bq) unit And V volume samples(m^3) and r^2 radius equal 2.58 (cm) .

5- Finding the number of radon atoms N_{Rn} in the samples :

$A_{Rn} = N_{Rn}\lambda_{Rn}$ ……. 6

After determining the number of radon atoms, the number of uranium atoms is found through the law of radiation balance

$\lambda_U N_U = \lambda_{Rn} N_{Rn}$ ……. 7

6- Determining the mass of uranium in the samples :

$W_U = N_U A_U / N_{av}$ ……. 8

Where W_U represents the mass of uranium in the samples, A_U represents the mass number of uranium, and N_{av} represents the number of Avogadro, N_U number of uranium atoms in the samples .

7- Determining the uranium concentration(CU) in samples in ppm units :

$ppm = \frac{W_U}{W_S}$ ……. 9

Where W_S represents the mass of the sample in (gm) .

Results and Discussion

Table 1 shows the location of the sample, its type, the sample weight, number of traces, and the radon concentration in the air and the samples. The results show that the highest radioactivity of radon gas and the concentration of uranium in soil, where the radon value soil samples in Alksak refinery were 18.06 Bq/kg , the uranium concentration was 0.941 ppm, while the lowest values of radioactivity were for radon gas and the concentration of uranium in Alksak refinery; the radon value was of 8.7 Bq/kg and the value of uranium concentration was 0.703 ppm. when comparing the results that were obtained with the permissible limits UNSEARS (32) Bq/kg [6] .As the results were within the permissible global limit, therefore they pose no risk to humans. In sludge samples, the highest radioactivity of radon gas (12.62 Bq/kg) and the concentration of uranium was in Ain Zala field 1.019 ppm, and the lowest radioactivity of radon gas and the concentration of uranium were in sample entry number 19 in table 1 for Ain Zala field with a value of 10.52 Bq/kg and uranium with a concentration of 0.849 ppm. The sludge results obtained with IAEA ($8 - 5 \times 10^5$ Bq/kg [1] show that the results are within the universally permitted limit. As for the crude oil sample, the highest radioactivity of radon gas and the concentration of uranium in Ain Zala field where the radon value was 11.56Bq/kg, uranium with a concentration of 0.785 ppm. The lowest radioactivity value of radon gas and the lowest concentration of uranium was in AlKasik refinery and the lowest concentration of uranium was in AlKasik refinery 9.73 Bq/kg for radon and 0.933 ppm for uranium concentration, compared to the permissible limit. For the OGP ($800 - 4 \times 10^5$) Bq/kg [15] , we find that it is within the universally permitted limit. As for the water samples associated with the production of sludge, the highest radioactivity value was for radon gas in Ain Zala field 8.88 Bq/kg, and the highest uranium concentration was in Ain Zala and uranium with a concentration of 0.716 ppm. The lowest radioactivity of radon and the uranium concentration was in Alksak refinery 8.44 Bq/kg and uranium at a concentration of 0.681 ppm When comparing the results we obtained with the permissible limit UNSEAR (50) Bq/kg [16] we find that what we obtained is within the permissible limits globally. As for the level of risk of alpha rays, all results were within the global permissible limit 1≤ and thus do not form a risk to the health of workers in the oil industry. Figure 4 shows the rate of radioactivity of radon in the samples, where it was found that the rate of radioactivity of radon in soil is 12.81 Bq/kg , water produced 8.66 Bq/kg and sludge 11.81 Bq/kg **,** and Crude Oil 10.65 Bq/kg

Table 1- the location of the sample, its type, the sample weight, number of traces, and the radon concentration in the air and the samples

NO	Location	Material	A_{Rn} (Bq)	A_{Rn} (Bq/kg)	$N_U \times 10^{17}$	$W_U \times 10^{-6}$	CU (ppm)
1	Alksak refinery	Soil	0.9372	11.67	1.913	75.630	0.941
2	Alksak refinery	Soil	1.4508	18.06	2.961	117.063	1.457
3	Alksak refinery	Soil	0.9941	12.55	2.435	96.267	1.215
4	Alksak refinery	Soil	1.2045	14.99	2.458	97.177	1.209

Table 2- the radioactivity of radon (Bq), the radioactivity of radon (Bq/kg), the number of uranium atoms, the mass of uranium in the samples, the concentration of uranium(ppm) in the samples, and the alpha index.

