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Vol.29; No.2. May-August | 2021

Determining the Natural Radioactivity Level of Soil Samples from Halabja City

Kamal O. Abdullah ¹	Adel M. Hussien ¹	
Oday K. Azzawi ²	Shalaw Zrar Sedeeq ³	

1Physic Dep.- College of Science-University of Sulaimani- Iraq kamal.abdullah@univsul.edu.iq ²University of Halabja - College of Science- (Physic Dep.). ³University of Koya - College of Science- (Physic Dep.)

*Corresponding author email: : kamal.abdullah@univsul.edu.iq

D 1 1 45 (44 (0000		
Received: 15/11/2020	Accepted: 15/4/2021	Published: $1/7/2021$

<u>Abstract:</u>

The samples of soil were collected from 7 locations in Halabja city, in February 2019. The natural radioactivity of the soil samples has been studied using High Pure Germanium (HPGe) detection system. The activity concentration (A_s) of the natural series; ²³⁸U, ²³²Th and ⁴⁰K was determined for the samples. The values of A_s were ranged (45.641 ± 3.252 - 92.862 ± 4.295), (61.287±2.996 - 135.773 ± 18.366) and (227.9 ± 4.874 - 413.0±7.060) Bq kg⁻¹, respectively. The radium equivalent activity ($R_{(eq)}$) was determined with range (52.006 – 130.518) Bq kg⁻¹. The total absorbed dose rate (*H*) calculated from the activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K in the studied area ranges from 25.53 nGy h⁻¹ to 58.921 nGy h⁻¹. The calculated values of total annual effective dose (H_{ann}) for the soil samples of the study area ranged from 0.156 to 0.361. Since these values are less than unity, the EC report (European Commission, radiation protection report) concludes that the soil from these areas is healthy and poses no substantial radiological risk to the population.

Keywords:

Soil, Specific Activity (As), HPGe–Detector, Total Annual Effective Dose (Hann), Halabja- City

Citation:

Kamal O. Abdullah^{1,} Adel M. Hussien^{1,} Oday K. Azzawi², Shalaw Zrar Sedeeq². **Determining the Natural Radioactivity Level of Soil Samples from Halabja City**. Journal of University of Babylon for Pure and applied science (JUBPAS). May-August, 2021. Vol.29; No.2; p:177-187.



1.INTRODUCTION

The main radioactive materials in naturally occurring radioactive materials are long–lived radionuclides such as ²³⁸U, ²³⁵U and ²³²Th and ⁴⁰K. Natural radioactivity and terrestrial gamma dose resulting from naturally occurring radioactive in building materials are largely dependent on geological and geographical conditions, so natural radioactivity concentrations in materials, especially soil, differ from one area to the next around the world [1].

The three heavy nuclides, 238 U, 235 U and 232 Th, also are the origins of the three natural radioactivity chains, the chains proceed through a series of α and β decays that end at three isotopes of lead. Natural radioactivity can be divided into "fossil" radioactivity, which results from elements present during the Earth's formation, and "cosmogenic" radioactivity, which results from elements continuously released in the atmosphere by cosmic-rays [2].

Radioactive decay is a spontaneous change within the nucleus of an atom which results in the emission of particles or electromagnetic radiation. Charged particle decay is the most common form of radioactive decay, with spontaneous fission being one of a few rarer processes. The mass of the product is lower than the mass of the initial nuclide, which drives radioactive decay[3].

Nuclear radiation comes in a variety of forms, concentrations, and energies. Furthermore, a radionuclide may have several modes of decay. The existence of significant activities of more than one radionuclide in a sample will make the study much more difficult. In addition, the various parent– daughter nuclide decay schemes, equilibria between parent and daughter radionuclides, and the rates of radionuclide decay are all factors to consider [4].

The properties of nuclear radiation and the mechanisms whereby nuclear radiation dissipates its energy in matter, form the basis for the methods of detection and measurement of radionuclides [4]. There are many techniques for analyzing radioactivity and many types of detectors for measuring which they designed in the gaseous, liquid and solid state [5]. In this study, we used a method of high accuracy Gamma-Spectroscopy Analyzer (HPGe-System) to identify the concentration of some natural radionuclide in the soil samples of some areas inside and surrounded the Halabja city, because HPGe detector has a high resolution if compared with the other gamma detectors, especially other semiconductor detectors [3].

