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Assessment of natural radioactivity and associated radiation hazards of some used engine oil samples

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Abstract

To assess the natural radioactivity and associated radiation hazards of used and unused engine oil in the environmental, some samples have been collected from fuel station and local markets in Baghdad city of Iraq. The specific activities of ^{238}U, ^{232}Th, ^{40}K and ^{235}U were measured by using a gamma ray spectrometer based on a high purity germanium (HPGE) detector. The results showed that the activities concentration of Ra-226 (^{238}U series), ^{232}Th, ^{40}K and ^{235}U were (0.59-18.41) Bq/L and average (10.15) Bq/L, (0.00-35.47) Bq/L and average (14.87) Bq/L , (14.16-245.27) Bq/L and average (88.97) Bq/L,(0.03-0.85) Bq/L and average (0.47) Bq/L, respectively. The hazard indices have been calculated in the present work which include (radium equivalent activity (Ra_{eq}), absorbed dose rate in Air (AD),annual effective dose equivalent (A.E.D) which include indoor and outdoor effective dose rate, activity concentration Index (Iγ), external hazard index (H_{ext}) and internal hazard index (H_{int})), respectively. For the used engine oil samples, the highest values of (Ra_{eq} (Bq/l), AD (nGy/h) , $A.E.D_{in}$(mSv/y) , $A.E.D_{out}$ (mSv/y) , Iγ, H_{ext}, H_{int}) which were equal to (52.57, 24.18, 0.12, 0.03, 0.38, 0.19, 0.14), respectively , for Iraqi used engine oil sample, while the lowest values of (Ra_{eq} (Bq/l), AD (nGy/h) , $A.E.D_{in}$ (mSv/y) , $A.E.D_{out}$ (mSv/y) , Iγ,H_{ext}, H_{int}) was found in (U.A.E) sample which was equal to (5.21, 2.49, 0.01, 0.003, 0.04, 0.03, 0.01), respectively. While the unused engine oil samples, the highest values of (Ra_{eq} (Bq/l), AD (nGy/h) , $A.E.D_{in}$ (mSv/y) , $A.E.D_{out}$ (mSv/y) , Iγ, H_{ext}, H_{int}) which were equal to (70.21, 33.03, 0.16, 0.04, 0.52, 0.24, 0.19), respectively ,for Iraqi unused engine oil sample, while

the lowest values of (Ra_{eq} (Bq/l), AD (nGy/h) , $A.E.D_{in}$ (mSv/y) , $A.E.D_{out}$ (mSv/y) , Iγ, H_{ext}, H_{int}) were found in (U.A.E) sample which were equal to (27.73, 13.12, 0.06, 0.02, 0.20, 0.11, 0.08), respectively. The results comparison with worldwide average and it is found that the levels of radiation and radiation hazards indices values were within the permissible limits globally , the study showed that the used and unused engine oil are safe radiological and does not constitute a danger to the environmental.

Keywords: Used engine oils, NORM, (HPGe) Detector, Radiological hazards, Natural radioactivity

Introduction

In recent years the problem of the contamination of soil, water and air with petroleum products has become ever more increasing [1, 2]. Used engine oils are particularly dangerous for the environment due to their contamination with products of thermal decomposition and mechanical impurities. The above products become severely biodegradable and have carcinogenic properties, therefore, information on the content of radioactive elements (radionuclides) in these products are critical, taking into account the environmental aspect. petroleum products mainly contain liquid hydrocarbon compounds (carbon and hydrogen) and also gaseous hydrocarbons (natural gas) [3], but also can be present in small quantities the naturally occurring radioactive materials (NORM) .The primary radionuclide of concern in oil and gas stream are Ra-226 (U-238 series), Th-232, and K-40 which are responsible for most of the external exposure in such facilities [4]. Radioactive decay of U-238 and Th-232 produces several series of daughter radioisotopes of different physical characteristics and of different elements with respect to their half-lives, modes of decay, and types and energies of emitted radiation [5]. The radium isotopes are the decay products of uranium and thorium exist in subsurface formations from which hydrocarbons are industrialized. Since Radium-226 is slightly soluble, it is mobilized in the fluids, these properties for radium depend on the salinity of the formation water; higher salinity is aligned with a greater solubility [6].

