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Qualitative analysis of natural radioactive isotopes concentrations in crude oil from Saba and Al-Nasiriyah fields, Southern Iraq

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Abstract: The existence of naturally occurring radioactive materials (NORM) along with oil and gas creates a major risk to public health and the environment. Employees who have been extracting crude oil and the surrounding environment could be seriously affected if the amounts of these radioactive materials were higher than what is permitted globally. The current study was conducted to verify that the natural radioactive content of one of the fields in Thi Qar province, specifically the Saba oil field, is higher than that of the nearby fields in the same area. In this investigation, a gamma-ray spectrometry system was used based on a 3" x 3" NaI (Tl) scintillation detector to measure the natural radioactivity concentrations of 238U, 232Th, 226Ra, 228Ra, and 40K in crude oil samples taken from the Saba and al-Nasiriyah oil fields in Thi Qar province, Iraq. In the crude oil samples under examination, the highest activity concentrations of 238U, 232Th, 226Ra, 228Ra, and 40K were observed in the Saba field compared with the al-Nasiriyah field. Additionally, the maximum concentrations of the equivalent activity of 226Ra, 232Th, and 40K were also found in the Saba field. Moreover, the highest and lowest absorbed doses, which were calculated based on BECK, UNSCEAR, and ICRP, along with all other parameters such as annual effective dose (AED), the external and internal hazard indices, and lifetime cancer risks (ELCR), were identified to have maximum values in the Saba field compared to the al-Nasiriyah field. The results for every sample of crude oil showed that there were no serious radiation threats to the environment or the employees. Thus, all parameters that were measured and computed fall below the acceptable global limits recommended by the ICRP, WHO, and UNSCEAR.

Keywords: *Activity concentration, Crude oil radioactivity, Gamma-ray spectroscopy, Radiation hazard, Radionuclides.*

1. Introduction

A liquid that was originally made of hydrocarbons and consists of trace amounts of nitrogen, sulfur, and oxygen is called crude oil or petroleum [1]. The amount of crude oil differs significantly from field to field due to it is created underground and remains there until it is released onto the surface of the earth through a variety of natural processes, such as land cracks or fractures. Additionally, it is extracted through human activity, such as drilling wells and extracting the oil at a certain pressure or by external pumping [2]. Crude oil can exist in nature as liquid or gaseous substances, such as natural gas or crude oil, or as solid or semi-solid substances, such as an asphalt crater. Hydrocarbons, or replacement hydrocarbons, represent the majority of crude oil; the two main components are carbon (83–87%) and hydrogen (10–14%). There are three other substances that are less important, such as nitrogen (usually less than 0.1 and occasionally up to 2%), oxygen (up to 1.5%), and sulfur (0.1 to 3% and rarely up to 7%) [3, 4].

People are generally concerned about protecting themselves against ionizing radiation due to its numerous sources in daily life and its negative impacts on both the environment and humans [4]. One of the most important requirements for defending against ionizing radiation may be understanding what

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radiation is and how it impacts humans, in addition to being aware of the sources of industrial and natural radiation [5, 6]. Technologically Enhanced Natural Occurrence Radioactive Materials, or TENORMs, are another name for naturally occurring radioactive materials, or NORMs [7]. These terms are used to characterize naturally occurring radioactive elements that are found in crude oil and related equipment and contain (NORM) in the petroleum and natural gas industries. Several radioactive elements, such as potassium, uranium, and thorium, as well as some of the radioactive decay products of these elements, such as radium and radon [8, 9]. These elements are found in both the earth and the tissues of all living things. Naturally occurring radioactive material is frequently found in natural gas and oil as well as in its byproducts, which include sand, mud, soil, rock, coal, groundwater, metallic and nonmetallic minerals, fertilizers, and raw materials like phosphate and apatite a metal used as a source of phosphorus. It is found that NORM is associated with metallic ores, including those for tin, niobium, rare earth elements, some copper, and gold [10, 11].

The aim of the present study is to verify that the natural radioactive content of one of the oil fields of Thi Qar province, which is the Saba field, is higher than the nearby oil fields in the same area. Furthermore, enhancement the implementations of radiological safety standards.

2. Materials and Methods

2.1. Experimental Detail

2.1.1. Activity Concentration

The activity concentration, measured in (Bq/L). Using the following Equation 1 [12]:

$$A(Bq/L) = \left(\frac{N}{t.I_\gamma(E_\gamma).\varepsilon(E_\gamma).m}\right) \times DCO \qquad (1)$$

Where (N) is the net area under the peak, (m) represents the weight of the sample in kilograms, I_γ (E_γ) is the intensity, (t) is measurement time, $\varepsilon(E_\gamma)$ refers to the efficiency, and (DCO) is the density of the crude oil [13].

