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Analytical Study Of The Radioactivity Of Some Asphalt Springs In The Heet City, Anbar Governorate, Iraq

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Abstract
This research aims to estimate pollution levels of the concentrations of ^{238}U, ^{232}Th, and ^{40}K in the soil, water, and asphalt for some Heet City in the west of Anbar Governorate - Iraq to find out the natural and industrial radioactivity contaminations. In this study, the gamma-ray spectroscopy technique was used with a Germanium detector to calculate the levels of radioactivity of natural radionuclides (^{238}U, ^{232}Th, ^{40}K) in different samples of soil, water, and asphalt collected from (8) areas of Heet City and its nearby regions in Anbar Governorate - Iraq. Through the study results, it was shown that the range of concentration values for ^{238}U, ^{232}Th, and ^{40}k in soil samples ranged from (1.2, 2, and 26) Bq/kg to (810, 506.6, and 435.7) Bq/kg and at an average (147.2, 94,1, and 309.28), respectively. While in asphalt samples, the concentrations ranged between (17 and 48) Bq/kg for both ^{238}U and ^{232}Th, and for ^{40}k, they ranged between (220 and 326) Bq/kg. As for water samples, the study did not detect any concentrations of ^{238}U and ^{232}Th, and the detected concentrations of ^{40}k were very low. In general, from this study, we conclude that the Abu Al-Jeer region contains relatively high radioactivity levels, higher than the permissible global limit in some of its areas. Therefore, periodic and continuous monitoring must be conducted to verify radioactivity in these areas to avoid any potential danger to the population.

Keywords: *radioactivity, asphalt, soil, water, gamma-ray spectroscopy, Anbar Governorate - Iraq.*

INTRODUCTION

Natural radiation is something people are exposed to regularly, and it mainly comes from two large sources: Radiation from outer space and the decay of radioactive substances in the soil and rocks are the main sources. When radionuclides break down into other nuclides, ionising radiation in the forms of gamma rays, beta particles, and cosmic rays is given off. The daughter nuclides can either be stable or radioactive and can also break down further. Rainwater, surface water, and groundwater may have radioactive substances known as radionuclides. Since radionuclides present in water are often toxic and may lead to cancer, they can be risky for human health. Radioisotopes (18O, 2H, 3H, 13C, and 14C) assist in defining how groundwater moves, how long it remains in one place, how different layers of rock interact, the rate at which it recharges, how quickly radioactivity breaks down, water pollution, contamination rates, and how old sediments are in dam sediments or aquifers [1–3]. Rainwater, surface water, and shallow and deep water all contain different levels of radionuclides that have natural origins. Their composition (anions, cations, and various isotopes) is further impacted by their discharge from the rocks that comprise water reservoirs: Because of atmospheric pollution, radioactive and poisonous materials travel via the following processes: oxidation-reduction, settling,

transportation, erosion, and breakdown. As a result, these elements end up in rain and water. [4–6]. The environment has both terrestrial and alien types of radiation. From the point of view of environmental safety and public education, collecting radioactivity data and calculating radiological factors is extremely important. The natural radioactivity in soil, rock, sand, and other parts of the environment greatly determines the dose a biological system gets [7]. Gamma-ray spectrometry is used most often to find out how much 238U, 232Th, and 40K is in environmental samples. How radionuclides, trace elements, organic compounds, and other substances from precipitation interact with soil depends on things like soil type, density, texture, and humidity, according to [8]. Radiation is present everywhere and people have been in contact with it all the time. Nature is one of the major origins of radiation. Different studies indicate that significant radiation comes from radionuclides such as 238U, 232Th, and single instances of 87Rb, 40K, which are found in nature [9].

The amount of radon gas in shallow groundwater samples from Abu-Jir, Anbar governorate, was determined using the Rad-7 detector. Radon gas levels in the samples ranged from a high of 9.3 Bq/L to a low of 2.1 Bq/L, with an average of 6.441.8 Bq/L. Although annual values can range from 33.945 μSv/y to 7.66 μSv/y, people typically receive an annual dose of 0.145±0.06 μSv/y.[10].