The gamma radiation emitted from naturally occurring radionuclides materials NORMs is called terrestrial background radiations, which exist as trace levels in all ground formations, represents the main external source of irradiation to the human body [7]. However, if there is any human interference which leads to increase the level of NORM by any means or processes (such as dumping, extracting or producing radioactive materials from uranium mines or in oil production etc.), then it is called technically enhanced radioactive materials [8,9].

The aim of the present study is to determine the activity concentrations and the associated radiation hazards such as radium equivalent activity, absorbed dose rate in air, indoor and outdoor annual effective dose equivalents, activity concentration Index, internal and external hazard indices in different types for used and unused engine oil samples from different origins by using high purity germanium detector.

2. Materials and methods

A. Collection and Preparation of the samples

Engine oil samples were collected from fuel stations and markets in Baghdad city of Iraq; all most of these were foreign origins and the others from Iraqi origins. The samples were weighed, stored and sealed in polyethylene Marinelli beakers of

half liter capacity, and kept for one month in order to reach the radioactive secular equilibrium for U-238 and Th-232 with their respective progenies before starting the measurements [10-13].

The instrumentation used to measure gamma-rays from engine oil samples consists of a HPGe semiconductor detector (p-type, Ge, 3"x3" crystal diameter, operation voltage 3600 volt D.C, resolution =1.77 keV at 1332 keV ^{60}Co), associated electronics, and a computer-based multichannel analyzer (MCA) as shown in Fig.1. The gamma ray lines of 186.21 keV from Ra-226, 295.21 and 351.95 keV gamma-rays from Pb-214, 609.32 from Bi-214 were used to determine the U-238. The gamma ray lines of 583.19 keV from Pb-212 and 911.16 keV gamma rays from Ac-228 were used to determine the

Th-232. The activity of K-40 was evaluated using its 1460.80 keV gamma ray line. The detector was shielded with lead blokes of 10 cm thick to reduce the background radiation effect.

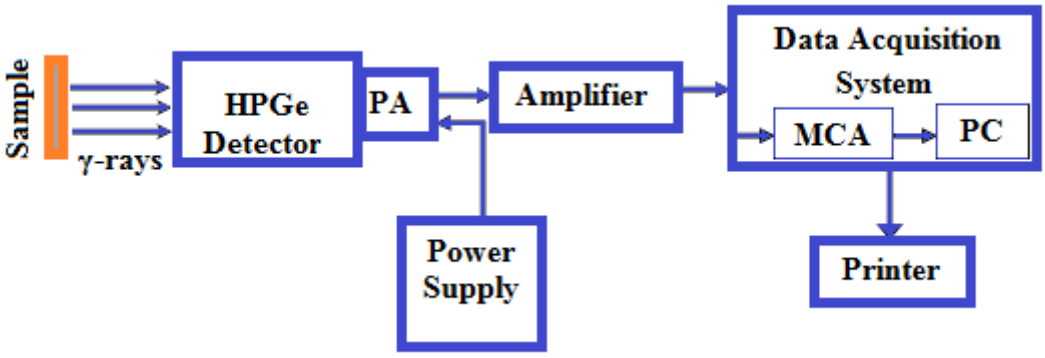

Figure .1: Blocked diagram of gamma-ray spectrometer system.

B. Activity concentrations Calculation

The activity concentrations of the radionuclides in the measured samples were computed using the following relations [14]:

$$A = N_{net} / \varepsilon . I\gamma . m . t \ (Bq/l) \quad \dots (1)$$

A: Activity concentrations of the radionuclides measured in (Bq/l) units.

N_{net}: Area under the photo-peak.

ε: efficiency of the detectors at energy Eγ.

M: mass engine oil samples (0.5 liter).

Iγ: is the abundance at energy Eγ.

t: time of measurement which was equal to (7200 s).

$$A_{u\text{-}235} = A_{u\text{-}238} / 21.7 \quad \dots (2)$$

C. Radiation Hazard indices Calculation

1. Radium Equivalent Activity (Ra_{eq})

The Radium equivalents (Ra_{eq}) in Bq/ l are used to assess the radiological hazards associated with materials that contain natural radioactivity in Bq/l using the following equation to calculate Ra_{eq} [15-16]:

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

Where, A_{Ra}, A_{Th} and A_K are the specific activity concentrations of Ra-226, Th-232 and K- 40 in (Bq/l) units respectively. Maximum value of Ra_{eq} must be less than 370 Bq/ l that is equivalent to the annual dose equivalent of 1.5 mSv/y.