2.1.2. The equivalent activity concentration

The equivalent activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K (Ra_{eq}, Th_{eq}, and K_{eq}) in (Bq/L) respectively, can be calculated by using the following Equations according to [14]:

$$Raeq=ARa+1.43ATh+0.077AK \qquad (2)$$

$$Theq=ATh+0.7ARa+0.055AK \qquad (3)$$

$$Keq=AK+18.46ATh+13.24ARa \qquad (4)$$

Where A_{Ra}, A_{Th}, and A_K are the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K respectively.

2.1.3. Gamma Ray Absorbed Dose (D_γ)

The absorbed dose's term represents the amount of energy that is a unit of mass absorbs in the radiation-exposed body, which can be calculated by several mothed including the recommendations of ICRP Smith, et al. [15]; Beck, et al. [16] and UNSCEAR [17] respectively in (nGy/h) as following:

$$D_{\gamma(ICRP)} = 0.427A_{Ra} + 0.662A_{Th} + 0.043A_K \qquad (5)$$

$$D_{\gamma(Beck)} = 0.420A_K + 0.429A_{Ra} + 0.666A_{Th} \qquad (6)$$

$$D_{\gamma(UNSCEAR)} = 0.533A_{Ra} + 0.827A_{Th} + 0.0537A_K \qquad (7)$$

2.1.4. The external (H_{ex}) and Internal (H_{in}) Indices

The external and internal hazard indices determine the potential dose levels that workers in sites with radiation activity may be exposed to, which are calculated using the Equations 8 and 9 [18, 19]:

$$H_{ex}\left(\frac{Bq}{L}\right) = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \qquad (8)$$

$$H_{in}\left(\frac{Bq}{L}\right) = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \qquad (9)$$

2.1.5. Excess Lifetime Cancer Risk (ELCR)

Excess Lifetime Cancer Risk (ELCR) is a measure used to estimate the probability of an individual developing cancer over their lifetime due to exposure to radiation, beyond normal background levels. The total number of cancer cases that may occur by annual effective dose given by the following Eq. (10):

$$ELCR = AEDxDLxRF \qquad (10)$$

Where (DL) is the estimated life expectancy, and (RF) refers to the hazard of fatal harm per Sievert [20, 21].

2.1.6. The Annual Effective Dose (AED)

In order to identify the annual effective dose of the gamma-emitting substances, UNSCEAR2000 [22] suggested a conversion coefficient (0.7 Sv. Gy^{-1}) to convert the absorbed dose in air to the annual effective dose received by humans. Thus, the annual effective dose can be calculated as the following Equation 11.

$$AED(mSv/y) = D_{\gamma}(nGy/h)x10^{-6}x\,8760h/y\,x0.2x0.7\,Sv/Gy \qquad (11)$$

The number (8760) refers to the hours per year, where the (0.2) represents the occupational factor. The annul effective dose must be less than the world average of (0.458) mSv/y.

2.1.7. Index of Representative Level ($I_{\gamma r}$)

The Index of Representative Level is an indicator utilized to estimate gamma radiation exposure from natural radioactivity in materials created by Organization for Economic Co-operation and Development (OECD). It refers to the levels of radionuclides (^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{40}K) and assists determining if radiation concentrations are within acceptable safety limits. If Iγr is 1 or below, the material is generally considered safe. Equation 12 can be used to determine this factor [23]:

$$I_{\gamma r(OECD)} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \qquad (12)$$

2.1.8. Minimum Detectable Activity

Detecting low concentrations of NORM requires determining the minimum detectable activity (MDA), which is based on the background radiation and the presence of gamma rays from other radionuclides. To determine the (MDA), Equation 13 is used:

$$MDA = \frac{(4.66x\sigma_b)+3}{\varepsilon(E_\gamma).I_\gamma(E_\gamma).W.t} \qquad (13)$$

Where (σ_b) is the standard deviation, ε(Eγ) represents the efficiency of the spectroscopy system, Iγ(Eγ) refers to the intensity of the targeted radionuclide energy, (W) is the weight of the sample in kilograms, (t) is the measurement time for the sample. This equation is applicable when the sample and background counting times are the same [24, 25].

2.2. Sample Preparation and Measurements

Twenty crude oil samples were collected and prepared from the two crude oil fields, each weighing around (1.5) liter. The samples were placed into the containers for one month in order to get the radioactive equilibrium. Every sample was approximately weighed to fill a one-liter container (Marielle Becker) and counted for 18 hours. Using a 76 mm x 76 mm Teledyne isotope NaI (Tl) scintillation detector with a 7.5% keV resolution at 661.76 keV Cs-137 source gamma-ray spectroscopy system as shown in Figure 1, the samples were measured and analysis to obtained qualitative of naturally occurring radioactive materials concentrations. In the current study, four methods were used to calibrate the measurement system to obtain the highest possible accuracy, namely calibrating the energy to its location (channel), the detector resolution, the experimental efficiency, and the minimum detectable activity. The minimum detectable activity for the targeted radionuclides was measured and calculated as illustrated in Table 1. After determining the Minimum Detectable Activity (MDA), Equation 1 was applied to calculate the activity concentration of the targeted radionuclides. Based on the activity concentrations of ^{238}U

(measured through the gamma emissions of ^{214}Bi at 1120.3 keV and ^{226}Ra at 609.3 keV) and ^{232}Th (measured through the gamma emissions of ^{208}Tl at 583 keV and ^{228}Ac at 911 keV), as well as the single gamma energy of ^{40}K at 1460.8 keV, the radionuclide concentrations were calculated to assess the radioactive content.