NO	Location	Material	Track $Tr.cm^{-2}$	C_s $Bq.m^{-3}10^3$	C_a $Bq.m^{-3}10^3$	Sample weight (gm)	The coordinates of the site
1	Alksak refinery	Soil	1402	0.824	11.2105	80.33	36°27'43.2"N 42°40'26.4"E
2	Alksak refinery	Soil	1809	1.063	17.3545	80.33	36°27'39.6"N 42°40'19.2"E
3	Alksak refinery	Soil	1487	0.874	11.8908	79.23	36°27'28.8"N 42°40'12.0"E
4	Alksak refinery	Soil	1801	1.059	14.4077	80.33	36°27'30.3"N 42°40'12.3"E
5	Alksak refinery	Soil	1046	0.615	8.3671	80.33	36°27'32.4"N 42°40'15.6"E
6	Alksak refinery	Sludge	1522	0.895	12.1765	81.00	36°27'35.0"N 42°40'20.7"E
7	Alksak refinery	water produced	987	0.579	7.877	78	36°27'32.4"N 42°40'15.6"E
8	Ain Zala field1	Soil	1759	1.034	14.0676	79.23	36°44'20.2"N 42°35'44.3"E
9	Ain Zala field1	Soil	1517	0.892	12.1357	79.23	36°44'18.1"N 42°35'47.5"E
10	Ain Zala field1	Soil	1784	1.049	14.2716	79.23	36°44'16.6"N 42°35'46.5"E
11	Ain Zala field1	Soil	2175	1.278	17.3872	79.23	36°44'17.8"N 42°35'51.2"E
12	Ain Zala field1	Soil	1805	1.061	14.4349	79.23	36°44'21.6"N 42°35'50.4"E
13	Ain Zala field2	Soil	1340	0.788	10.7207	78.23	36°43'28.8"N 42°35'32.3"E
14	Ain Zala field2	Soil	1670	0.982	13.3601	79.23	36°43'28.5"N 42°35'31.5"E
15	Ain Zala field2	Soil	1786	1.049	14.2716	78.23	36°43'29.6"N 42°35'30.2"E
16	Ain Zala field2	Soil	1410	0.829	11.2785	78.23	36°43'30.1"N 42°35'32.3"E
17	Ain Zala field2	Soil	1145	0.673	9.1562	78.23	36°43'28.4"N 42°35'31.2"E
18	Ain Zala field1	Sludge	1461	0.859	11.6867	77.40	36°43'14.1"N 42°35'34.9"E
19	Ain Zala field2	Sludge	1276	0.749	10.1901	81.00	36°43'15.0"N 42°35'39.6"E
20	Ain Zala field	water produced	1038	0.609	8.285	78	36°43'22.3"N 42°35'37.6"E
21	Ain Zala field	oil	1350	0.667	10.7888	78	36°43'12.5"N 42°35'37.6"E
22	Alksak refinery	oil	1135	0.793	9.0745	78	36°27'35.0"N 42°40'20.7"E