Halabja is a city in Iraqi Kurdistan-Region and the capital of Halabja Governorate, located about 240 km northeast of Baghdad and 14 km from the Iranian border as shown in Fig.1. This city was bombarded by a chemical bomb during 1988 by a Saddam regime more than 5000 people were dying.

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Fig.1 location of Halabja in map of Iraq and in Kurdistan region map

2. Materials and methods

2.1 The study area

The province of Halabja located in the east of Kurdistan-Region- Iraq between longitude 45° 58' 59" E and latitude 35° 10' 59" N, and surrounded by Hawraman and Shinrwe mountains on the north stretching across the Iran–Iraq border, and Balambo mountain and Darbandikhan dam on the south. In the west it surrounded by Sirwan River as shown in Fig.1. Approximately 50% of Halabja area are agricultural land, the study area has an elevation of 690 m above sea level [6]. Soils formed over Quaternary alluvial deposits, transported from surrounding high mountains which consist of different ages of limestones. Soils are classified as chestnut soils with deep soil profile, rough, broken and stony soil surface. Land use in Sharazur plain is predominantly arable, and forests surround the flat arable lands of the plain[7].

2.2 Sample preparation and Measuring Technique

Thirteen samples of soil were collected from a depth (10 and 20) cm in different locations inside of Halabja city, as shown in Table 1.



No	D ogions and Location	Pos	sition	Depth of	Code of
INO.	Regions and Location	Latitude(N)	Longitude (E)	samples	samples
1	Halabja-Zamaqi south	35° 1327.5	45° 5808.7	10cm	Z1
2	Halabja-Zamaqi south	35° 1327.5	45° 5808.7	20cm	Z2
3	Halabja- Center (Julakn)	35°1031.1	45° 5912.4	10cm	J3
4	Halabja-Center (Julakan)	35°1031.1	45° 5912.4	20cm	J4
5	Halabja-Center (Jaleela)	35° 3229	45° 1455.4	hillside	J5
6	Halabja-Outside (Ababailee)	35° 1025.6	46° 0140.3	10cm	Q6
7	Halabja-Center (Ababailee)	35° 1025.6	46° 0140.3	20cm	Q7
8	Halabja-outside (Anab)	35° 1230.5	46° 0058.02	10cm	A8
9	Halabja-outside (Anab)	35° 1230.5	46° 0058.02	20cm	A9
10	Halabja-outside (Anab-group grave)	35° 1152.9	46° 0109.1	10cm	A10
11	Halabja-outside (Anab-group grave)	35° 1152.9	46° 0109.1	20cm	A11
12	Halabja-Center (Shahidan)	35° 1038.06	45° 5943.38	10cm	S12
13	Halabja-Center (Shahidan)	35° 1038.06	45° 5943.38	20cm	S13

Table (1) Position,	location,	codes and	depth of	the samples.
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The samples were crushed to obtain a homogenous powder and dried in an oven to ensure that any moisture or dampness was removed. All samples were then screened to pass through a 2 mm sieve, then they weighted by 1kg and putting in a polyethylene plastic Marinelli beaker were sealed with a tape tightly to prevent gas escape, and stored for about one month to allow secular equilibrium between ²²²Rn and ²²⁶Ra and its daughter products in uranium chain. This common method of sample preparation steps was done by some previous studies [8, 9]

Gamma spectrometry analyzer connected to HPGe detector to measure the natural radionuclide (gamma) concentration of the soil samples, by using a HPGe detector with large crystal diameter 70.6 mm and length 70.7 mm. (41 % efficiency p-type detector, accurate energy resolution1. 97 keV for the energy 1333 keV ⁶⁰Co line). The whole gamma system was manufactured by Canberra Industries, Inc. The detector fed by the high voltage that fourth time greater than the power supply on NaI (Tl) detectors [10, 11].

The Gamma spectrometry was shielded completely by a thick shield (5cm) of lead (5x10x50) cm, one can find elsewhere the method about. The system's gamma energy and performance calibrations were carried out with the help of standard sources such as (⁶⁰Co, ¹³⁷Cs). The time it took for each soil sample's spectrum to be generated was 21,600 seconds [12].



On the basis of the energy peaks of the progenies, radionuclide analyses of soil samples were carried out. The concentrations of ²¹⁴Pb's decay products (295.1 keV with 18.7% and 352 keV with 35.8%), ²¹⁴Bi (609.3 keV with 45%, and 1120.2 keV with 15%) and ²²⁶Ra (186.1 keV with 3.3%) were taken to calculate total concentration of ²²⁶Ra, while the specific activity of ²³²Th has been calculated based on the energy peaks of ²¹²Bi (727.3 keV with 6.7%), ²²⁸Ac (338.4 keV with 12.4%, 463.1 keV with 4.6%, 911.2 with 29%) and ²⁰⁸Tl (583.0 with 30.58%). However, the activity concentration of ⁴⁰K was calculated using its gamma ray peak (1460.83 KeV, 10.67 %), which matched studies by Darwish D. A. E. and others [13].