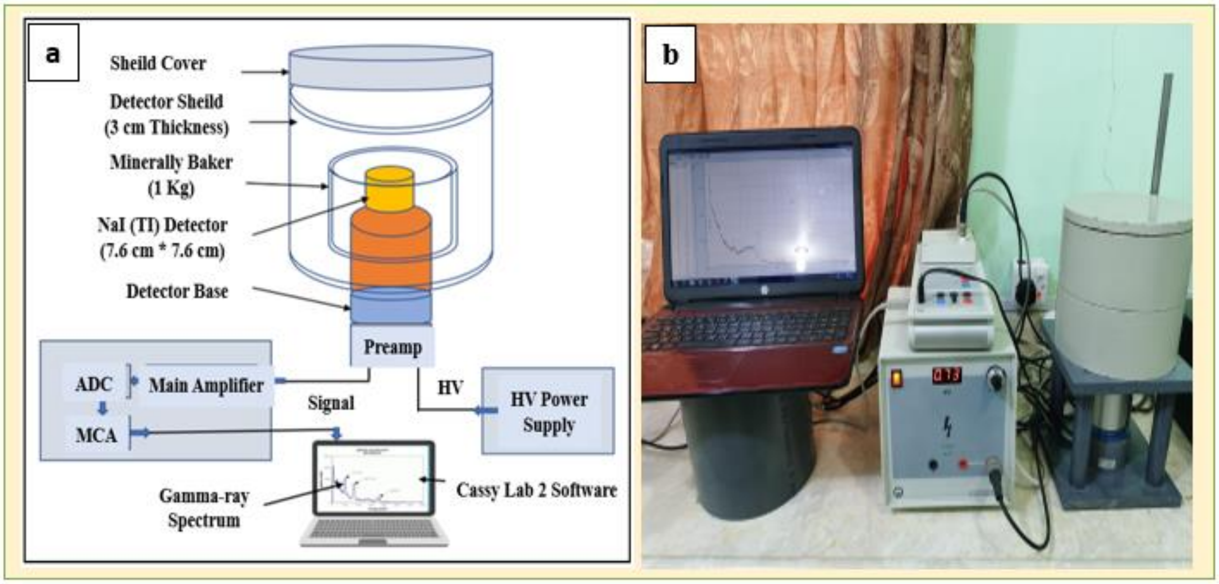

Figure 1.
a) Diagram of experimental setup of gamma spectroscopy system used in the present study. b) An image of the gamma spectroscopy system used in the present study.

Table 1.
Minimum detection activity (MDA) measured in present Study.

Parent nuclide	Daughter nuclide	Energy (keV)	Abundance (%)	$\varepsilon(E_\gamma)$	MDA (Bq)
$^{40}_{19}$K	Natural	1460.8	10.66	0.021	9.328
$^{226}_{88}$Ra	$_{83}$Bi^{214}	609.3	45.49	0.065	0.112
$^{238}_{92}$U	$_{83}$Bi^{214}	1120.3	14.7	0.0276	0.410
$^{232}_{90}$Th	$_{81}$Tl^{208}	583.2	85	0.089	0.106
$^{228}_{88}$Ra	$_{89}$Ac^{228}	911.2	25.8	0.0459	0.114

3. Results and Discussion

The activity concentrations of targeted natural radionuclides ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{40}K were obtained from crude oil samples that were taken from (20) locations of the Saba and al-Nasiriyah oil fields. The highest activity levels with an average of natural radionuclides for ^{238}U and ^{232}Th from the two oil fields were illustrated in Figure 2. While the lowest activity concentrations with their averages were determined in Figure 3. In Figure 4, the highest levels of activity concentrations for ^{226}Ra and ^{228}Ra of the Saba and al-Nasiriyah oil fields were clearly indicated, but the lowest activity concentrations of the two oil fields were illustrated in Figure 5. Moreover, Figure 6 demonstrated the highest and the lowest levels of activity concentrations with an average of ^{40}K for both oil fields. The maximum values of the highest and the lowest levels of activity concentrations were found in the Saba oil field in comparison with the al-Nasiriyah field. The reason behind having the Saba oil field the maximum values in both cases may be due to the fact that it has the highest level of salinity as illustrated in the database of Thi-Qar Oil Company and the research [26]. While al-Nasiriyah oil field has the lowest level of accompanied salinity with crude oil, which means that high natural radionuclides concentration will be found in the Saba oil field.

