Naturally occurring radioactive materials and related hazard indices in Ahdeb oil field

Hadi D. Al-Attabi, Zainab Mohammed Hassan

Department of Physics, College of Science, University of Waste, Waste Iraq

E-mail: www.zainabh11@yahoo.com

Abstract

In this work, measurements of activity concentration of naturally occurring radioactive materials **(**NORM) isotopes and their related hazard indices for several materials such as crude oil, sludge and water in Ahdeb oil fields in Waste governorate using high pure germanium coaxial detection technique. The average values for crude oil samples were174.72Bq/l, 43.46Bq/l, 355.07Bq/l, 264.21Bq/l, 122.52nGy/h, 0.7138, 1.1861, 0.601 mSv/y, 0.1503mSv/y and 1.8361 for Ra-226, Ac-228, K-40, Ra eq, D, H-external and H-internal respectively. According to the results; the ratio between ^{238}U to ^{232}Th was 4, which represents the natural ratio in the crust earth; therefore, one can be strongly suggested that the geo-stricture of the Ahdeb oil fields dose not contents any kind of rocks. Although the results indicate the rising in the activity concentration of NORM isotopes, the national and international comparisons proved that it is still in the world range limits.

Key words

NORM isotopes, hazard indices, sludge, crude oil, Ahdeb oil field.

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النظائر المشعة الطبيعية ومعاملات الخطورة في حقل الاحدب النفطي

هادي دويج العتابي، زينب محمد حسن

قسم الفيزياء، كلية العلوم، جامعة واسط، واسط العراق

الخلاصة

في هذا البحث تم قياس تراكيز النظائر المشعة الطبيعية المنشأ ومعاملات الخطورة المرافقة لها لبعض المواد مثل النفط الخام والحمأة والماء في حقل الاحدب النفطي في محافظة واسط بأستخدام تقنية الجرمانيوم عالي النقاوة المحوري. متوسط القيم لعينات النفط الخام هي 174.72Bq/l و 43.46 Bq/l و 355.07Bq/l و 264.21Bq/l و 122.52 nG/h و 0.8317 و 1.1681 و 0.106mSv/y و 0.1503 mS/y و 1.8361 الى Ra-226 أو AC-228، K-40، مكافئ الراديوم، والجرعة، H-الخارجية، H-الداخلية، على التوالي. كانت النسبة بين U-238 إلى Th-232 هي 4، وهو ما يمثل النسبة الطبيعية في قشرة الأرض . وبالتالي، يمكن أن نستنتج أن تركيب حقل نفط الاحدب لا تحتوي على أي نوع من الصخور. على الرغم من أن النتائج تشير إلى ارتفاع في تركيز النشاط الاشعاعي للنظائر المدروسة في هذا البحث، أثبتت المقارنات الوطنية والدولية ان هذه القيم لا تزال في حدود القيم العالية.

Introduction

Naturally occurring radio nuclides are present at varying concentrations in the earth's crust and can be concentrated and enhanced by processes associated with the recovery of oil and gas. This ''enhanced'' NORM, often known as TENORM (Technologically-Enhanced Naturally Occurring Radioactive Materials) can be created when industrial activity increases the concentrations of radioactive materials or when the material is redistributed as a result of human intervention or some industrial processes, TENORM also can be the by-product or waste product of oil, gas and geothermal energy production. Sludge, drilling mud, and

pipe scales are examples of materials that can contain elevated levels of NORM, and the radioactive materials may be moved from site to site as equipment and materials are reused[1]. In addition, the oil can be classified as useful and good if it contained high concentration of natural gas, if not it called the dead oil. Further, there are many evidences that high concentrations of radon gas can be associated with the natural gas. Therefore, the measurements of the concentration of radon gas in the oil fields are important if one needs to evaluate the overall NORM and TENORM [1].

