NATURAL RADIOACTIVITY LEVELS OF AGRICULTURAL AND VIRGIN CLAY SOIL SAMPLES AT AL NAJAF GOVERNORATE

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http://dx.doi.org/10.26739/2573-5616-2017-8-8-1

Abstract

The gamma radiation has been measured to determine natural radioactivity of ^{238}U, ^{232}Th and ^{40}K in the agricultural and virgin soils in Najaf city, south western part of Iraq. Thirty soil samples were collected from agricultural clay and virgin soil for different location and at (10,20,30,40 and 50 cm) depth. These samples have been studied by using gamma-ray spectroscopy which consists of high counting efficiency NaI(Tl) detector. The specific activity of the primordial nuclides for ^{238}U ranged from 10.89±0.61 to 25.98±1.00 Bq/kg with an average of 17.65±0.80 Bq/kg for clay soil and from 0.56±0.13 to 9.38±0.60 Bq/kg with an average 4.72±0.39 for virgin soil. While the ^{232}Th radioisotopes ranged from 3.19±0.20 to 12.94±0.38 Bq/kg with an average of 8.28±0.33 Bq/kg for clay soil, and from 4.42±0.23 to 10.89±0.39 Bq/kg with an average 6.79±0.29 for virgin soil. The specific activity for ^{40}K ranged from 340.4±3.54 to 522.8±4.97 Bq/kg with an average of 419.83±4.10 Bq/kg for clay soil, and from 107.16±1.93 to 372.33±3.95 Bq/kg with an average 244.25±2.09 for virgin soil.

All values of radium equivalent dose, absorbed dose, internal and external hazard index an activity concentration index were lower than the world wide average, also compare with the values recommended by the international agencies and found to be within the permissible limits.

KEYWORLDS: natural radiation, virgin clay soil and its environmental, Nal(Tl).

Introduction

Terrestrial gamma radionuclide is a considerable part of the total from natural sources. Only nuclides with half-lives comparable with the age of the earth and present in terrestrial materials such as ^{232}**Th**, ^{238}**U**, and ^{40}**K** are of great interest as they cause external and internal hazards because of gamma ray emissions[1]. We currently live in a world of radionuclide's, where we inhale and ingest these radioactive substances daily in the form of food, water, and air as no place on the earth is free of radioactivity [2]. Fission product ^{137}Cs is strongly absorbed and maintained by soil particles, similar to natural radionuclide's, which are widely distributed at different soil depths. Therefore, knowledge on radionuclide distribution in soils is essential to control health risks to the affected population[3]. Soil is one of the most important sources of radioactivity. It is used as a raw material in the construction of roads, buildings, landfills, and playgrounds. Numerous studies conducted worldwide have shown that, ^{238}**U**, including its decay products in soils , rocks, and ^{232}**Th** in found in one type from sand such as monazite sands are the main sources of high natural background area [4]. The main sources of phosphorus for fertilizers and the primary material for the production of phosphate products are phosphate rocks. Phosphate fertilizers contain elevated natural uranium and its decay products [5]. Radionuclide's in phosphate rocks can enter the environment through different ways, such as usage of phosphogypsum in building materials and agriculture or fertilization of agricultural lands [6].

SAMPLES PREPARATION AND MEASUREMENTS :

The natural radionuclides were determined in (20) agricultural and virgin, Soils samples collected from (4) different regions in Najaf province as shown in table (1). Three types of soil were collected at depth (0- 50) cm from farms and virgin. The samples dried at (100°C) for 24 hours in oven as shown in figure (1). the containers were washed with dilute hydrochloric acid and rinsed with distilled water. Samples prepared for analysis by drying, sieving and kept moisture free by keeping (24) hours in the oven at 100C°. They were mechanically crushed and sieved through of 0.8mm pore size diameter to get sieved to get homogeneity. After that samples milled by soil blender, then about (1kg) of the homogenous soil sample was packed in Marinelli (polyethylene) beaker, carefully sealed and stored for at least 4 weeks before counting time to achieve secular equilibrium. Before use, the containers have been washed with dilute hydrochloric acid and rinsed with distilled water and coded to the specific code in order to distinguish between samples [7,8]. The specific activity of each radionuclide is calculated using the following equation [9, 10].

$$A(Bq \backslash kg) = \frac{C}{\varepsilon \cdot \gamma \cdot m \cdot t} \qquad \text{--------------------------(1)}$$

where **A** the specific activity of the radionuclide in Bq/kg, **C** background subtracted , **ε** the counting efficiency, **γ** the percentage of gamma emission probability of the radionuclide under study, **t** the counting time in second and **m** the mass of the sample in kg.

Radiation hazards due to the activity concentration of ^{238}U, ^{232}Th and ^{40}K were showed by two different indices. The most widely used radiation hazard index Ra_{eq} is called the Radium equivalent activity. The Radium equivalent activity is a weighted sum of activities of the above three radionuclides based on the estimation that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same γ-ray dose rates . Ra_{eq} is given by [11]:

$Ra_{eq} = A_U+(1.43\ A_{Th}) + (0.077A_K)$ ----------------------(2)

A_U, A_{Th}, and A_K are the specific activity values of uranium, thorium, and potassium in Bq/kg, respectively.