2. Activity Concentration Index (Iγ)

The Activity Concentration Index (Iγ), is used to estimate the level of gamma radiation hazard associated with the natural radionuclides in specific construction materials. The gamma index (Iɣ) was calculated by using the following relation [17]:

$$I = A_{Ra} / 150 + A_{Th} / 100 + A_K / 1500 \ldots (4)$$

Where, A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/l, respectively. The safety value for this index is ≤ 1.

3. External Hazard Index (H_{ex})

The external hazard index H_{ex} can be calculated by the following equation [18]:

$$H_{(ext)} = A_{Ra} / 370 + A_{Th} / 259 + A_K / 4810 \leq 1 \ldots (5)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/l, respectively, the values of this index must be less than unity in order to keep the radiation hazard without posing any significant radiological threat to public.

4. Internal Hazard Index (H_{in})

The internal exposure to ^{222}Rn and its daughter products is represented by the internal radiation hazard index, H_{in} which is given as [18]:

$$H_{(int)} = A_{Ra} / 185 + A_{Th} / 259 + A_K / 4810 \leq 1 \ldots (6)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/l, respectively, this index value must be less than unity in order to keep the radiation hazard to be insignificant.

5. Absorbed Dose Rate in Air (AD)

In order to assess radiological risk, external exposure to radiation arising from naturally occurring radionuclides can be determined in terms of the absorbed dose rate in air at 1 m above the ground surface. The conversion factors used to compute absorbed dose rate in air per unit activity concentration in Bq/l corresponds to 0.462 nG/h for ^{226}Ra,0.621 nG/h for ^{232}Th and 0.0417 nG/h for ^{40}K [18].

$$D_\gamma (nGy/h) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \ldots (7)$$

Where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/l respectively.

6. The Annual Effective Dose (A.E.D)

Annual estimated average effective dose equivalent received by a member is calculated using number of hours in year (24 x 365 = 8760 h/year), a conversion factor of (0.7 Sv/Gy), which is used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20% and 80% for indoors [19].

The annual effective doses are determined as follows:

(AED) in (mSv/y) = Absorbed Dose (nGy/h) × 8760 h/y × 0.7 Sv/Gy × 0.8 × 10^{-6} … (8)

(AED) out (mSv/y) =Absorbed Dose (nGy/h) × 8760 h/y × 0.7 Sv/Gy × 0.2 × 10^{-6} … (9)

3. Results and Discussions

3.1 The activity concentration

The activity concentrations of natural radionuclide for all samples were calculated using equation (1, 2) and summarized in table (1, 2) and fig (2).The activity concentration of Ra-226, Th-232, and K-40, U-235 were 0.59-18.41 Bq/L, with the average value of 10.15Bq/L, 0.00-35.47Bq/L, with the average value of 14.87Bq/L, 14.16-245.2Bq/L, with the average value of 88.97 Bq/L, 0.03-0.85 Bq/L, with the average value of 0.47 Bq/L, respectively.

Table.1: Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{235}U in Bq/l in engine oil samples and other petroleum products.

Sample Code	Origins	Kinds	Ra-226 (Bq/l)	Th-232 (Bq/l)	K-40 (Bq/l)	U-235 (Bq/l)
Used engine oils						
UO1	Iraq	Al-Dora	17.24	19	105.97	0.79
UO2	Germany	Liqui Moly	5.15	13.45	50.1	0.24
UO3	U.A.E	Super Nevad	4.12	B.D.L	14.16	0.19
Unused engine oils						
NO4	Iraq	Al-Dora	18.41	23.02	245.27	0.85
NO5	Germany	Liqui Moly	13.04	18.2	230.41	0.60
NO6	U.A.E	Super Nevad	11.16	6.62	92.3	0.51
Other petroleum products						
HO7	U.A.E	Hydraulic oil (Super Nevad)	1.9	5.82	15.23	0.70
GO8	U.A.E	Gear oil (Super Nevad)	0.59	24.34	35.47	0.03
CO9	Iraq	Crude oil (wasit)	6.63	15.06	49.53	0.31
CO10	Iraq	Crude oil Salahuddin	11.22	6.28	51.22	0.52
Min			***0.59***	***0.00***	***14.16***	***0.03***
Max			***18.41***	***35.47***	***245.27***	***0.85***
Ave			***10.25***	***14.64***	***88.97***	***0.47***