The Kut City (the capital of Wasit province) is located in eastern Iraq, on the left bank of the Tigris river, about 160 kilometers south east of Baghdad, between 32˚30'20"N latitude and of 45˚49'29"E longitude, with a population of approximately 374,000. Kut city depends on the oil fields, which are: the first of two oilfields Ahdab field and Badra and this study was limited to the Ahdab oil field that is an oil field located in the hand - Ahrar 27 km west of the Kut city. Estimated oil reserves of the field by more than a billion barrels. It started producing oil from the Ahdeb field in the July 1, 2011, and operated by China National Petroleum card production to 60 thousand barrels per day, and will increase production capacity to 120 thousand barrels per day within six months, and will be exported most of the production Ahdab field, but some of it will be used as fuel for power plants nearby to ease the shortage of electric power.

However, this paper dedicated to answer some important questions;

- What about the NORM and TENORM concentrations in Ahdeb oil fields?
- What about the values of radiological hazard indices and are these values within the allowed international limit?

All the results of the NORM measurements, which were measured in this work, can be categorized into ^{226}Ra, ^{228}Ac, and ^{40}K isotopes. Specific activity (SA) and its radiation hazard indices, which contented the gamma dose rate (D), the radium equivalent (Ra_{eq}), the external and internal hazard indices (H_{ex}, H_{in}), the representative level index, ($I_{\gamma r}$), and the annual effective dose equivalent (AEDE), were estimated.

In order to get accurate and confident results, measurements for each parameter such as S.A (for each isotope such as ^{226}Ra) have been done for three different samples, then the average of these measurements have been taken as a final result.

Collection of samples

A number of 26 samples of crude oil, sludge, corrosion, and ground water samples were collected form Ahdeb oil field. These samples were exposed to sunlight for an extended period of time were quite dry, while those that were recently removed from separation tanks or stored in barrels, tended to be more oily following collection.

Results

1-The crude oil samples

The overall results of the specifications for the investigated samples (the crude oil samples) have been presented in Table 1. The obtained average specific activity (SA) for ^{226}Ra, ^{228}Ac and ^{40}K were 174.72, 43.46 and 355.07 Bq/l. The maximum SA values of ^{226}Ra and ^{228}Ac were 212.30 and 52.42 Bq/l in O5 sample (the Crude oil taken from the well 32 of the layers of Musharraf oil field), while the maximum SA value of ^{40}K was 465.35 Bq/l in O13 sample (Crude-oil taken from the Rumaila layer of the first well). The minimum

SA values of ^{226}Ra, ^{228}Ac and ^{40}K were 126.37, 30.59, and 229.98 Bq/l in O1 sample (the Crude oil under Hydrostatic pressure). However, there is an important observation from the NORM results of these samples, which is the ratio between ^{226}Ra (which belongs to ^{238}U chain) and ^{228}Ac (which belongs to ^{232}Th chain). This ratio approximately have value near to the natural value, about 4, and this can be attributed to the nature of the Ahdeb land, which have a plane surface and do not have any kind of rocks. However, the differences in the activity concentrations of NORM isotopes of the crude oil samples can be attributed to the differences in the depths of oil wells. If the oil well has high depth, then the activity concentrations of NORM isotopes have large values. The average value of radium equivalent activities (Raeq) was 264.21Bq/l. This value is also considered to be normal [2], while the highest Raeq was 311.01 Bq/l for O13 sample (taken from the Rumaila layer of the first well) and the minimum Raeq was 187.82 Bq/l for O1 sample. The overall average value of gamma dose rate (D) for the full sample set was found to be 122.52 nGy/h which was 144.51 nGy/h for O13 sample that was higher than the permissible value and the minimum value was 86.97 nGy/h for O1 sample. The overall average values of the H-external and H-internal hazard indices (Hex and Hin), were 0.7138 and 1.1861 for Hex and Hin, respectively. The highest values of Hex and Hin were 0.8403 and 1.3861 for the O13 sample and the minimum values were 0.5075 and 0.8403 for O1 sample, respectively. The values of the Hex and Hin indices must be less than unity (≤1) for the radiation hazard to be negligible, [3]. So, it is noted that all values of Hex and Hin had difference values, and Hin had high values, which were more than the allowable limits (≥1), while Hex had normal values.

Table 1: The overall results of the crude oil samples.