Overall, based on a comprehensive comparison of the measured activity concentrations with the data that were reported in David [27] as shown in Table 2, it can be concluded that the concentrations in both oil fields pose no significant risk of radiation exposure to workers or the environment because they fall within the limits that are supported by the World Health Organization (WHO), the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), and the International Commission on Radiological Protection (ICRP). The equivalent concentrations of ^{226}Ra (Ra_{eq}), ^{232}Th (Th_{eq}), and ^{40}K (K_{eq}) were determined, and the results were presented in Table 3 for both oil fields. In the Saba oil field, the highest levels of equivalent concentrations for ^{226}Ra, ^{232}Th, and ^{40}K were illustrated in the sample (SFT1), which were greater than the ones that were indicated in the al-Nasiriyah oil field in the sample (NFT1). Moreover, the lowest levels of equivalent concentrations in the Saba field for the same elements were presented in the sample (SFW1) which were also greater than the ones that were found in the al-Nasiriyah oil field in the sample (NFW1). All the equivalent concentrations in both oil fields are within the allowable limitations recommended by UNSCEAR, ICRP, and WHO. However, the Saba oil field has the maximum quantities for the highest and the lowest levels and it can be clearly shown in Figure 7 in comparison with the al-Nasiriyah oil field. The Saba oil field still has the highest level of equivalent concentration. In both oil fields, the absorbed doses of natural radionuclides D_{ICRP}, $D_{UNSCEAR}$, and D_{BECK} were calculated in nGy/h. The outcomes have been determined and displayed in Table 4. The highest and the lowest levels were demonstrated in the Saba field and both of them also have the maximum values compared with the al-Nasiriyah oil field for the same reason as illustrated in the activity concentrations. The external (H_{ex}) and internal (H_{in}) hazard indices were calculated, which are displayed in Table 5. The highest and the lowest levels were observed in the Saba oil field in the sample (SFT1), which still has a greater concentration than the al-Nasiriyah oil field. According to the obtained results, the external hazard level is less than the internal hazard level, but hazard indices in both oil fields are lower than the unity of the globally permissible level, showing that dealing with crude oil in both fields is still safe for both employees and the worksite. The excess lifetime cancer risk (ELCR) was determined and showed that the Saba oil field in samples (SFT1) and (SFW2) had the highest and lowest concentrations as determined in Table 5, but the al-Nasiriyah field had the highest and the lowest once as found in samples (NFT1) and (NFW11). The annual effective dose (AED) for ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{40}K was computed, in Saba field, especially in the sample (SFT1) the highest value was found, while sample (SFW2) had the lowest once, but al-Nasiriyah crude oil samples had the minimum levels comparing with the Saba oil field. The current study showed that all samples had a result that was below the global limitations. Hence, Table 5 included the representative level index I_{yr}. The highest and lowest representative level indices were also indicated in the Saba field compared with the al-Nasiriyah field.