From table (2) it can be noticed that, the average activity concentration of Ra-226, Th-232, K-40 and U-235 for used engine oil were 8.84 Bq/L , 16.23 Bq/L , 56.74 Bq/L , 0.41 Bq/L, respectively, while the average activity concentration of same

natural radionuclide for unused engine oil were14.20 Bq/L, 15.95 Bq/L , 189.33 Bq/L , 0.65 Bq/L , respectively, which is below the world wide averaged value of Ra-226, Th-232, and K-40 in normal oil samples which are 50, 50, and 500 Bq/kg or Bq/l , respectively cited by UNSCEAR [20].

Table .2: Minimum, maximum and mean activity concentration values for used and unused engine oil samples.

Radionuclide	Minimum (Bq / l)	Maximum (Bq / l)	Mean (Bq / l)
Used engine oils			
^{226}**Ra**	4.12	17.24	8.84
^{232}**Th**	0.00	19.00	16.23
^{40}**K**	14.16	105.97	56.74
^{235}**U**	0.19	0.79	0.41
Unused engine oils			
^{226}**Ra**	11.16	18.41	14.20
^{232}**Th**	6.62	23.02	15.95
^{40}**K**	92.30	245.27	189.33
^{235}**U**	0.51	0.85	0.65
worldwide Ave.	^{226}***Ra***	^{232}***Th***	^{40}***K***
	50*[20]*	***50****[20]*	***500****[20]*

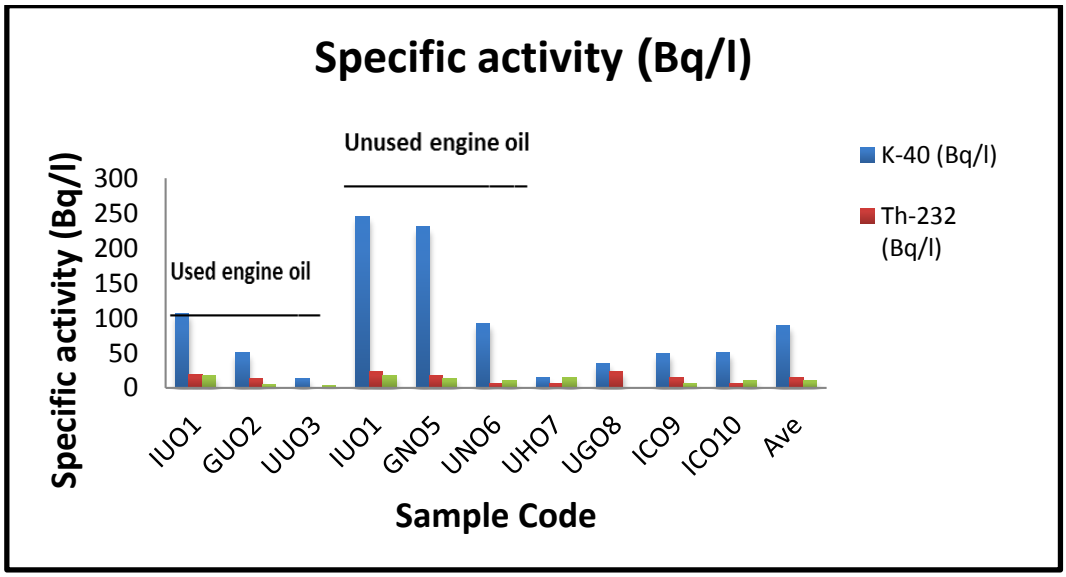

Figure .2: Specific activity of (^{226}Ra, ^{232}Th and^{40}K) for all samples.