The sample details	***The sample code***	*Ra-226 (Bq/l)*	***Ac-228 (Bq/ l)***	***K-40 (Bq/ l)***	***Ra eq (Bq/ l)***	***D (nGy/h)***	***H-external***	***H-internal***	***In Eff (mSv/y)***	***out Eff (mSv/y)***	***Iγr***
Crude oil under Hydrostatic pressure	O1	126.37	30.59	229.98	187.82	86.97	0.5075	0.8490	0.4266	0.1067	1.3017
Crude oil from the Moudud oil field layer Mi4	O2	180.09	44.02	310.50	266.95	123.49	0.7212	1.2080	0.6058	0.1514	1.8478
Crude oil from the layer of the first well of Moudud oil field	O3	174.73	43.19	342.90	262.90	121.85	0.7103	1.1825	0.5977	0.1494	1.8254
Crude oil from the layer of the second well of Moudud oil field	O4	181.63	45.04	309.60	269.88	124.79	0.7292	1.2200	0.6122	0.1530	1.8677
Crude oil taken from the well 32 of the layer Musharraf oil field	O5	212.30	52.42	289.54	309.56	142.71	0.8364	1.4102	0.7001	0.1750	2.1326
Crude oil taken from the well 47 of the layer Musharraf oil field	O6	195.93	49.14	296.32	289.02	133.39	0.7809	1.3104	0.6544	0.1636	1.9951
Crude oil taken from the well 55 of the layer Musharraf oil field	O7	184.36	45.06	273.91	269.89	124.58	0.7292	1.2275	0.6111	0.1528	1.8623

The sample details	*The sample code*	*Ra-226 (Bq/ l)*	*Ac-228 (Bq/ l)*	*K-40 (Bq/ l)*	*Ra eq (Bq/ l)*	*D (nGy/h)*	*H-external*	*H-internal*	*In Eff (mSv/y)*	*out Eff (mSv/y)*	*Iγr*
Crude oil from the Alkhaseeb oil field-layer kh2	O8	158.29	38.11	374.95	241.66	112.43	0.6529	1.0807	0.5515	0.1379	1.6863
Crude oil taken from the well first sequence dome Line 7 Line 1 sequence and be horizontal drilling of the Alkhaseeb layer	O9	148.94	37.23	438.57	235.95	110.22	0.6375	1.0400	0.5407	0.1352	1.6576
Crude oil from the well taking second dome Line 1 and Line 1 sequence and be horizontal drilling of the Alkhaseeb layer	O10	154.05	40.51	419.32	244.27	113.81	0.6599	1.0763	0.5583	0.1396	1.7116
Crude oil from the well taking second dome Line 3 and Line 3 sequence and have a horizontal drilling of the Alkhaseeb layer	O11	163.74	39.93	421.73	253.31	118.03	0.6844	1.1269	0.5790	0.1448	1.7721
Crude oil taken from the dome of the well 1 Line 13 and Line 5 sequence of drilling a horizontal Alkhaseeb layer	O12	157.32	40.33	398.58	245.68	114.35	0.6638	1.0890	0.5609	0.1402	1.7178
Crude-oil from the Rumaila layer: that the sample taken from the Rumaila layer of the first well	O13	201.95	51.21	465.35	311.01	144.51	0.8403	1.3861	0.7089	0.1772	2.1687
Crude oil taken from the well layer 46 of Rumaila	O14	193.71	48.29	437.39	296.44	137.72	0.8009	1.3245	0.6756	0.1689	2.0659
Crude-oil pre-treatment: a crude oil has not been any treatment or attic is separated from the gas or water process	O15	187.44	46.85	317.48	278.88	128.93	0.7535	1.2601	0.6325	0.1581	1.9298
Average		**174.72**	**43.46**	**355.07**	**264.21**	**122.52**	**0.7138**	**1.1861**	**0.6010**	**0.1503**	**1.8361**

