

Studying the Radioactivity of Local and Imported cars oil in Baghdad using (HPGe) detector

دراسة النشاط الإشعاعي لزيوت السيارات المحلية والمستوردة في مدينة بغداد باستخدام كاشف الجرمانيوم عالي النقاوة

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الخلاصة

تم في هذا البحث دراسة النشاط الإشعاعي لخمسة عشر عينة من زيوت المحركات، ثمان منها محلية جمعت من محطات الوقود الموجودة في مدينة بغداد، وسبعة منها مستوردة من مناشيء مختلفة جمعت من الاسواق المحلية. تم التحري عن النويدات المشعة الموجودة في العينات وحساب فعاليتها النوعية باستخدام تقنية التحليل الطيفي لاشعة كما بواسطة منظومة كاشف الجرمانيوم عالي النقاوة (HPGe)، كما تم ايضا حساب معاملات الخطورة وعامل النوعية النسبية الذي تم حسابه لأول مرة في هذا البحث.

كشفت الدراسة عن وجود ثمان نويدات مشعة في العينات المدروسة هي (الرادون-222، الرصاص-214 والبزموت-214) التابعة لسلسلة اليورانيوم-238 و(الرصاص-212، الثاليوم-208 والاكينيوم-228) التابعة لسلسلة الثوريوم-232 بالاضافة الى البوتاسيوم-40 والسيزيوم-137.

بينت النتائج المستحصلة ان الفعالية المكافئة للراديوم تراوحت بين (1.73-13.72) Bq/l، ومعدل جرعة كما (0.82-6.24) nG/h ومعامل تمثيل كما (-0.0120-0.096) وقيم الجرعة الفعالة السنوية المكافئة الداخلية (-0.004-0.031 mSv/y) والخارجية (0.002-0.037 mSv/y) ومعامل الخطورة الداخلي (0.009-0.052) و الخارجي (-0.009-0.052) اما معامل النوعية النسبية فقد تراوح بين (1.76-13.26) Bq/l.

كما ان هناك معامل جديد قد تم حسابه لأول مرة في هذا البحث وهو الفعالية النوعية المؤثرة حيث تراوحت قيمته بين (1.76-13.26) Bq/l.

تم مقارنة معدل نتائج البحث الحالي مع معدل الحد العالمي فوجد انها ضمن الحدود المسموحه اي ان الزيوت المستخدمة في السيارات والمحركات امنة من الناحية الاشعاعية وصالحة للاستهلاك ولا تشكل خطر على عموم المجتمع و البيئة.

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Abstract

The radioactivity of fifteen cars oil samples were be studied in this research, eight of them are local collected from the fuel station in Baghdad, the remaining seven are imported from different origins collected from local markets of Baghdad.

The specific activity of the detected radionuclides were be measured by the spectral analysis technique of gamma-ray using high purity germanium (HPGe) detector. It was also a calculation of radiation risk factors; and the specific effective activity (A_{eff}), which was calculated for the first time in this research.

Eight radionuclides were be detected in this research ; (^{226}Ra , ^{214}Pb , ^{214}Bi) which belong to the uranium-238 series and (^{212}Pb , ^{208}Tl , ^{228}Ac) which belong to Th-232 series in addition to ^{40}K and ^{137}Cs .

The results shown that the radium equivalent activity (Ra_{eq}) were (1.73-13.72) Bq/l, the absorbed gamma doserate (D_v) were (0.82- 6.24) nG/h, the representative gamma index ($I_{\square r}$)

were(0.012-0.096), the indoor annual effective dose equivalent (AEDE)_{in} were(0.004-0.031) mSv/y,the outdoor annual effective dose equivalent (AEDE)_{out} were(0.002-0.037)mSv/y, and the internal hazard index(H_{int}) (0.009-0.052), the external hazard index (H_{ext}) (0.005-0.037).

Another factor has been calculated for the first time in this research it's the specific effective activity (A_{eff}) which is ranged between 1.76 and 13.26Bq/l.