Soil, rock, water, granite, and building materials, along with 40K, 238U, 232Th, and their decay products, emit gamma radiation. This radiation makes up part of what is called terrestrial background radiation and is largely responsible for the cumulative dose received by humans [11]. Other nuclides do not contribute much to the sum of the total activity. Once radionuclides are in the environment, whether man-made or natural, they can enter the food chain as plants and animals take them up [12]. Radiation from sediments might affect the body as a whole, either when inhaled or when exposed to its emissions externally [13]. A less obvious risk to all living organisms is ionising radiation from the sun, other celestial bodies, soil, and the Earth's surface. Of all the radiation individuals are exposed to, about 87% comes from natural sources. To avoid health harm, it is necessary to measure the amount of radiation that people are exposed to from various sources. [14].

Several studies have been made to measure the amount of radioactivity in soil samples with different techniques. Basim Khalaf Rejah & et al. (2020) [15] are an example, as they studied Southern Al-Dora Region soil in Baghdad using a NaI (Tl) gamma ray detector to find natural and artificial radioactive elements. They found that most of the study locations are not at risk from radiological hazards. Haneen A. Jassam and Salih The study shows that radiation exists along the Abu- Jeer Fault Zone but cannot isolate its source from the other phenomena present.

Being close to the Abu-Al-Jeer Fault Zone is important in the research area, mainly because this fault relates to several things such as springs and leaks of hydrocarbons. This study proved that radiation exists on this fault, however, the source cannot be determined from the linked events. There is a noticeable difference in the soil samples, especially in the bituminous samples from Abu-Jir village, which are made up of clay minerals and organic matter [16]. Being present in soil, natural radionuclides lead to exposure both inside and outside. Soil is an important part of our environment used for many purposes, such as production of goods, playground surfacing, and street construction. Finding radionuclides in local soil is crucial since the findings from this study may serve as a foundation for upcoming environmental research. In order to learn more about natural and industrial radioactive pollution, the study aims to determine the levels of 238U, 232Th, and 40K contamination in soil, water, and asphalt in a few cities in Heet, west of the Anbar Governorate, Iraq. Additionally, we research the rate of external gamma doses and ensure that the public is informed about radiation safety.

EXPERIMENTAL PART

Study area

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The district of Heet and the sub-district of Abu Al-Jeer, which is west of Anbar Governorate, were studied. Most of the study was focused on places like residential, traffic, and industrial areas where people live. Figure (1) identifies the first place of sample collection in Heet, Figure (2) identifies the second place in Abu Al-Jeer, and Figure (3) shows a map of all locations. Table (1) contains both the coordinates and symbols for some areas of Heet and Abu Al-Jeer in the study area.

Table (1) Coordinates and symbols of some areas of Heet and Abu Al-Jeer under study

	Code	Sample type	Study areas	Coordinates	
				E	N
1	Q 1	soil, water	Al-Khader neighborhood - Heet	42.88697°	33.26556°
2	Q 2	soil, water	Al-Askari neighborhood 1 - Heet	42.88590°	33.26755°
3	Q 3	soil, water	Al-Askari neighborhood 2 - Heet	42.88979°	33.25957°
4	Q 4	soil, water	Al-Maamoura neighborhood - Heet	42.83230°	33.62244°
5	Q 5	soil, water, asphalt	Al-Khalidiya neighborhood - Heet	42.83049°	33.62545°
6	R 1	soil, water, asphalt	Abu Al-Jeer	42.77385°	33.61014°
7	R 2	soil, water, asphalt	Abu Al-Jeer	42.73523°	33.70679°
8	R 3	soil, water, asphalt	Abu Al-Jeer	42.81466°	33.63970°

Figure (1) Locations of (Q) group

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Figure (2) Locations of (R) group