The average value of the representative level index (Iγr) [4] for the crude oil samples was 1.8361. The highest value was 1.3017 for O13 sample and the minimum value was 2.1687 O1 sample. To estimate the annual effective dose rates, the conversion coefficient from absorbed dose (D) in air to effective dose with 0.7 Sv⁄Gy value, indoor occupancy factor with 0.8 value and outdoor occupancy factor of 0.2 proposed by UNSCEAR 2000 [5] were used. The average values of the annual effective dose equivalent (AEDE) due to terrestrial gamma radiation indoors and outdoors obtained for crude oil samples set to be 0.259 and 0.0648 mSv⁄y. The highest

values of E_(ied) and E_(oed)[6] (in mSv)/y were 0.7089 and 0.1772 in the O13 sample and the minimum values were 0.4266 and 0.1067 in the O1 sample.

2- Results of the sludge samples

Six samples of the sludge were collected from different Ahdeb oil field and the activity concentration has been measured. The overall results for the activity concentration and the hazard indices are presented in Table 2. The results of these samples can be classified as TENORM, because some of it deduced from refinery stations and the others precipitated when oil extracted.

Table 2: The overall results of the sludge samples.

The sample details	*Sample code*	*Ra-226 (Bq/kg)*	*Ac-228 (Bq/kg)*	*K-40 (Bq/kg)*	*Ra eq (Bq/kg)*	*D(nGy/h)*	*H-external*	*H-internal*	*In Eff (mSv/y)*	*out Eff (mSv/y)*	*Iγr*
Clays of drilling operations: or (drilling fluids) used in drilling water component of industrial materials and processes are added canonical	SL1	316.84	83.38	484.77	473.40	218.37	1.279	2.135	1.071	0.268	3.269
Clays of insulation operations: a sediment deposited in Insulators wet during oil processing operations and be in the form of fat (Sludge)	SL2	573.38	150.89	705.26	843.46	388.01	2.279	3.829	1.903	0.476	5.802
Dust from drilling the first well of residues 16-AD1	SL3	240.34	63.25	367.72	359.10	165.65	0.970	1.620	0.813	0.203	2.480
Dust from inside the well of the first 16-AD1	SL4	373.47	98.28	571.41	558.01	257.40	1.508	2.517	1.263	0.316	3.854
Waste oil	SL5	358.17	94.26	548.00	535.15	246.86	1.446	2.414	1.211	0.303	3.696
Extract water with oil residue	SL6	178.64	54.13	273.32	277.10	127.55	0.749	1.231	0.626	0.156	1.914
Average		**340.14**	**90.70**	**491.75**	**507.71**	**233.98**	**1.372**	**2.291**	**1.148**	**0.287**	**3.502**

The obtained data revealed that the average activity concentrations for ^{226}Ra, ^{228}Ac and ^{40}K for these set of samples had the values; 340.1, 90.70 and 491.75Bq/kg, respectively.
The maximum activity concentrations of ^{226}Ra ^{228}Ac and ^{40}K were 573.38, 150.89, and 705.26 Bq/kg appeared in the sample SL2 and the minimum concentrations were 178.64, 54.13, and 273.32Bq/kgappeared in the sample SL6.
The calculated hazard values, which can be estimated according to ref [6], of the sludge samples, can be illustrated as follow;

Radium equivalent (Ra_{eq}):
The average value of Ra_{eq} was 507.71Bq/kg , the maximum value was 843.46 Bq/kg for SL2 and the minimum value was 277.10Bq/kg for SL6.

Gamma dose rate:
The average value of gamma dose rate (D) for the full sample set was found to be233.98nGy/h. While the maximum value was 388.01 for SL2 sample and the minimum value was 127.55 nGy/h for SL6 sample.

External and internal hazard indices: The average values of H_{ex} and H_{in} were 1.372 and 2.291, respectively. The maximum values of H_{ex} and H_{in} were 2.279 and3.829 found in the sample SL2 and the minimum values were 0.749 and 1.231 found in the sample SL6. Through the obtained values of H_{ex} and H_{in}, one can note that all the H_{ex} and H_{in} values went over the allowable limits ($\geq$1) and, therefore, authorized person must be followed the radiation safety requirements.