The obtained results were compared with the worldwide average;it was within the recommended values. That mean, oils used in automobile engines are safe, valid for consumption and does not pose a danger to society and the environment.

Key words

Radionuclides, high purity germanium detector, car and engine oil,radiation risk factors,specific effective activity.

1. Introduction

The presence of naturally occurring radioactive material (NORM) has been known since early 1930s in petroleum tanks, in oil and gas production, and in their processing facilities [1].Petroleum is not a single chemical compound, Liquid petroleum or oil comprises a variety of liquid hydrocarbon compounds (carbon and hydrogen). There are also gaseous hydrocarbons (natural gas) [2].

Trace quantities of the radionuclides ²³⁸U, ²³²Th and ⁴⁰K have been present in the earth's crust since its formation [1], Radioactive decay of ²³⁸U and ²³²Th produces several radioisotopes of different elements and of different physical characteristics with respect to their half-lives, modes of decay, and energies of emitted radiation [3].

NOR activity concentrations can vary from one facility to another depending on geological formation and operational conditions and may also change over the lifetime of a single well[4].

In the exploration and extraction processes of oil and gas, the natural radionuclides ²³⁸U, ²³²Th, ²¹⁰Pb, ⁴⁰K, as well as the radium isotopes (²²³Ra, ²²⁴Ra, ²²⁶Ra -the only important isotope of radium because its average concentration in the earth's crust "40 Bq/kg"-and ²²⁸Ra), etc., are brought to the slurry surfaces and may contain levels of radioactivity above the surface background [1,5]. There are several exposure pathways to humans from oilfield NORM, such as inhalation of radon gas.

The major radionuclides of concern in oil and gas NORM are ²²⁶Ra and ²²⁸Ra; these isotopes are the decay products of uranium and thorium exist in subsurface formations from which hydrocarbons are industrialized. Although Ra is mobilized in the fluids, uranium and thorium are immobile to a large extent. In addition, ²¹⁰Pb and ²²²Rn particularly exist in gas processing equipment [6].

Gamma radiation emitted from (NOR), such as ⁴⁰K, and radionuclides from the ²³²Tl and ²³⁸U series and their decay, represent the main external source of irradiation to the human body. Sources of further exposure to radiation in the fields are radon inhalation, dust inhalation, ground-water ingestion, and skin beta exposure[7].

The objectives of this study, is to investigate the radionuclides and measure their specific activities and to estimate the hazard indices; radium equivalent activities, absorbed gamma dose rate, representative level index, external and internal hazard index. The results obtained will be compared with reported values in literatures in order to confirm the radiological hazards concerning with the automobiles oil consumed in Baghdad city involving natural radionuclides present in them are safe and within the world wide range.

2. Experimental details

2.1 Collection and preparation of the Samples

Fifteen samples of several types of the oils used in the engine cars in Baghdad were collected in this study, eight of them are local collected from the fuel stations, the remaining are imported from different origins collected from local markets.

The samples were weighed, stored and sealed in Marinelli beakers of one liter capacity, and kept for thirteen days in order to reach the radioactive secular equilibrium between ^{222}Rn , ^{220}Rn and their respective parent nuclei, ^{226}Ra , ^{224}Ra . before starting the investigations.

2.2 Coding the samples

The coding of the samples and the required information; date of collection, mass, types and country of origin, are listed in table (1).

Table 1. Sample code, date of collection, mass, state, type and the country of origin.

Sample code	Date	Mass(gm)	Type	Country of Origin
B1	9/9/2015	680	Kerosene	Iraq
B2	4/10/2015	746	Motor oil (Jumbo Dubai)	U.A.E
B3	4/10/2015	739	Motor oil (Furtcs)	U.A.E
B4	4/10/2015	743	Motor oil (Vulcan 330)	Iran
B5	4/10/2015	769	Motor oil (Super GT)	U.A.E
B6	8/10/2015	735	Motor oil (Fuchs)	Germany
B7	8/10/2015	782	Motor oil (Crafft)	K.S.A
B8	8/10/2015	693	Motor oil (Al Khaleej)	Kuwait
B9	8/10/2015	691	Gasoline	Iraq
B10	8/10/2015	541	Gasoline	Iraq
B11	8/10/2015	506	Gasoline	Iraq
B12	8/10/2015	715	Kerosene	Iraq
B13	8/10/2015	646	Gasoline	Iraq
B14	8/10/2015	687	Gasoline	Iraq
B15	8/10/2015	625	Gasoline	Iraq