3.2 Radiological risk

The Hazard indices for all samples in the present work were calculated using the equations (3-9) and listed in table (3, 4) and figs (3-7) which include (Radium equivalent activity (Ra_{eq}), Absorbed Dose Rate in Air (AD), Annual effective dose equivalent (A.E.D) which include indoor and outdoor effective dose rate, Activity concentration Index (Iγ), external hazard index (H_{ext}) and internal hazard index (H_{int})), respectively. The results shows that the average values of hazard indices ($Ra_{eq,}$ AD, $A.E.D_{in,}$ $A.E.D_{out,}$ Iγ, H_{ext}, H_{int}) were (37.52, 17.37, 0.085, 0.021, 0.27, 0.130, 0.101), respectively. The results have been

shown that the average values of hazard indicts for all samples of the present study were less than the worldwide average values for the hazard indicts given by [20, 21].

Table . 3: Radiation risk indices in used, unused engine oil and some other petroleum products samples

Sample Code	Ra_{eq} (Bq/l)	DɣR (nGy/h)	(A.E.D) (mSv/y)		Iγr	Hazard index	
			$(A.E.D)_{in}$	$(A.E.D)_{out}$		H_{in}	H_{out}
UO1	52.57	24.18	0.119	0.030	0.38	0.189	0.142
UO2	28.24	12.82	0.063	0.016	0.20	0.090	0.076
UO3	5.21	2.49	0.012	0.003	0.04	0.025	0.014
NO4	70.21	33.03	0.162	0.041	0.52	0.239	0.190
NO5	56.81	26.93	0.132	0.033	0.42	0.189	0.153
NO6	27.73	13.12	0.064	0.016	0.20	0.105	0.075
HO7	24.70	11.27	0.055	0.014	0.17	0.108	0.067
HO8	53.19	23.31	0.114	0.029	0.37	0.145	0.144
CO9	31.98	14.48	0.071	0.018	0.23	0.104	0.086
CO10	24.14	11.22	0.055	0.014	0.17	0.096	0.065
Min	***5.21***	***2.49***	***0.012***	***0.003***	***0.04***	***0.025***	***0.014***
Max	***70.21***	***33.03***	***0.162***	***0.041***	***0.52***	***0.239***	***0.190***
Ave	***37.52***	***17.37***	***0.085***	***0.021***	***0.27***	***0.130***	***0.101***

For the used engine oil samples, table (4) shows that the highest values of hazard indices (Ra_{eq} (Bq/l), AD (nGy/h), $A.E.D_{in}$(mSv/y), $A.E.D_{out}$ (mSv/y), Iγ, H_{ext}, H_{int}) were found in Iraqi sample (UO1) which were equal to (52.57 Bq/l, 24.18 nGy/h, 0.119 mSv/y, 0.030 mSv/y, 0.38, 0.189, 0.142), respectively , while the lowest values of the same hazard indices were found in (U.A.E) sample (UO3) which were equal to (5.21 Bq/l, 2.49 nGy/h, 0.012 mSv/y, 0.003 mSv/y, 0.04, 0.025, 0.014), respectively, with average value of (28.76 Bq/l, 13.23 nGy/h, 0.07 mSv/y, 0.02 mSv/y, 0.21, 0.10, 0.08), respectively.

Table . 4: Minimum, maximum and mean hazards indices values for used and unused engine oil samples.

Sample Type	Ra_{eq} (Bq/l)	DɣR (nGy/h)	(A.E.D) (mSv/y)		Iγr	Hazard index	
			$(A.E.D)_{in}$	$(A.E.D)_{out}$		H_{in}	H_{out}
Used engine oils							
Max	52.57	24.18	0.12	0.03	0.38	0.19	0.14
Min	5.21	2.49	0.01	0.00	0.04	0.03	0.01
Ave	28.76	13.23	0.07	0.02	0.21	0.10	0.08
Unused engine oils							
Max	70.21	33.03	0.16	0.04	0.52	0.24	0.19
Min	27.73	13.12	0.06	0.02	0.20	0.11	0.08
Ave	50.54	23.85	0.12	0.03	0.37	0.18	0.14
worldwide Ave.	***370 [20]***	***55 [20]***	***20 [20]***	***1 [20]***	***1[21]***	***1 [21]***	***1 [21]***