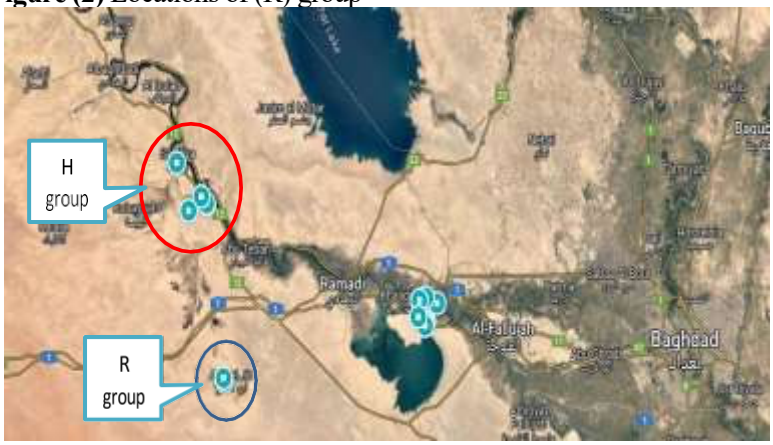

Figure (3) Locations of all groups

SAMPLE PREPARATION

Soil samples collection

Random soil samples were taken to 30 cm depth with a stainless-steel shovel, with each spot's GPS location being written down. For laboratory use, the soil samples were put into plastic bags provided with a modelling form containing the sample number, area's name, and details about the sites, including labs and factories, garages, and population sizes.

Natural asphalt samples

Natural asphalt was collected randomly from different places including residential, commercial, industrial, and rural areas. The samples were placed in special boxes that had a modelling form for each area, which showed the sample number, area name, information about the area, what it has like labs, factories, car garages, population number, and was numbered by area and district for laboratory work.

Collection of water samples

The samples were taken in 500 mL plastic bottles and then filtered to take out impurities and suspended particles. The samples came from (residential, industrial, traffic, and rural) areas in the district of Heet and Abu Al-Jeer area.

Radioactivity analysis

To remove all the moisture, each soil sample was roasted for two to three hours at 105 °C in an electric oven. After being processed in a mill, the samples were sieved through a 2 mm screen. To prevent the radon gases from escaping, the samples were put within a cylindrical plastic jar that

had screw covers. Prior to being filled, all samples were measured using an electronic weighing balance. To meet the requirements for secular equilibrium, a 1 kilogramme sample of each soil was stored for at least a month [17]. The 3"×3" crystal Hyper Pure Germanium Detector (HPGe) was then used to analyse each sample.

To find the activity concentrations for each radionuclide in the soil, we used a Canberra (USA) gamma spectrometer with relevant accessories. The contaminating radionuclides in the soil were recognised through each sample. The HPGe detector is easy to move and has an efficiency higher than 42%. It is a state-of-the-art instrument made by ORTEC. At 60Co energy, the resolution of the detector is 1.32 MeV. To cheque the energy and efficiency calibration, the gamma-ray spectrometer is tested using a 60Co radioactive source. The detector efficiency measures the proportion of photons the detector picks up from the photons emitted by the source. So, it is necessary to carry out the calibration with care.

The spectrum from the models studied in the field was recorded after energy and efficiency calibration, and then the specific effectiveness (concentrations) was figured out as the ratio of activity to the mass of the sample, following [18].

$$SA(E_\gamma) = \frac{\text{Net}}{T \times I(E_\gamma) \times \varepsilon(E_\gamma) \times m} \quad (1)$$

Where (m) is the sample mass in kilograms. , $I(E_\gamma)$ is the intensity of the isotope at energy E_γ, and (T) is the measurement time in seconds (in this work, 7200 s).

Radiological parameters (Hazard Indices)

Radium equivalent activity (Ra_{eq})

The Raeq is applied to calculate the dangers connected to the materials containing 238U, 232Th and 40K in Bq/kg. This parameter will be very useful in having information on materials for housing construction, based on radiation safety standards. The Raeq value is an integral quantity of the natural radionuclides activities that include 238U, 232Th, 40K. It is calculated on the assumption where 370 Bq/kg 238U, 259 Bq/kg 232Th and 4810 Bq/kg 40K will approximate to give us similar gamma dose. It is determined by the following equation [18, 19]:

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_K \quad (2)$$

where C_K, C_U, and C_{Th} are the specific activities, measured in becquerels per kilogram, of ^{40}K, ^{238}U, and ^{232}Th, respectively. The globally permissible value for this parameter is 370 Bq/kg [18].