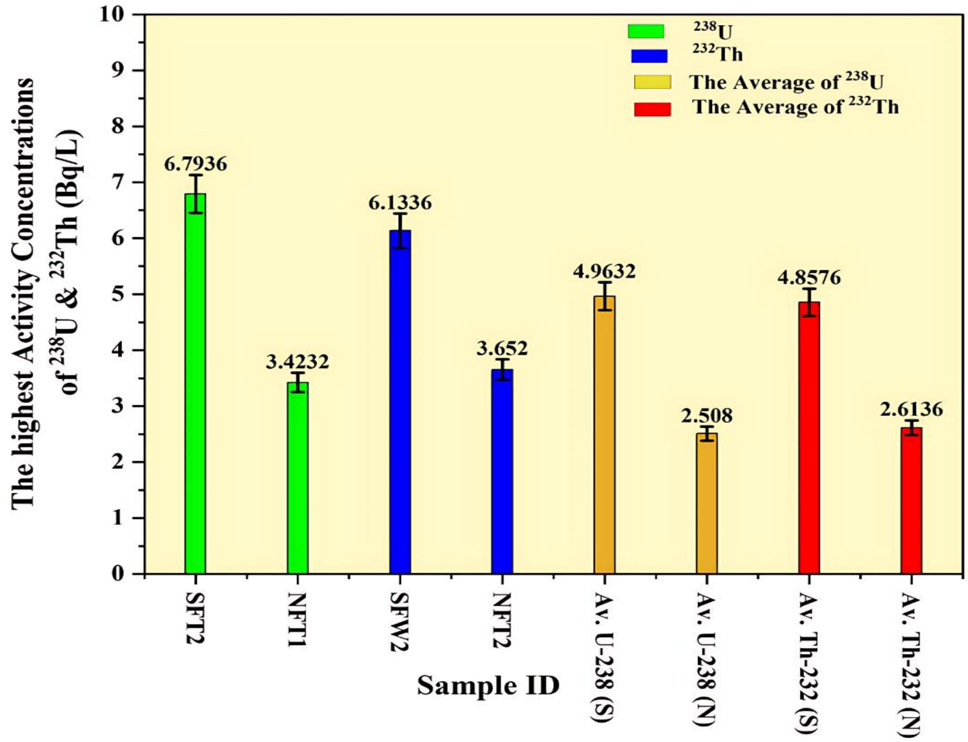

Figure 2.
The highest activity concentrations of ^{238}U and ^{232}Th (Bq/L) in Saba and al-Nasiriyah oil fields.

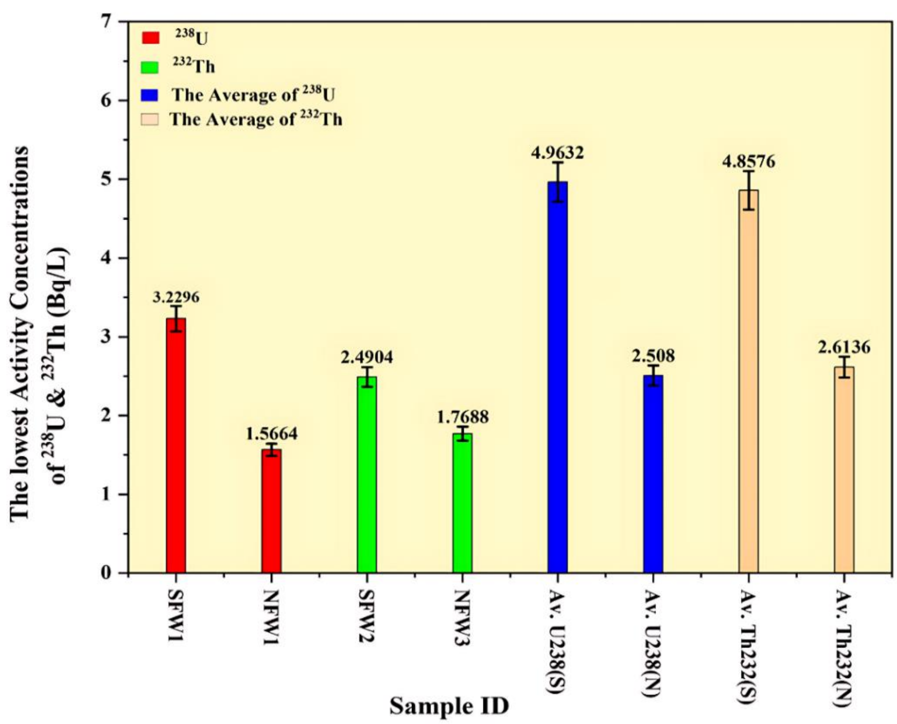

Figure 3.
The lowest activity concentrations of ^{238}U and ^{232}Th (Bq/L) in Saba and Al-Nasiriyah oil fields.

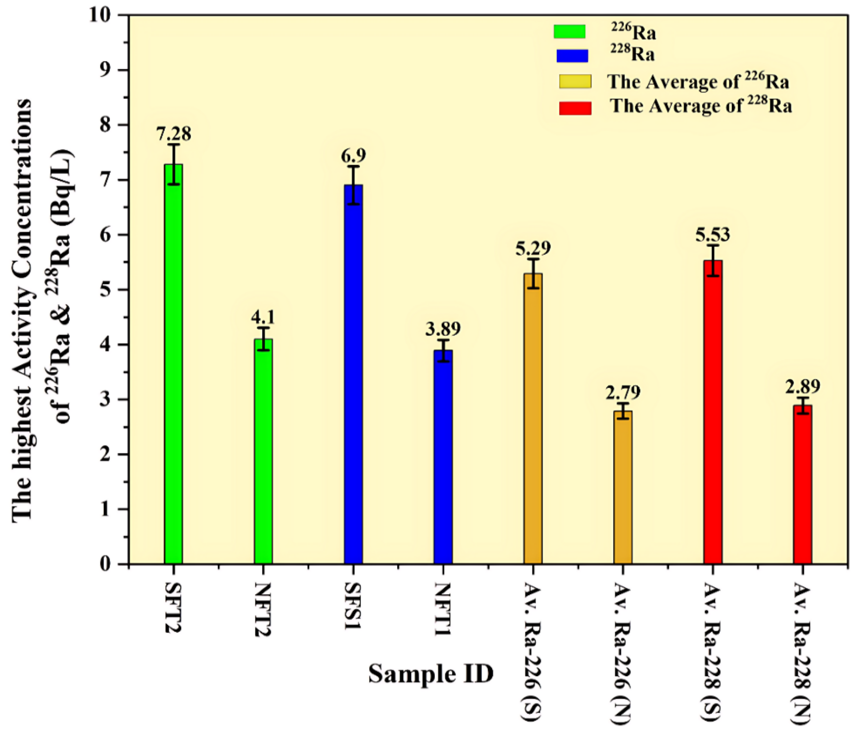

Figure 4.
The highest activity concentrations of ^{226}Ra and ^{228}Ra (Bq/L) in Saba and al-Nasiriyah oil fields.