2.3 Activity concentrations and Gamma Dose Rate

The radionuclide activity concentrations were calculated using the equation [14],

$$A_{\mathcal{C}} = T_{YM\varepsilon}$$
(1)

where Ac is the radionuclide's activity concentration in Bq/kg, C_n is the net count area per second, T_{γ} is the absolute transfer likelihood of the specific gamma, M is the mass of the sample (kg), and ε is the specific gamma energy's detector effeciency. A radiological index that represents the existing of natural radionuclide (²²⁶Ra, ²³²Th and ⁴⁰K) activity in the terrestrial is called radium equivalent activity (Ra_{eq}) [15, 16].

 $Ra_{eq} = A_{Ra} + 1.43 A_{Th} + 0.077 A_{K}$ (3) The natural radionuclides participation ratio as; 10 B_q kg⁻¹ of ²²⁶Ra, 7 B_q kg⁻¹ of ²³²Th

and 130

 $B_q k_{g^{-1}}$ of ${}^{40}K$, to produce an equal gamma dose rate (Ra_{eq}), its maximum limit is 370 $B_q k_{g^{-1}}$.

The absorbed dose rate (H) is formulated from the As of ^{226}Ra , ^{232}Th , and ^{40}K , and using the conversion factor in units (nGy hr⁻¹ / Bq kg⁻¹) with the values for soil; 0.4551226 for ^{226}Ra , 0.5835, for ^{232}Th and 0.0429 for ^{40}K , as given.

 $H_{(nGy h-1)} = 0.4551 A_{Ra} + 0.5835 A_{Th} + 0.0429 A_K$ (4) The effective dose exposure to the human beings in one year due to the soil radioactivity can be estimated using the conversion factor from absorbed dose in air to effective dose received by adults (0.7 Sv Gy⁻¹) given in the UNSCEAR report [17]. The total annual effective dose rate (H_{ann}) in unit (mS_v y⁻¹). The H_{ann} consider as the best radiation risk indicator can be calculated from [18].



 $H_{ann}(mS_v y^{-1}) = H(nGy h^{-1}) * 8760 h y^{-1} * 0.7 Sv Gy^{-1} * 10^{-6}$

 $= 0.006132 * H(nGy hr^{-1})$

(5)

The outdoor (or indoor) H_{ann} calculate by multiplying the above equation by 0.2 for outdoor and 0.8 for indoor.

3. Result and Discussion

An empty Marinelli beaker with the same volume (1000 cm³) was counted with the same geometrical conditions as the samples to determine the background distribution due to naturally occurring radionuclides in the atmosphere around the detector. The background spectra were used to correct the net -ray peak areas for the studied isotopes after the activity and background spectra were measured for a total of 21600 seconds. Table 2 shows the average radionuclide concentrations in the seven unique Halabja locations. Figures 2, 3 and 4 display the specific activities (A_s) of ²³⁸U, ²³²Th, and ⁴⁰K in Bq kg⁻¹ dry weight. The unique activities for ²³⁸U ranged from (45.641 to 92.862) Bq kg⁻¹ with sample J5 from the mountain having the highest value (Halabja-Jaleela).). The activities range for ²³²Th were from (61.287 to 135.773) Bq kg⁻¹ in which sample A11 has the highest value which is the outside of Halabja (Anab). The ⁴⁰K activities are in the range from (227.9 to 413) Bq kg⁻¹ in which sample J3 and J4 has the highest value which is from (Halabja-Julakan).