Also from the table (4) it can be noticed that, the highest values of hazard indices (Ra_{eq} (Bq/l), AD (nGy/h) , $A.E.D_{in}$(mSv/y) , $A.E.D_{out}$ (mSv/y) , Iγ, H_{ext}, H_{int}) of the unused engine oil samples were found in Iraqi sample (NO4) which were equal to (70.21Bq/l, 33.03 nGy/h, 0.16 mSv/y, 0.04 mSv/y, 0.52, 0.24, 0.19), respectively, while the lowest values of the same hazard indices were found in (U.A.E) sample (NO6) which were equal to (27.73 Bq/l , 13.12 nGy/h , 0.06 mSv/y , 0.02 mSv/y , 0.20

, 0.11, 0.08), respectively, with average value of (50.54 Bq/l , 23.85 nGy/h , 0.12 mSv/y , 0.03 mSv/y, 0.37 , 0.18 , 0.14) respectively.

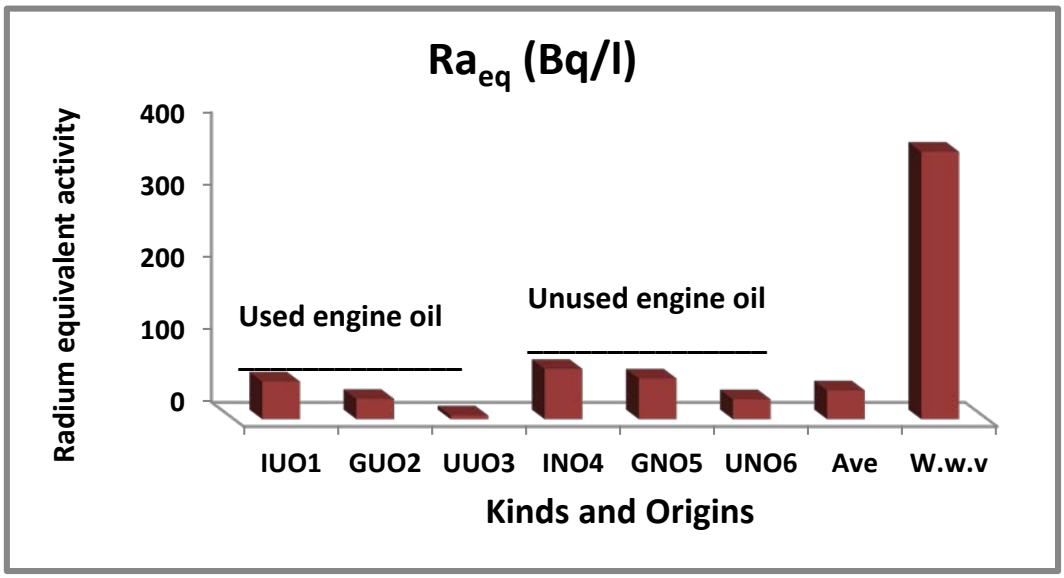

Figure .3: Radium equivalent activity (Ra_{eq}) for the engine oil samples

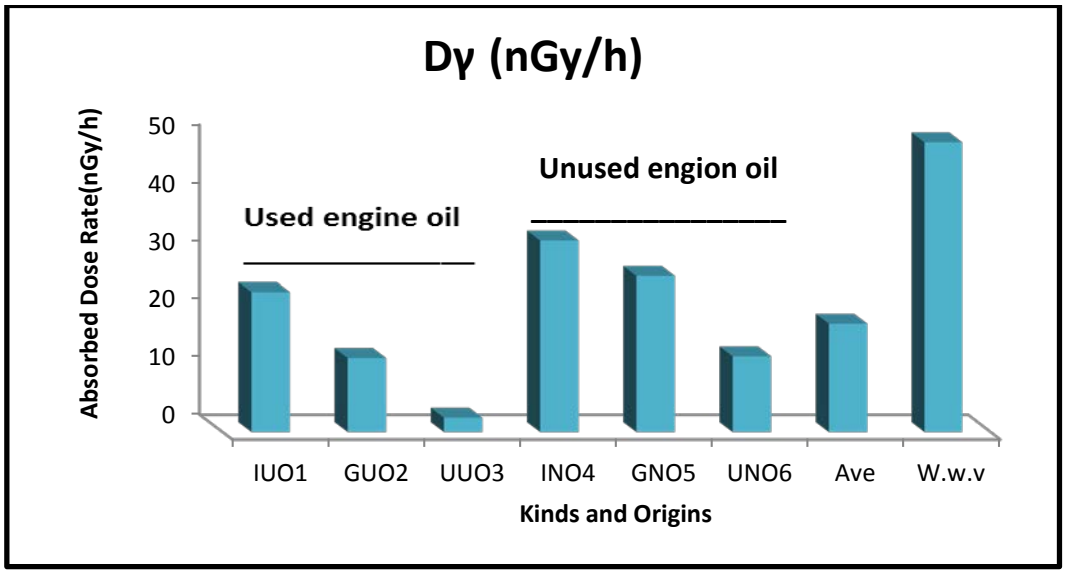

Figure .4: Absorbed dose rate in air (AD) for the engine oil samples

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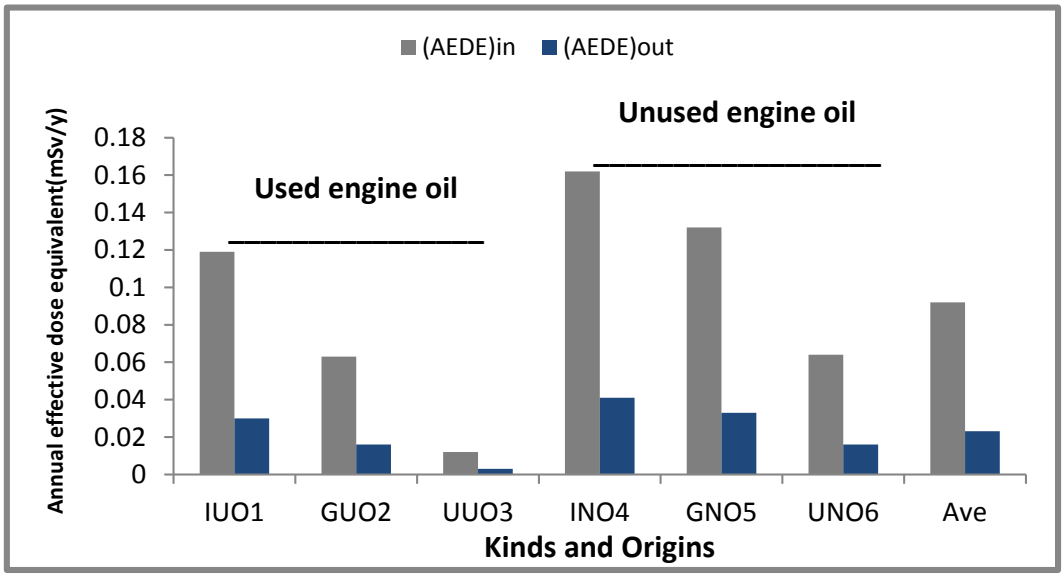

Figure .5: Annual effective dose equivalent (indoor and outdoor effective dose rate) for the engine oil samples