Annual effective dose (in mSv/y):
The average values of the annual effective dose equivalent (AEDE) due to indoors (E_{ied}) and outdoors (E_{oed}) terrestrial gamma radiation were calculated to be 1.148 and 0.287mSv/y. The maximum values of E_{ied} and E_{oed} were 1.903 and 0.476mSv/yfor SL2 sample, and the minimum values were 0.626 and 0.156 for SL6 sample.

Representative level index, ($I_{\gamma r}$):
The overall average value of $I_{\gamma r}$ was 0.289, whereas the maximum value was 5.802 for SL2 sample and the minimum value was 1.914 for SL6 sample.
In order to establish the works of sludge samples, corrosion sample was taken from the tubes of oil and investigated against NORM isotopes (we do not illustrate the result of this sample with the sludge samples because it resulted from the chemical effects of oil on the tubes). The overall results for the measured corrosion samples have been presented in Table3.The obtained data revealed that the overall average value of the specific activity for ^{226}Ra, ^{228}Ac and ^{40}K of this sample were 473.95, 124.72 and 725.14 Bq/kg .
However, all the hazard indices of this sample showed high values comparing with other samples such as crude oil and sludge samples, as shown in Table3. Therefore, these results compel us to deal cautiously with the oil pipelines.

Table 3: The overall results of the corrosion sample.

The sample details	*Sample code*	*Ra-226 (Bq/kg)*	*Ac-228 (Bq/kg)*	*K-40 (Bq/kg)*	*Ra eq (Bq/kg)*	*D (nGy/h)*	*H-external*	*H-internal*	*In Eff (mSv/y)*	*out Eff (mSv/y)*	*Iγr*
corrosion of one of the oil pipes	C1	473.95	124.72	725.14	708.14	326.66	1.913	3.194	1.602	0.401	4.890

3- The water samples

As you know, that in the oil extraction process, the extracted oil was mixed with some quantities of ground water. Therefore, these waters must be refined in specific stations. However, measurements of NORM isotopes in these water samples give us some indications about the last fate of it and about the ability of how can one get rid these waters?

Four samples of the extracted ground water were collected from deferent Ahdeb oil fields and the activity concentration have been measured. The overall results for the activity concentration and the hazard indices have been presented in Table 4. The obtained data revealed that the average activity concentrations for ^{226}Ra, ^{228}Ac and ^{40}K for these set of samples had the values; 25.05, 7.59 and 35.17Bq/l, respectively.

Table 4: The overall results of the water samples.

The sample details	*Sample code*	*Ra-226 (Bq/ l)*	*Ac-228 (Bq/ l)*	*K-40 (Bq/ l)*	*Ra eq (Bq/ l)*	*D (nGy/h)*	*H-external*	*H-internal*	*In Eff (mSv/y)*	*out Eff (mSv/y)*	*Iγr*
Water extracted with oil	W1	14.36	4.35	21.97	22.27	10.25	0.060	0.099	0.050	0.013	0.154
Water treatment is 100%	W2	21.84	6.62	33.42	33.88	15.59	0.092	0.151	0.076	0.019	0.234
The second stage water separation	W3	13.70	4.15	8.36	20.28	9.26	0.055	0.092	0.045	0.011	0.138
Water Mkmena	W4	50.29	15.24	76.94	78.01	35.91	0.211	0.347	0.176	0.044	0.539
Average	***Average***	***25.05***	***7.59***	***35.17***	***38.61***	***17.75***	***0.104***	***0.172***	***0.087***	***0.022***	***0.266***

The maximum activity concentrations of ^{226}Ra, ^{228}Ac and ^{40}K were 50.29, 15.24, and 76.94 Bq/l appeared in the sample W4 and the minimum activity concentrations were 13.70, 4.15, and 8.36 Bq/l for W3 sample.