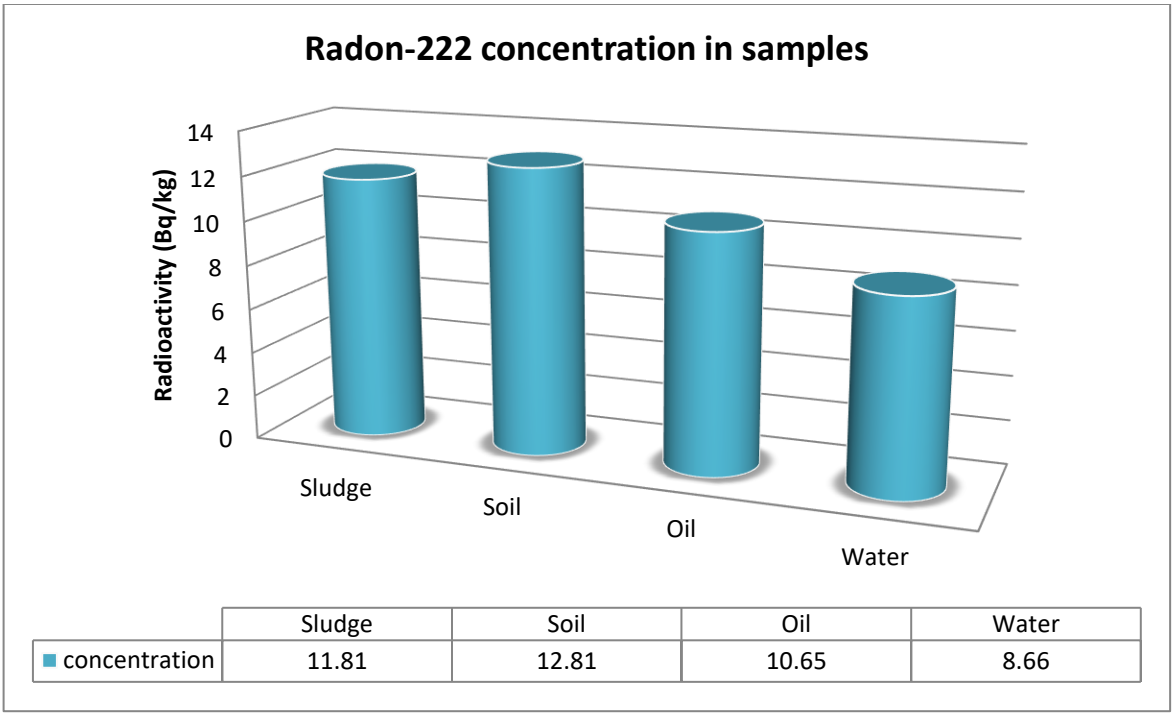

Figure 3 shows the rate of radioactivity of radon in the samples, where it was found that the rate of radioactivity of radon in soil is 12.81 Bq/kg , water produced 8.66 Bq/kg and sludge 11.81 Bq/kg **,** and Crude Oil . 10.65 Bq/kgThe radon rate obtained in the collected samples is within the permissible limits when compared with global values .

Table 3- Comparison of NORM concentration with unit Bq/kg of the current study with other studies

NO	Sample	$Rn_{222} - (Ra_{226})$	Refrence
1	Worldwide average(soil)	32	**[6]**
2	Oman NORM(Sludge)	547	**[17]**
3	China (Surface soil)	12.6	**[18]**
4	OGP (Crude oil)	$800 - 4 \times 10^5$	**[15]**
5	IAEA (Sludge)	$8 - 5 \times 10^5$	**[1]**
6	Iraq (sludge)	68.7-312.8	**[19]**
7	Iraq (Soil)	49.8-97.6	
8	Iraq (Produced water)	20.3-67.3	
9	Oil (Iraq)	18.6-33.6	
10	Iraq NORM (Sludge)	1.8-252	**[20]**
11	Iraq NORM (Crude Oil)	2.3-5.8	
12	Iraq (sludge)	6.8-14.4	
13	Iraq (Surface soil)	3.7-43.3	
14	Iraq (Formation water)	8.8	
15	Iraq (Soil)	8.71-18.35	**This work**
16	Iraq NORM (Sludge)	10.52-12.62	
17	Iraq NORM (Crude Oil)	9.73-11.56	
18	Iraq (Produced water)	8.44-8.88	
19	Worldwide average(Produced water)	50	**[16]**

Conclusions

The radioactivity of radon gas and the concentration of uranium were calculated in the samples collected from Al-Kasak refinery and Ain Zala field. The results showed that the average concentration of uranium for soil samples, produced water, slag and crude oil was within the internationally permissible limit. The level of danger of alpha rays was calculated. The results we obtained were less than 1 and therefore do not constitute any danger to the health of workers and the environment.

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