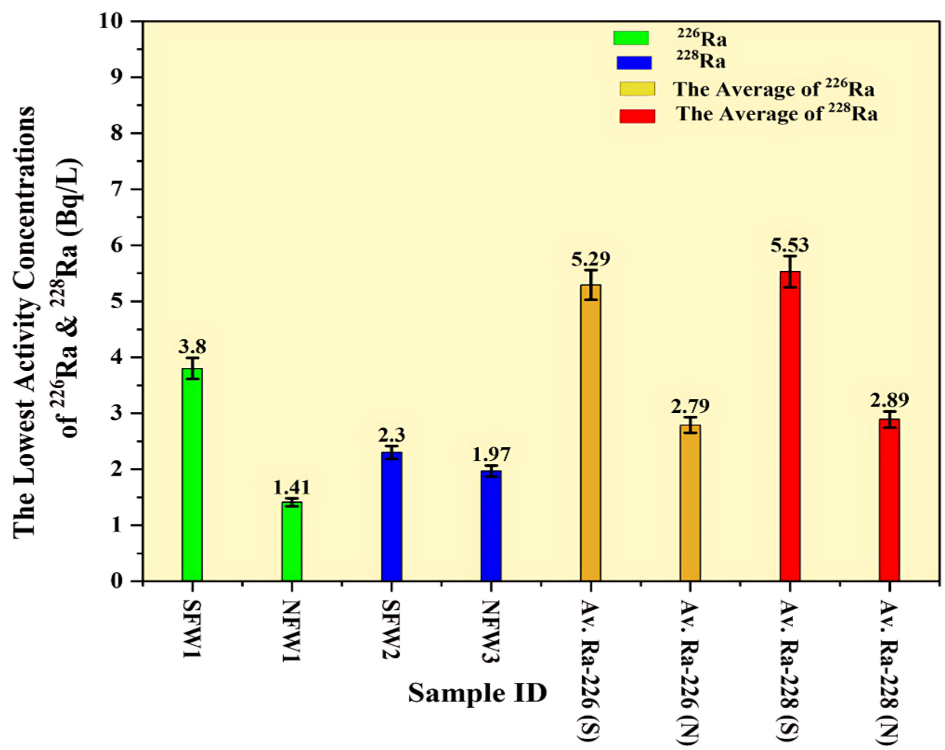

Figure 5.
The lowest activity concentrations of ^{226}Ra and ^{228}Ra (Bq/L) in Saba and al-Nasiriyah oil fields.

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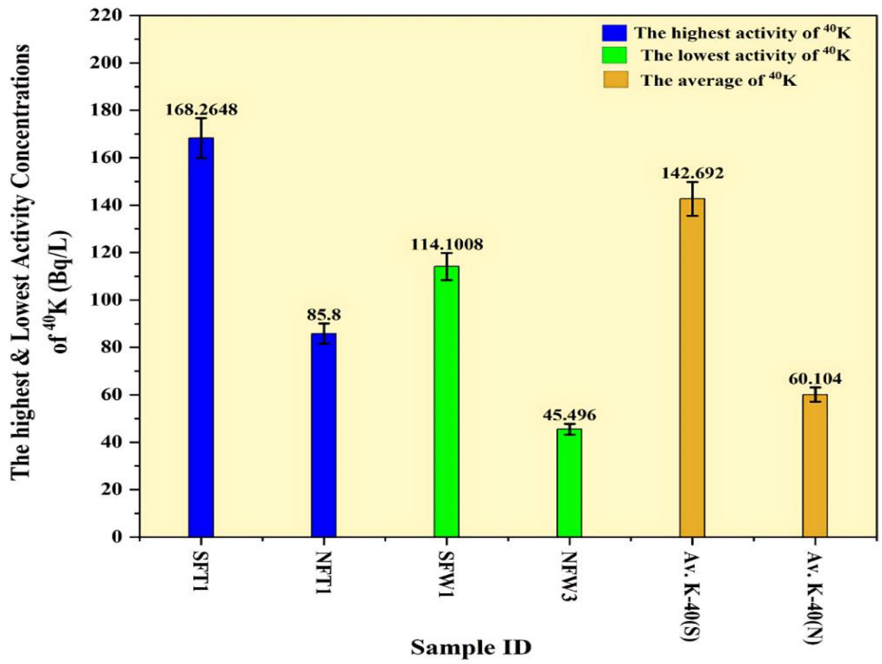

Figure 6.
The highest and lowest activity concentrations of ^{40}K (Bq/L) in Saba and Al-Nasiriyah oil fields.