2.3 High purity germanium (HPGe) detector

The oil samples were analyzed by gamma spectrometry of high purity germanium (HPGe) detector, of semiconductor type (p-type), Ge, 3"x3" crystal diameter, operation voltage 3600 volt D.C, relative efficiency 20% and peak/Compton ratio of 52:1 at 1.33 MeV (Co-60), No. of channels was 16.400 ch. A software program called MAESTRO- version (7.1) was used to accumulate and measure the data. The detector was shielded with lead blocks of 10 cm thick to reduce the background radiation effect.

The efficiency calibration achieved by placed a Marinelli beaker(1000cm³) of a standard mixed element of ten radionuclides on the crystal detector for (86.400s =2hr), the radionuclides in the beaker are (²⁴¹Am, ¹⁰⁹Cd, ¹³⁹Ce, ⁵⁷Co, ⁶⁰Co, ¹³⁷Cs, ¹¹³Sn, ⁸⁵Sr, ⁸⁸Y and ²⁰³Hg). The efficiency calibration curve drawn between energy and efficiency is shown in fig.1.

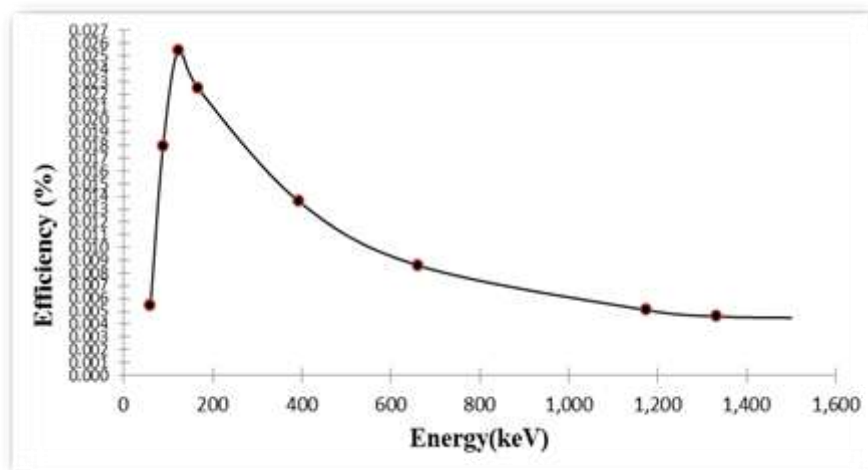


Figure 1. Efficiency calibration curve of (HPGe) detector using standard mixed source.

The efficiency of each radioisotopes detected in this research were identify from the efficiency calibration curve of the detector and listed in addition to their half-lives, energy, and abunds in table (2).

Table 2. radionuclides, gamma energies used in the research, abunds and efficiency.

Series	Isotope	Half-life	E _γ (keV)	I _γ (E _γ) %	Efficiency (%)
²³⁸ U	²²⁶ Ra	1600 y	186.21	3.64	0.0219
²³⁸ U	²¹⁴ Pb	26.80 m	295.21	19.20	0.0196
²³² Th	²¹² Pb	10.64 h	238.63	43.50	0.0173
²³⁸ U	²¹⁴ Pb	26.80 m	351.92	35.10	0.0150
²³² Th	²⁰⁸ Tl	3.07 m	583.19	30.58	0.0096
²³⁸ U	²¹⁴ Bi	19.90 m	609.32	44.60	0.0093
-----	¹³⁷ Cs	30 y	661.61	87.50	0.0086
²³² Th	²²⁸ Ac	6.13 h	911.16	26.60	0.0069
-----	⁴⁰ K	1.28×10 ⁹ y	1460.80	10.67	0.0049