Gamma dose rate (Dγ)

Based on the concentrations of 238U,232Th, and 40K in the soil, it is possible to calculate the roof rates of the gamma rays from the Earth surface to outdoor air of 1 metre. For the calculation of Dγ in the air at the height of 1 m from ground the following equation [19] can be used:

$$D\ [nGy/h] = 0.462C_U + 0.621C_{Th} + 0.0417C_K \quad (3)$$

Annual effective dose equivalent

The AEDE for a person as a result of being outdoors is given at the constant value per AEDE for Gamma Dose Rate: 0.7 Sv/Gy. The times spent outdoors are by the use of occupancy factors of 0.8 (19/24) and 0.2 (5/24) respectively. The AEDE is given as: [18, 19].

$$AEDE_{outdoor}\ [mSv/y] = D\gamma[nGy/h] \times 8760[h/y] \times 0.7[Sv/Gy] \times 0.2 \times 10^{-6} \quad (4)$$

External hazard index

The radioactive elements are present in natural form in rocks and soil and when the decay, they radiate which may be harmful to human beings. The most important naturally occurring radioactive elements are 40K and elements of uranium and thorium series, which cause significant exposure of the humans to radiation. The below equation is used to calculate external radiation hazard index [19]:

$$H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (5)$$

The radiation hazard is negligible if the index value is less than one. The upper allowed limit for H_{ex} is equal to one [19].

Representative Gamma index (I_γ)

The I_γ is an important parameter used to determine the risk associated with gamma rays from natural radionuclides in samples, and it can be calculated with the following equation:

$$I_\gamma = \frac{C_U}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500} \leq 1 \quad (6)$$

From this factor, the values of the annual dose rate can be predicted; for example, an effective annual dose less than 0.3 mSv corresponds to $I_\gamma < 0.5$, and a value less than 1 mSv corresponds to $I_\gamma < 1$ [19].

RESULTS AND DISCUSSION

In the present work, the natural radioactive activity of the nuclides (238U, 232Th, 40K) were determined in (19) various samples of soil, water, and asphalt utilising a gamma-ray spectroscopy method; the results were placed in tables (2,3 and 4).

Table (2) Specific activity of ^{238}U, ^{232}Th, and ^{40}K in soil samples.

Sample Code	Specific Activity (Bq/kg)		
	^{238}U	^{232}Th	^{40}K
Q1	29 ± 5	19.6 ± 3.5	305 ± 50
Q2	27.6 ± 6.9	10.8 ± 2	399.6 ± 52
Q3	1.2 ± 0.3	2 ± 0.7	26 ± 5
Q4	23 ± 2	48.8 ± 6	398.2 ± 35
Q5	-	39.4 ± 15	343.2 ± 32
R1	810 ± 69	108.8 ± 18	382.6 ± 52
R2	108.9 ± 5	506.6 ± 89	435.7 ± 50
R3	30.4 ± 4	17 ± 3	184 ± 29
Average	147.2 ± 13.17	94.1 ± 17.15	309.28 ± 38.1
permissible global limit [17]	35	30	400

Table (3) Specific activity of ^{238}U, ^{232}Th, and ^{40}K in asphalt samples.

Sample Code	Specific Activity (Bq/L)		
	^{238}U	^{232}Th	^{40}K
R1	46.2 ± 10	17.1 ± 4	220 ± 45
R2	41.7 ± 6.5	48.4 ± 12.1	326.2 ± 51
R3	-	-	22.8 ± 5

.

Table (4) Specific activity of ^{238}U, ^{232}Th, and ^{40}K in water samples

Sample Code	Specific Activity (Bq/L)		
	^{238}U	^{232}Th	^{40}K
Q1	-	-	30 ± 5
Q2	-	-	26.8 ± 4
Q3	-	-	36 ± 5
Q4	-	-	33 ± 6
Q5	-	-	34 ± 3.8
R1	-	-	22 ± 3.1
R2	-	-	7.4 ± 1
R3	-	-	24.1 ± 7

Average	-	-	26.66 ± 4.36

From table (2) The specific activity values of the above elements in the soil samples and the standard deviations of these values are shown along with it, we observe that the values of concentration of 238U ranged from (810 ± 69) Bq/kg in the R1 sample to Abu Al-Jeer region to (1.2 ± When we compare this result with the results of the past studies we find it close to the study (Amin, R.M., 2012) [20] and less than (Orgun, Y. & et al.,2007) [21] and (Petrescu & et al., 2011) [22], as for the 232Th concentration, the maximum value was (506.6 ± 89) Bq/kg in R2 sample located in (Abu Al-Jeer region), the minimum value is (2 ± 0.7) Bq/kg in Q3 sample and average is about(94.1 ± 17.15 On the contrary, the maximum value of 40K was (435.7 ± 50) Bq/kg in R2 sample (in the (Abu Al-Jeer region)). The lowest recording (26 ± 5) Bq/kg was found in Q3 sample, which had average of approximately (309.28 ± 38.1) Bq/kg, and it is lesser than the acceptable global limit (400 Bq/kg), while comparing the levels with the previous studies we see that it lies closer to the study Figure 4 shows the activity specific from the present study and the allowable global limit values.