Table 2.
Natural radionuclides in Saba and Al-Nasiriyah oil fields measured and compared with the concentrations in studies which reported globally.

Radionuclides	Activity concentrations (Bq /L) [14]	Saba activity concentrations (Bq /L) (Present work)	Al-Nasiriyah activity concentrations (Bq /L) (Present work)
^{238}U	0.0001 - 10	3.229 – 6.793	1.584 – 3.44
^{232}Th	0.03 - 20	2.49 – 6.133	1.768 – 3.652
^{226}Ra	0.1 - 40	3.344 – 6.406	1.240- 3.608
^{228}Ra	0.05 - 20	2.024 – 6.072	1.733 – 3.423
^{40}K	1.0 - 623.0	114.1 – 168.26	45.49 – 85.8

Table 3.
The equivalent concentrations of ^{226}Ra, ^{232}Th, and ^{40}K (Ra_{eq}, Th_{eq}, and K_{eq}) in (Bq/L).

Saba oil field				Nasiriyah oil field			
Sample ID	**Ra_{eq}**	**Th_{eq}**	**K_{eq}**	**Sample ID**	**Ra_{eq}**	**Th_{eq}**	**K_{eq}**
SFW1	16.496	11.67	214.74	NFW1	7.567	5.35	98.37
SFW2	17.461	12.37	227.42	NFW3	8.062	5.69	104.93
SFW3	20.521	14.51	266.84	NFW5	11.14	7.86	145.05
SFM	19.234	13.60	250.17	NFW7	9.476	6.69	123.61
SFS1	24.172	17.08	314.50	NFW9	8.768	6.19	114.06
SFS2	23.928	16.91	311.55	NFW11	8.386	5.93	109.14
SFT1	27.456	19.40	357.34	NFS1	11.30	7.98	147.02
SFT2	27.568	19.47	358.97	NFS2	13.47	9.51	175.40
SFP1	23.157	16.35	301.41	NFT1	14.99	10.59	195.24
SFP2	25.913	18.30	337.27	NFT2	14.94	10.55	194.65
Average	22.591	31.93	294.02	Average	10.81	7.63	140.75
Max	27.568	19.47	358.97	Max	14.99	10.59	195.24
Min	16.496	11.67	214.74	Min	7.567	5.35	98.37
STDEV	4.132	5.31	53.81	STDEV	2.887	2.04	37.61

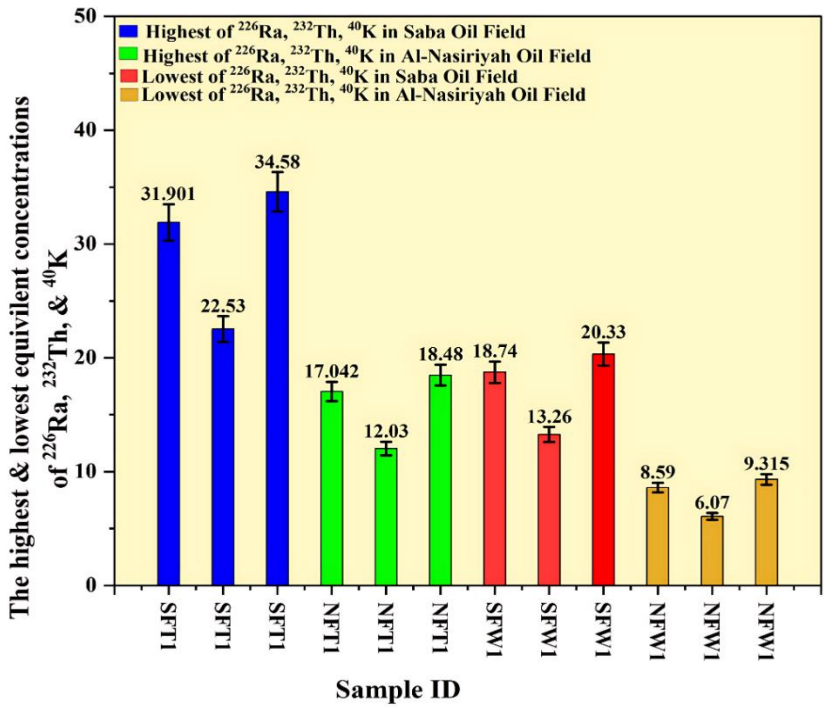

Figure 7.
The highest & lowest equivalent concentrations of ^{226}Ra, ^{232}Th, & ^{40}K in Saba & Al-Nasiriyah oil fields.

Table 4.
The gamma ray absorbed doses of natural radionuclides D_{ICRP}, $D_{UNSCEAR}$, and D_{BECK} in (nGy/h) in Saba & Al-Nasiriyah oil fields.