2.4 Specific activity calculations:

The specific activities of the detected radionuclides were calculated by putting the Marinelli beaker of the samples on the crystal detector for (7200s=2hr) from the following equation [8]:

$$A_i(E_\gamma) = \frac{N}{\epsilon \cdot I_\gamma \cdot M \cdot t_c} \quad \dots (1)$$

Where $A_i(E_\gamma)$ is the specific activity of the radionuclides, N is the net area under the peak, t_c is the counting life time, I_γ is the abundance at energy E_γ , ϵ is the detection efficiency at energy E_γ , (M) is the mass of the oil sample in (liter).

The background correction was accomplished by measure the counts when placed an empty Marinelli beaker on the detector crystal for the same period of time of the samples.

2.4.1 The specific effective activity (A_{eff})

The specific effective activity (A_{eff}) was calculated by equation (2), where A_{Ra} , A_{Th} and A_K are specific activities of ^{226}Ra , ^{232}Th and ^{40}K in (Bq/kg) or (Bq/l). (1.31) and (0.085) are weighted coefficients for ^{232}Th and ^{40}K , respectively [9].

$$A_{eff} = A_{Ra} + 1.31 A_{Th} + 0.085 A_K \quad \dots\dots(2)$$

2.5 Calculation of the Radiation Hazard Indices

The radiological hazard indices were be calculated for each samples as follows:

2.5.1 Radium equivalent activity index (Ra_{eq})

It estimate that (1Bq/kg of ^{226}Ra), (1.43Bq/kg of ^{232}Th) and (0.077Bq/kg of ^{40}K) produce the same γ -ray dose [10]. It is calculated by equation (3),

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \dots\dots (3)$$

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

2.5.2 Gamma absorbed dose rate (D_γ)

It is the absorbed dose rate at a height of 1 m above the ground surface due to natural radioactivity. It refers to the radiation hazard to people from the steel slug in building materials, and calculated with equation (4) [11],

$$D_\gamma = 0.462C_{Ra} + 0.621C_{Th} + 0.0417 C_K \text{ (nGy/h)} \dots\dots (4)$$

Where C_{Ra} , C_{Th} and C_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K respectively [8].

2.5.3 Representative gamma level index ($I_{\gamma r}$)

Another radiation hazard index defined by equation (5) where, C_{Ra} , C_{Th} and C_K are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in Bq/kg, respectively [8,12].

$$I_{\gamma r} = (1/150)C_{Ra} + (1/100)C_{Th} + (1/1500)C_K \quad \dots\dots(5)$$

The safety value for this index is ≤ 1

2.5.4 Annual effective dose equivalent (AEDE)

The annual effective dose equivalent received by a member is calculated from gamma absorbed dose rate by applying dose conversion factor of (0.7Sv/Gy) and the occupancy factor for indoor and outdoor which is 0.8(19/24), and 0.2(5/24) respectively. It is determined with equations (6) and (7) respectively [8].

$$(AEDE)_{in} = D_\gamma \text{ (nGy/h)} \times 8760 \text{ h/y} \times 0.7 \text{ Sv/Gy} \times 0.8 \times 10^{-6} \quad \dots\dots(6)$$

$$(AEDE)_{out} = D_\gamma \text{ (nGy/h)} \times 8760 \text{ h/y} \times 0.7 \text{ Sv/Gy} \times 0.2 \times 10^{-6} \quad \dots\dots(7)$$

2.5.5 External (H_{ext}) and internal hazard indices (H_{int})

External and internal radiation hazards due to ^{40}K , ^{232}Th , and ^{226}Ra are denoted by H_{ext} is defined by the following expressions in equations (8) and (10)[12]:

$$H_{(ext)} = A_{Ra}/370 + A_{Th}/259 + A_k/4810 \leq 1 \quad \dots (8)$$

$$H_{(int)} = A_u/185 + A_{Th}/259 + A_k/4810 \leq 1 \quad \dots (9)$$

3. Results and discussion

Eight radionuclides were detected in the examined oil samples they are ;(^{226}Ra , ^{214}Bi , ^{214}Pb) which are the decay products of the ^{238}U Series, (^{212}Pb , ^{208}Tl , ^{228}Ac) which are the decay products of ^{232}Th series, the single radionuclides ^{40}K and the artificial ^{137}Cs .