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Code of	As (238U)	As (²³² Th)	$\mathbf{A}_{\mathbf{s}}({}^{40}\mathbf{k})$	
samples	Bq k _{g-1}	Bq k _{g-1}	Bq k _{g-1}	
Z1	71.892 ± 3.997	98.734 ± 17.098	312.2 ± 5.739	
Z2	87.865 ± 3.982	77.259±4.27	330.3 ± 5.896	
J3	75.383 ± 4.29	75.819±3.575	413.0 ± 7.059	
J4	74.757 ± 3.869	91.652±3.917	413.0 ± 7.060	
J5	92.862 ± 4.295	97.413 ± 17.169	227.9 ± 4.874	
Q6	81.341 ± 4.384	80.641±3.994	268.4 ± 5.261	
Q7	72.984 ± 4.13	81.185±3.562	274.4 ± 5.270	
A8	45.641 ± 3.252	79.439 ± 16.124	286.6 ± 5.751	
A9	49.805 ± 3.225	68.161±3.39	291.7 ± 5.557	
A10	86.493 ± 4.548	89.196±4.161	402.8 ± 7.319	
A11	74.422 ± 4.108	135.773 ± 18.366	357.6 ± 6.686	
S12	51.837 ± 3.697	61.28 <mark>7±</mark> 2.996	287.1 ± 5.781	
S13	72.740 ± 3.807	80.67 <mark>5±</mark> 3.824	354.4 ± 6.532	

Table 2 Radioactivity concentration (A_s) of $(^{238}U, ^{232}Th \text{ and } ^{40}k$) of the soil samples

Table 3 presents the specific activity in the soil samples of the other work of different regions that surrounding Halabja city [19] for comparison with our results.

Table 3 Radioactivity concentration (As) of $(^{238}U, ^{232}Th \text{ and } ^{40}k$) from different areas of the region

T	²³⁸U (Bq kg ⁻¹)		²³² Th (Bq kg ⁻¹)		⁴⁰ k (Bq kg ⁻¹)	
Locations	10cm	20cm	10cm	20cm	10cm	20cm
Halabja	107.632	103.044	16.810	15.621	287.49	256.41
Serwan	68.2	69.688	15.908	15.908	209.79	209.79
Byara	45.508	42.532	16.646	17.384	217.56	202.02
Khurmal	84.568	82.46	17.302	16.236	264.18	248.64

The distribution of ²²⁶Ra, ²³²Th, and ⁴⁰K in soil is not standardized due to radiation exposure. To compare the specific activity of samples containing different concentrations



of 226 Ra, 232 Th, and 40 K, the radioactivity was measured in terms of radium equivalent activity (Ra_{eq}) in the unit Bq kg⁻¹.

The average Ra_{eq} in the soil samples ranged between (52.002 to 130.518) Bq k_g^{-1} . The determined radium equivalent is found to be less than the permitted maximum value of 370 Bq kg^{-1} [20]. The other radiological indexes, as shown in table 4, were determined for external -radiation due to a particular combination of various natural activities in a sample, especially the total annual effective dose rate (H_{ann}) in (mS_v y⁻¹). Finally, according to the Radiation Protection (European Commission, Radiation Protection Report), soil from these regions is safe and without being poisonous to population.

Table 4 Radium equivalent (a(eq)) in Bq kg-1, absorbed dose rate (H) in , total annual effective dose rate (H_{ann}) in (mS_v y⁻¹) , and its out and in door for each sample.

Code of samples	Ra(eq)	H (nGy hr ⁻¹)	<i>H</i> ann (mS _v y ⁻¹)	Hann outdoor (mSv y ⁻¹)	Hann Indoor (mS _v y ⁻¹)
Z1	88.173	41.01	0.251473	0.050294	0.201178
Z2	63.786	31.624	0.193918	0.038783	0.155134
J3	74.615	36.56	0.224185	0.044837	0.179348
J4	87.629	41.972	0.257372	0.051474	0.205897
J5	119.291	53.163	0.325995	0.065199	0.260796
Q6	78.648	36.841	0.225909	0.045181	0.180727
Q7	70.159	34.888	0.213933	0.042786	0.171146
A8	75.653	35.047	0.214908	0.042981	0.171926
A9	59.93	28.854	0.176932	0.035386	0.141545
A10	64.677	32.599	0.199897	0.039979	0.159917
A11	130.518	58.921	0.361303	0.07226	0.289024
S12	52.006	25.53	0.156549	0.031309	0.125239
S13	75.049	36.125	0.221518	0.044303	0.177214

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Fig. 2 The activity concentrations of (²³⁸U) series



Fig. 3 The activity concentrations of ²³²Th series

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Fig. 4 The activity concentrations of ⁴⁰k

4. Conclusion

From this work we conclude that the area under investigation has a normal dose level of natural radionuclides and isotopes (background radiation), although there are some high values of concentrations of ²³⁸U, ²³²Th and ⁴⁰K but still within the worldwide standard value. These dose rate fluctuations return to the combination of the soil samples from these areas of Halabja city.

Conflict of interests.

There are non-conflicts of interest.

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