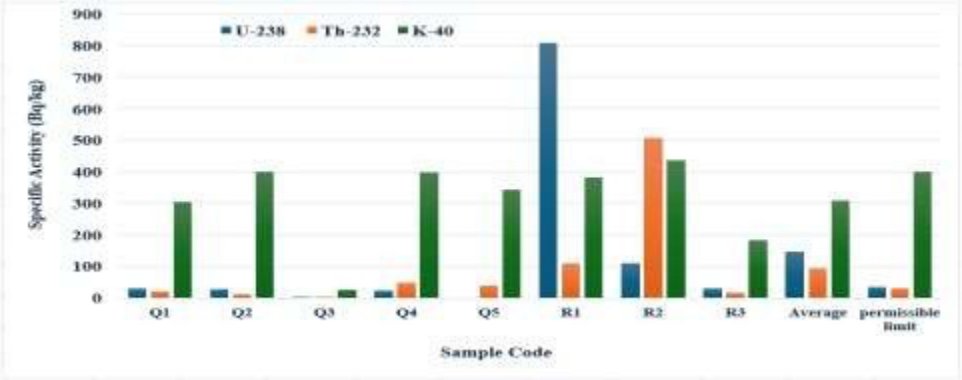

Figure (4) shows a chart of the specific activity of the soil samples compared to the permissible limits.

Table (5) The comparison of the current study results with the results of previous studies for various countries for the soil samples.

Specific activity (Bq/kg)				
Country	^{238}U	^{232}Th	^{40}K	Ref.
Egypt	137	82	1082	[20]
Turkey	174.78	204.69	1171	[21]
Romania	559.7	91.52	-	[22]
Nigeria	40.44	94.44	134.25	[19]
Nigeria	63.29	226.67	832.59	[23]
Malaysia	66	82	310	[24]
Kazakhstan	37	60	300	[24]
Turkey	95.5	110.4	1273	[25]
Iraq	38.22	42.99	16.64	[15]
Iraq	147.2	94.1	309.28	Present study

Regarding asphalt samples, three asphalt samples were studied and analysed, which were taken from the springs belonging to Abu Al-Jeer area. The results are given in the table (3). Sample R1 and R2 had the highest concentration of 238U that was (46.2 ± 10) Bq/L and (41.7 ± 6.5) Bq/L respectively and nothing was observed in R3 sample. As far as 232Th is concerned, the maximum value was recorded in sample R2 at (48.4 ± 12.1) Bq/L and the minimum value was in sample R1

at (17.1 ± 4) Bq/L, and its presence was not detected in sample R3. The activities of 40K were between (326.2 ± 51) Bq/L in sample R2 with the lowest activity of (22.8 ± 5) Bq/L in R3.

Also, (8) the samples of water were taken in varying locations as illustrated in table(4). The amount of radionuclides 238U and 232Th were not found in all samples as it was less than the value of the detection limit for the measurement system, while the specific activity for 40K isotope varied between (36 ± 5) Bq/L in R3 to (7.4 ± 1) Bq All the results were significant lower than the allowed global limit.

From the results received from this study it would be clear that the highest concentration among all the samples taken for measurement was in locations R1 and R2 while the lowest concentration was in place Q3. The reasons for this are that these areas squat down to the Abu-Al-Jeer Fault Zone, related to such phenomena as springs and hydrocarbon seepage. Previous works have indicated the presence of radiation as well as a variety of soils and rocks in this area. Also, geographical location and the difference in the conditions of the weather in different regions are among these factors.

Radiological risk factors were also calculated for soil and asphalt samples concentration levels and being (Raeq, Dγ, AEDEout, Iγ, Hex) and they were placed in tables (6 and 7). The findings reveal that the average of (Raeq, AEDEout and Hex) for the soil were (145.760 ± 10.284 Bq/kg, 0.082 mSv/y and 0.394), respectively, which is in less than the internationally permissible limit of (3 Though the result of the average of (D and Iγ), gave (66.667 nGy/h and 1.037), which was also a little above the global permissible limit of (55 nGy/h and 1), respectively. As a whole, the levels of radiological risk factors are rather high and could be dangerous in some regions, but in the others, they do not affect the public health.