The numbers of radionuclides in the samples are varying from 6 radionuclides in samples B5 and B8 to 2 radionuclides in sample B7. Some radionuclides can have highest specific activity in some samples and below the detection limits (B.L.D) in other samples. The radionuclide (K-40) appeared in all samples with average specific activity (6.10 ± 2.47) Bq/l, the artificial radionuclide ^{137}Cs was observed in 7 samples.

The comparison between the calculated specific activity values and the world wide average values recommended by (UNSCEAR) mention that our results were below these levels [14,15]. Three chosen spectrums of samples B1, B7 and B8 were shown below in figures (2),(3) and (4), B1 have the highest specific activity, B7 have the lowest number of nuclides, and B8 have the highest number of nuclides.

The detected radionuclides, their specific activity, the chains that decayed them, the minimum, maximum and the average values of the oil samples were listed in table (3).

The specific effective activity (A_{eff}) of ^{238}U , ^{232}Th and ^{40}K radionuclides for all the samples were calculated from equation (2) and listed in table (4).

The diagram of the specific effective activities (A_{eff}) of the oil samples shown in figure (5) clear the variation of the specific activity in these sample.

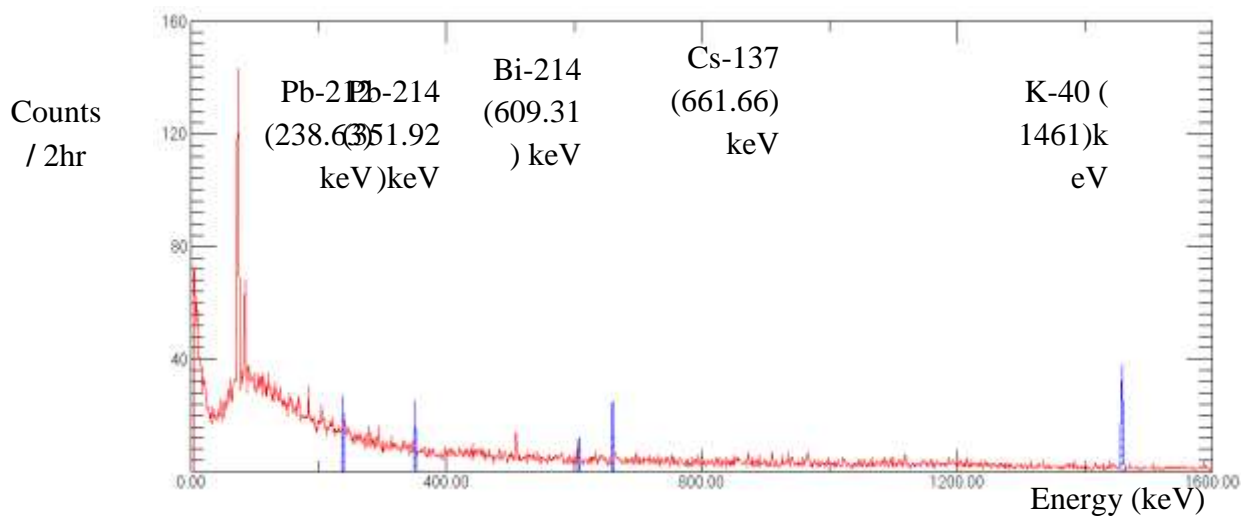


Figure 2. Gamma-rays spectrum for sample (B1).