Saba oil field				**Nasiriyah oil field**			
Sample ID	**D_{ICRP}**	**D_{Beck}**	**D_{UNSCER}**	**Sample ID**	**D_{ICRP}**	**D_{Beck}**	**D_{UNSCER}**
SFW1	3.628	8.396	7.92	NFW1	2.001	3.909	3.14
SFW2	3.071	8.629	9.30	NFW3	2.277	4.157	3.55
SFW3	4.590	10.363	9.58	NFW5	2.948	5.412	4.84
SFM	4.429	9.778	9.01	NFW7	2.580	4.737	4.51
SFS1	5.800	12.165	11.11	NFW9	2.593	4.504	3.65
SFS2	5.951	12.083	11.14	NFW11	1.986	4.218	3.94
SFT1	6.824	13.885	12.61	NFS1	3.125	5.650	4.74
SFT2	6.905	13.775	12.71	NFS2	3.633	6.629	5.77
SFP1	5.998	11.588	10.31	NFT1	3.808	7.389	6.73
SFP2	6.588	12.918	11.59	NFT2	4.053	7.370	6.50
Average	5.378	11.358	10.53	Average	2.900	5.398	4.74
Max	6.905	13.885	12.71	Max	4.053	7.389	6.73
Min	3.071	8.396	7.92	Min	1.986	3.909	3.14
STDEV	1.423	2.056	1.68	STDEV	0.771	1.353	1.31

Table 5.
The annual effective dose (mSv/y.), Excess lifetime cancer risk, representative level index, and hazard indices (Bq/L) for all samples in Saba and al-Nasiriyah oil fields.

Saba oil field						**Nasiriyah oil field**					
Sample ID	**AED**	**ELCR $*10^{-4}$**	**Iγr**	**H_{ex}**	**H_{in}**	**Sample ID**	**AED**	**ELCR $*10^{-4}$**	**Iγr**	**H_{ex}**	**H_{in}**
SFW1	4.449	1.557	0.101	0.045	0.054	NFW1	2.454	0.859	0.041	0.020	0.024
SFW2	3.766	1.318	0.116	0.047	0.057	NFW3	2.793	0.978	0.044	0.022	0.027
SFW3	5.630	1.970	0.119	0.055	0.065	NFW5	3.615	1.265	0.060	0.030	0.037
SFM	5.431	1.901	0.112	0.052	0.061	NFW7	3.165	1.108	0.056	0.026	0.034
SFS1	7.114	2.490	0.138	0.065	0.078	NFW9	3.180	1.113	0.045	0.024	0.029
SFS2	7.298	2.554	0.138	0.065	0.079	NFW11	2.435	0.852	0.049	0.023	0.027
SFT1	8.369	2.929	0.157	0.074	0.090	NFS1	3.833	1.342	0.059	0.031	0.037
SFT2	8.469	2.964	0.158	0.074	0.092	NFS2	4.455	1.559	0.072	0.036	0.045
SFP1	7.356	2.575	0.128	0.063	0.076	NFT1	4.670	1.634	0.084	0.041	0.050
SFP2	8.080	2.828	0.144	0.070	0.085	NFT2	4.971	1.740	0.081	0.040	0.050
Average	6.596	2.309	0.131	0.061	0.074	Average	3.557	1.245	0.059	0.029	0.036
Max	8.469	2.964	0.158	0.074	0.092	Max.	4.971	1.740	0.084	0.041	0.050
Min	3.766	1.318	0.101	0.045	0.054	Min.	2.435	0.852	0.041	0.020	0.024
STDEV	1.745	0.611	0.020	0.011	0.014	STDEV	0.946	0.331	0.016	0.008	0.010

4. Conclusion

The current study was conducted in Saba and al-Nasiriyah oil fields in Thi Qar province, and showed that both oil fields indicated that the measured activity concentrations for ^{238}U, ^{232}Th, ^{226}Ra, ^{228}Ra, and ^{40}K respectively, in the crude oil samples were computed and found to be less than the permitted globally limits. The study introduces an essential data base on the concentration levels and the corresponding radiation doses for naturally occurring radionuclides. The hazard indices concentration levels were found to be less than the permissible worldwide values. Furthermore, the results indicated there is a relationship between the level of salinity and the Natural radionuclides concentrations which means the high level of salinity leads to the high concentrations of radionuclides in crude oil. The recommended safety limits for all other estimated parameters were concluded that there is no potential risk to radiological health associated with the crude oil produced from these oil fields. The results of the present study might be valuable when oil companies establish radiation safety regulations meant to protect the humans and the environment from radiation hazards that may arise from sources of crude oil.

Transparency:

The authors confirm that the manuscript is an honest, accurate, and transparent account of the study; that no vital features of the study have been omitted; and that any discrepancies from the study as planned have been explained. This study followed all ethical practices during writing.

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