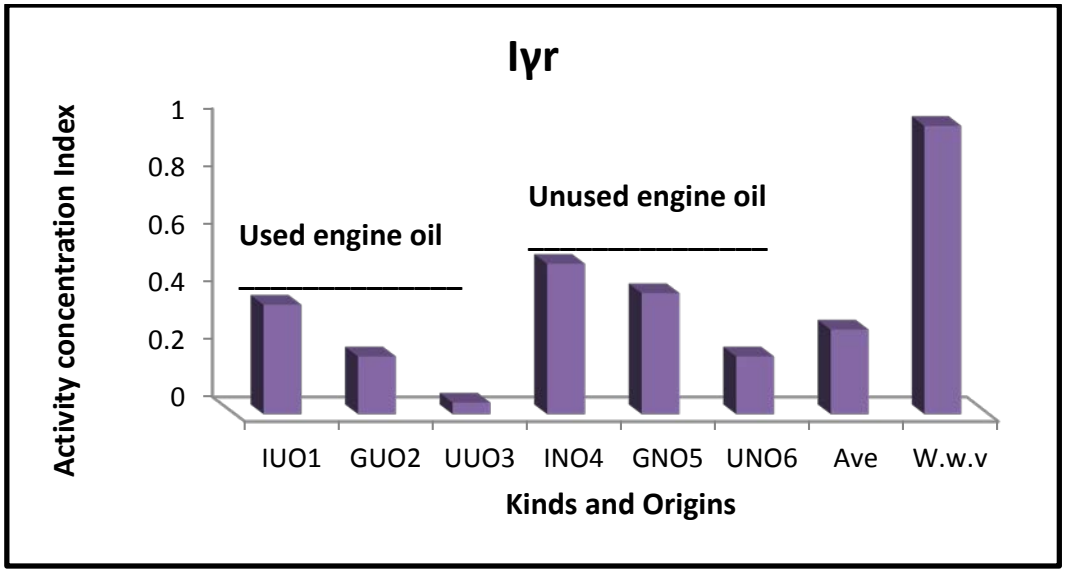

Figure. 6: Activity concentration Index (Iγ) for the engine oil samples

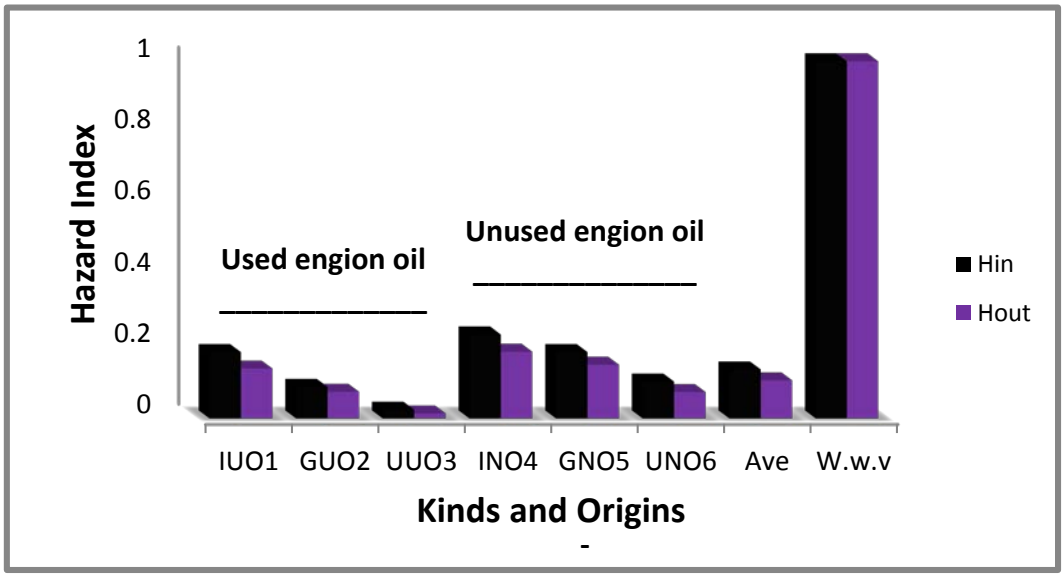

Figure.7: External hazard index (H_{ext}) and internal hazard index (H_{int}) for the engine oil samples

4. Conclusions

In the present study, gamma ray spectrometer based on a high purity germanium (HPGE) detector was exploited to determine activity concentration due to naturally occurring radionuclides materials NORMs ^{226}Ra (238U series), ^{232}Th and ^{40}K radioisotopes and the associated radiation hazard levels in some used and unused engine oil samples collected from fuel station and local markets in Baghdad city of Iraq. The average concentrations for ^{226}Ra, ^{232}Th, ^{40}K and ^{235}U were 8.84 Bq/L , 16.23 Bq/L , 56.74 Bq/L , 0.41 Bq/L, respectively, in used engine oil samples and in unused engine oil samples were 14.20 Bq/L, 15.95 Bq/L , 189.33 Bq/L , 0.65 Bq/L , respectively. These average activity concentrations of NORMs were lower than the world average values. The averages values of the obtained hazard indices such as: Radium equivalent activity (Ra_{eq}), Absorbed Dose Rate in Air (AD), Annual effective dose equivalent (A.E.D) which include indoor and outdoor effective dose rate, Activity concentration Index (Iγ), external hazard index (H_{ext}) and internal hazard index (H_{int}) were less than the world wide average values. This indicates that the used and unused engine oil samples are safe radiological and does not constitute a danger to the environmental.

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