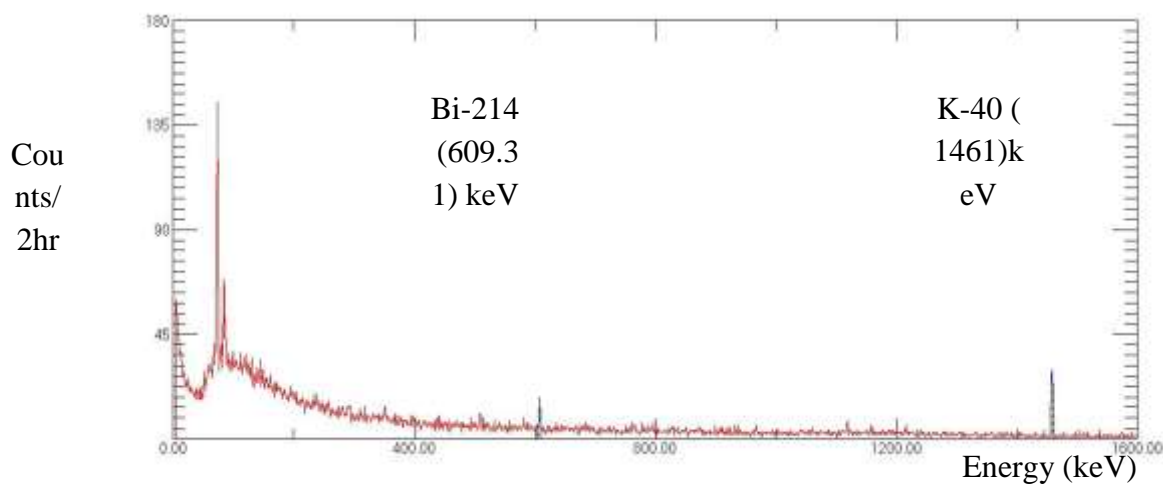


Figure 3. Gamma-rays spectrum for sample (B7).

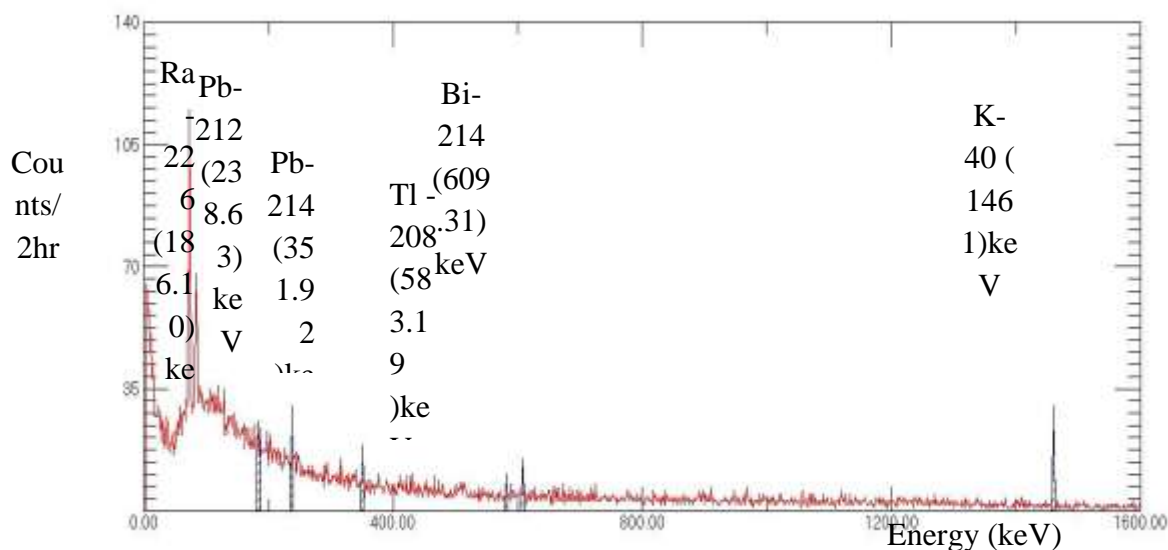


Figure 4. Gamma-rays spectrum for sample (B8).

Table 3. The specific activities of the detected radionuclides.