For asphalt samples, the average results of all risks parameters were below the global limit.

Table (6) The Radiological parameters (Ra_{eq}, Dγ, $AEDE_{out}$, Iγ and H_{ex})for soil samples.

No.	Sample Number	Ra_{eq} (Bq/Kg)	Dγ (nGy/h)	$(AEDE)_{out}$ (mSv/y)	I_γ	H_{ex}
1	Q 1	80.513 ± 8.973	37.95	0.05	0.59	0.22
2	Q 2	73.813 ± 8.591	35.94	0.04	0.56	0.20
3	Q 3	6.062 ± 2.462	2.85	0.00	0.05	0.02
4	Q 4	123.445 ± 11.111	56.71	0.07	0.91	0.33
5	Q 5	82.768 ± 9.097	38.11	0.05	0.62	0.22
6	R 1	995.044 ± 31.544	455.89	0.56	6.74	2.69
7	R 2	866.887 ± 29.443	374.47	0.46	6.08	2.34
8	R 3	68.878 ± 8.299	31.99	0.04	0.50	0.19
Average		**287.176 ± 12.653**	**129.238**	**0.181**	**2.006**	**0.776**
permissible global limit [26]		**370**	**55**	**0.08**	**1**	**1**

Table (7) The radiological parameters (Ra_{eq}, Dγ, $AEDE_{out}$, Iγ and H_{ex}) for asphalt samples.

No.	Sample Number	Ra_{eq} (Bq/Kg)	D (nGy/h)	$(AEDE)_{out}$ (mSv/y)	I_γ	H_{ex}
1	R 1	87.593 ± 9.359	40.85	0.05	0.63	0.24
2	R 2	136.029 ± 11.663	62.10	0.08	0.98	0.37

3	R 3	1.756 ± 1.325	0.95	0.00	0.02	0.00
Average		**75.126 ± 7.449**	**34.633**	**0.042**	**0.540**	**0.203**
permissible global limit [26]		**370**	**55**	**1**	**1**	**1**

CONCLUSIONS

This research will be used to see the estimation of pollution of the concentrations of 238U, 232Th and 40K in the soil, water and asphalt of some Heet city in the west of Anbar governorate to determine the natural and industrial radioactivity contaminations. That is why the Gamma-ray spectroscopy technique with the help of the Germanium detector was applied to estimate the levels of radioactivity of natural radionuclides (238U, 232Th, 40K) of different samples of soil, water, and asphalt taken from (8) locations of the Heet district and its neighbor.

The results determined that Abu Al-Jeer area showed the highest concentrations of U and Th in the soil samples collected, also the specific activity average of these nuclides exceeded the permissible global limit. The outcomes may be explained by the fact that the soil is a natural deposit, containing within it, minerals as well as organic materials that are subject to a number of influences, such as weather conditions, terrain and type of rocks from which the soil is created. Furthermore, its geographical location. All these factors take a significant and crucial role and make significant contributions to the level of radioactivity in the samples.

Regarding the water samples, the observed outcomes reveal that there is no radioactivity in water and the explanation for the fact lies somewhere in the fact that water is continuously renewed to what it is due to a number of causes: rain and groundwater, as well as human actions.

Based on calculations of the radiological parameters of soil samples, it was found that all the radiological parameters studied are less than the permissible global limit except the absorbed dose rate (Dγ) and the gamma factor (Iγ) that was found to be more than the permissible global limit at some samples as a result of high concentrations of Th and U in the samples. As far as the radiological parameters of asphalt samples are concerned, they were below the world limit.

On average, it can be summarised that, the Abu Al-Jeer region is relatively radioactive to some of its areas by a little above the world acceptable limits. Thus, there has to be some periodic and continuous involvement to test radioactivity in those areas studied to preclude any possibilities of harming the population.

RECOMMENDATIONS

1) Analyse the same region through the use of other techniques and compare with this study.

2) Carry out a similar study to the present study on various samples, for example, air, giving various approaches.

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