From another point of view, the health and environmental protection agencies have recommended safe limit of S.A of ^{226}Ra (as example) in drinking water for human beings; United States EPA has recommended 372 mBq/l of S.A of ^{226}Ra in water as safe limit, whereas WHO [7] has recently recommended 186 mBq/l as the safe limit for drinking purpose. However,

UNSCEAR 2000 [5] recommended safe limit as 111 mBq/l and ICRP [8] has recommended the safe limit as 23.56 mBq/l. These levels are set to represent a concentration that does not result in any significant risk to health over the lifetime drinking of water. Public community water supplies must comply with the maximum contaminated limits (MCL) recommended by these various National and International agencies. Hence, these results demonstrated that the authorized persons must get rid of from this water.
However, the hazard indices can be illustrated as follow;

Radium equivalent (Ra_{eq}): The average value of Ra_{eq} was 38.61 Bq/l , the maximum value was 78.01Bq/l and the minimum value was 277.10Bq/l for W3 sample.

Gamma dose rate: The average value of gamma dose rate (D) for the full sample set was 17.75 nGy/h. The maximum value founded in the W4 sample was 35.91 nGy/h and the minimum value founded in the W3 sample was 0.092 nGy/h.

External and internal hazard indices: The average values of the H_{ex} and H_{in} were 0.10 and 0.172, respectively. The maximum values of H_{ex} and H_{in} were 0.211 and 0.347 for W4 and the minimum values were 0.055 and 0.092 for W3.

Annual effective dose: The average values of the indoors and outdoors annual effective dose equivalent (AEDE) due to terrestrial gamma radiation for all samples of water samples were 0.087 and 0.022mSv/y, respectively. The maximum value ofE_{ied} was 0.176 mSv/y for W4 and the minimum value was 0.045 mSv/y for W3. Whereas the maximum value of E_{oed} was 0.044 mSv/y for W4 and the minimum value was 0.011 mSv/y for W3.

Representative level index, ($I_{\gamma r}$): The average value of $I_{\gamma r}$ for all water samples set was 0.226. The maximum value was 0.539 for W4 and the minimum value was 0.138 for W3.
Finally, NORM measurements in the oil fields were done in several sites in the world (one can be said in all oil fields in the world) therefore, if we compared our results with others, one can note that our results are still in the world range, as can be shown in Table5. In general, the world results showed that the ratio between the ^{238}U and ^{232}Th chains had different values ranged from the natural ratio (4) to different values, whereas our results had ratio about 4. These differences can be attributed to the nature of the ground and to the nature of the oil (if the oil has high concentration of natural gas, then the oil has high concentration of radon and, therefore, more radium, which belongs to ^{238}U chain).

Table 5: The specific activity measurements for some countries and for the present work.

State		Isotope	Crude Oil	Sludge
Oman	Al-Farisi, 2008 [9]		6380	547
		^{282}Ac	2920	271
		^{40}K	-	118
U. S. A.	George E.Pataki and Cahill, 1999 [10]	^{226}Ra	ND	22.2– 273.8
		^{282}Ac	ND-7.4	7.4-173.9
		^{40}K	ND	18.5-155.4
Romania	Elena Botezatu and Grecea, 2004 [11]	^{226}Ra	0.021-0.041	3.7-59.2
		^{282}Ac	0.0002-0.007	0.05-0.12
		^{40}K	0.2-0.83	710-1100
OGP	International Association of Oil and Gas Production, 2008 [1]	^{226}Ra	800- 4*10^{5}	50-8*10^{5}
		^{282}Ac	1-70	2-10
		^{40}K	-	-
IAEA	Pub. 1171, 2003 [12]	^{226}Ra	0.1-40	5-8 × 10^{5}
		^{282}Ac	0.03-2	2-10
		^{40}K	-	-
Iraq- Wasit Ahdeb Oil Field	Present work	^{226}Ra	174.72	340.14
		^{282}Ac	43.46	90.70
		^{40}K	355.07	491.75

Conclusions

According to the results, some remarkable conclusions can be listed;

1. For all NORM results, the ratio between ^{238}U to ^{232}Th was Approximately 4, which represents the natural ratio in the crust earth; therefore, one can be strongly suggested that the geo-stricture of the Ahdeb oil fields dose not contents any kind of rocks.
2. Although the results indicate the rising in the activity concentration of NORM isotopes, the national and international comparisons proved that it is still in the world range limits.

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