Sample code	Specific activity of the detected radionuclides (Bq/l)								
	238U- Series			232Th-Series			K-40	Cs-137	Nuclides No.
	Ra-226	Bi-214	Pb-214	Pb-212	Tl-208	Ac-228			
B1	B.D.L	3.22	1.95	2.80	B.D.L	B.D.L	8.14	1.27	5
B2	B.D.L	B.D.L	2.30	B.D.L	B.D.L	B.D.L	9.96	0.99	3
B3	0.14	4.87	2.35	B.D.L	B.D.L	B.D.L	1.50	0.37	5
B4	0.54	4.45	1.05	B.D.L	B.D.L	1.23	12.07	B.D.L	5
B5	1.32	B.D.L	5.62	4.84	B.D.L	0.36	15.32	0.36	6
B6	0.31	B.D.L	1.66	B.D.L	B.D.L	B.D.L	14.32	0.63	4
B7	B.D.L	4.60	B.D.L	B.D.L	B.D.L	B.D.L	4.61	B.D.L	2
B8	0.19	2.88	0.60	2.30	2.04	B.D.L	6.40	B.D.L	6
B9	2.50	2.13	2.63	B.D.L	B.D.L	B.D.L	5.77	B.D.L	4
B10	B.D.L	B.D.L	1.44	B.D.L	B.D.L	B.D.L	3.72	0.68	3
B11	2.31	4.23	0.77	5.34	B.D.L	B.D.L	1.87	B.D.L	5
B12	B.D.L	B.D.L	0.73	2.91	B.D.L	2.40	2.56	B.D.L	4

B13	0.47	B.D.L	B.D.L	2.47	B.D.L	B.D.L	1.93	0.77	4
B14	B.D.L	4.62	3.02	B.D.L	B.D.L	B.D.L	1.52	B.D.L	3
B15	B.D.L	3.13	1.16	2.16	B.D.L	B.D.L	1.79	B.D.L	4
Min.	B.D.L	B.D.L	B.D.L	B.D.L	B.D.L	BDL	1.50	B.D.L	2
Max.	2.50	4.87	5.62	5.34	2.04	2.40	15.32	1.27	6
Ave.	0.52±0.47	2.28±1.16	1.69±1.30	1.52±1.23	0.14±0.10	0.27±0.22	6.10±2.47	0.34±0.23	
worldwide Ave.	50 [14]			50 [14]			500[14]	14.8[15]	

Table 4. The specific effective activities of ^{238}U , ^{232}Th , ^{40}K radionuclides.

Sample code	^{238}U (Bq/l)	^{232}Th (Bq/l)	^{40}K (Bq/l)	A_{eff} (Bq/l)
B1	3.22	2.80	8.14	7.58
B2	2.30	B.D.L	9.96	3.15
B3	4.87	B.D.L	1.50	5.00
B4	4.45	1.23	12.07	7.09
B5	5.62	4.84	15.32	13.26
B6	1.66	B.D.L	14.32	2.88
B7	4.60	B.D.L	4.61	4.99
B8	2.88	2.300	6.40	6.44
B9	2.63	B.D.L	5.77	3.12
B10	1.44	B.D.L	3.72	1.76
B11	4.23	5.34	1.87	11.38
B12	0.73	2.91	1.87	4.70
B13	0.47	2.47	1.93	3.87
B14	4.62	B.D.L	1.52	4.75
B15	3.13	2.16	1.79	6.11
Min.	0.47	B.D.L	1.50	1.76
Max.	5.62	5.34	15.32	13.26
Ave.	3.12±1.70	1.60±0.90	6.05±2.27	5.74 ±2.32

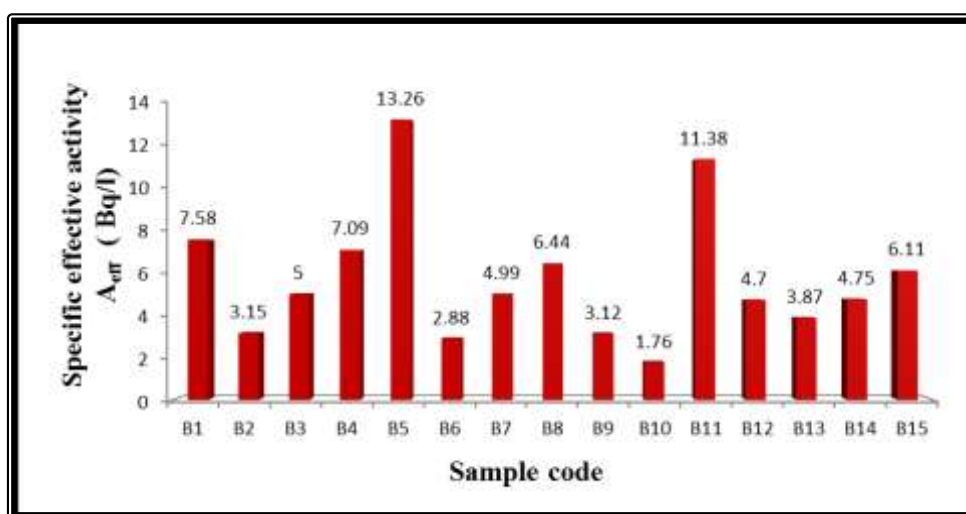


Figure 5. Specific effective activity (A_{eff}).

The hazard indices; Radium Equivalent Activity(R_{eq}), absorbed gamma dose rate (D_γ), representative gamma index (I_γ), annual effective dose equivalent(AEDE), external hazard index (H_{ext}), internal hazard index (H_{int}) were be calculated and listed in table (5).The average and the highest values of the radiation hazard indices are under the world wide average values [14, 16]as clear in table (5).

Table 5.Radiation hazard indices of samples.

Samples code	R_{eq} (Bq/l)	D_γ (nGy/h)	Annual effective dose Equivalent (mSv/y)		I_γ	Hazard index	
			(AEDE) _{in}	(AEDE) _{out}		H_{int}	H_{ext}
B1	7.85	3.57	0.017	0.009	0.055	0.030	0.021
B2	3.07	1.48	0.007	0.002	0.022	0.015	0.008
B3	4.99	2.31	0.011	0.003	0.033	0.027	0.013
B4	7.14	3.32	0.016	0.017	0.050	0.031	0.019
B5	13.72	6.24	0.031	0.008	0.096	0.052	0.037
B6	2.76	1.36	0.007	0.010	0.021	0.012	0.007
B7	4.95	2.32	0.019	0.020	0.034	0.026	0.013
B8	6.66	3.03	0.015	0.026	0.046	0.026	0.018
B9	3.07	1.46	0.007	0.012	0.021	0.015	0.008
B10	1.73	0.82	0.004	0.023	0.012	0.009	0.005
B11	12.01	5.35	0.026	0.007	0.083	0.044	0.032
B12	5.04	2.22	0.012	0.014	0.035	0.016	0.014
B13	4.15	1.83	0.009	0.032	0.029	0.012	0.011
B14	4.74	2.20	0.013	0.021	0.032	0.025	0.013
B15	6.36	2.86	0.019	0.037	0.044	0.026	0.017
Min.	1.73	0.82	0.004	0.002	0.012	0.009	0.005
Max.	13.72	6.24	0.031	0.037	0.096	0.0522	0.037
Ave.	5.88	2.69	0.014	0.016	0.041	0.024	0.017
Worldwide Ave.	370 [14]	55[14]	20[14]	1[14]	1 [15]	<1[16]	1[16]

4. Conclusions

1. Eight radionuclides were detected in the selected examined samples includes; three radionuclides (Ra-226, Bi-214, Pb-214) belong to uranium-238 series , three radionuclides (Pb-212, Tl-208, Ac-228) belong to thorium-232series,the single radionuclide K-40, and the artificial radionuclide Cs-137 which appeared in seven samples (B1,B2,B3,B5,B6 ,B10 ,B13).
2. The average values of the specific activities of the radionuclides in addition to the highest values were under the worldwide average recommended by (UNSCEAR).
3. The values of the six radiation hazard indices were be less than the recommended values given by worldwide average.
4. The addition values calculated in this research considered that radiation dose of the oil is safe for consumers.

5. References

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