Absorbed dose defined the amount of energy ionizing radiation deposits in a unit mass of matter, such as human tissue, it is expressed in a unit called the gray, symbol Gy, where one gray is equal to one joule per kilogram. , submultiples of the gray are often used, such as the milligray, mGy, 10^{-3}gray. The gray is named after the English physicist Harold Gray , the average absorbed dose rate in air 1m from terrestrial sources of gamma radiation in soil is estimated as 55 nGy/h. The corresponding values relative to different types of fertilizer are calculated by using the following relation [12]:

$A.D = 0.427\ A_U + 0.662A_{Th} + 0.043A_K$ ----------------------(3)

where, A.D is the dose rate in nGy/h.

Annual estimated average effective dose equivalent received by a member was measurement by using a conversion factor of 0.7 Sv/Gy, which was used to convert the absorbed rate to human effective dose equivalent with an outdoor occupancy of 20% and 80% for indoors. The annual effective doses were determined as follows [12]:

Indoor (mSv/y) =Absorbed Dose (nGy/h) × 8760 h/y × 0.8 × 0.7 Sv/Gy × 10^{-6} ------(4)

Outdoor (mSv/y) =Absorbed Dose (nGy/h) × 8760 h/y × 0.2 × 0.7 Sv/Gy × 10^{-6} ------(5)

In this case, the annual effective dose rate (in nSv/h) was evaluated from the specific activity (in Bq/kg) of the radioactive series and the ^{40}K in soils and fertilizers [12].

Activity concentration indexes are used to estimate the dangerous due to gamma radiation similar with the natural radionuclides (^{238}U, ^{232}Th and ^{40}K), in the study matter, another radiation hazard index ,the activity concentration index (I_γ), is used and is defined as [12].

$$I_\gamma = \frac{A_U}{300} + \frac{A_{Th}}{200} + \frac{A_k}{3000} \quad , \qquad \text{------(6)}$$

If the value of the activity concentration index is 1 or less, the corresponding material can be used, with regard to radioactivity, without restriction. If the value exceeds 1,. the corresponding material cannot be used in the building [13].

The external hazard index is another criterion to assess the radiological suitability of a material. It is known as follows [14].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{258} + \frac{A_K}{4810} \qquad \text{--------(7)}$$

Where A_{Ra} , A_{Th} and A_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K in (Bq/kg). The hazard indexes of the values should be <1.

The internal hazard index is a criterion for index radiation hazard. an important role for internal exposure in a room is ^{222}Rn in addition to gamma rays. Radon and its daughter products by the internal exposure and quantified by the internal hazard index H_{in} . By a factor of two to allow for the short lived progeny and contribution from ^{222}Rn increased the radio toxicity of ^{238}U [14].

$$H_{in} = \frac{ARa}{185} + \frac{ATh}{259} + \frac{AK}{4810} \qquad \text{---------(8)}$$

The values of the index must be less than the unity in order to keep the radiation hazard to be insignificant unity corresponds to the upper limit of radiation equivalent activity (370 Bq/kg) .

RESULTS AND DISCUSSION

The specific activity values at ^{238}U ,^{232}Th and ^{40}K radionuclides for 15 soil samples are tabulated in Table (1), the value have been found to lie in the range 10.89 ± 0.61 Bq/Kg to 25.98 ± 1.00 Bq/Kg. with an average of 17.65 ± 0.80 Bq/Kg

Table 1: Specific activity and radium equivalent dose for agricultural clay soil.

No	Depth (cm)	Sample Code	Specific activity in (Bq/kg) ^{40}K	^{238}U	^{232}Th	Ra_{eq} (Bq/kg)
1	10	A1	375.1±3.74	20.03±0.82	3.77±0.22	54.30
2	20	A2	374.3±3.65	24.53±0.89	3.19±0.20	57.91
3	30	A3	385.4±3.73	14.41±0.69	3.73±0.28	49.42
4	40	A4	431.5±3.68	12.17±0.64	6.36±0.29	54.49
5	50	A5	340.4±3.54	10.89±0.61	6.38±0.46	46.22
6	10	B1	522.8±4.97	23.96±0.02	12.94±0.38	82.72
7	20	B2	481.6±4.47	13.39±0.71	10.19±0.35	65.04
8	30	B3	520.0±4.79	23.33±0.97	8.14±0.36	75.01
9	40	B4	421.8±4.10	18.62±0.82	9.78±0.37	65.08
10	50	B5	488.3±4.52	19.33±0.86	9.71±0.36	70.81
11	10	C1	422.2±4.12	13.94±0.71	9.22±0.40	59.63
12	20	C2	393.0±4.06	25.98±1.00	11.07±0.38	72.07
13	30	C3	400.6±3.09	16.63±0.77	10.73±0.34	62.82
14	40	C4	374.5±3.91	14.48±0.73	10.54±0.31	58.39
15	50	C5	365.1±3.92	13.08±0.71	8.52±0.34	53.38
Minimum			340.4±3.54	10.89±0.61	3.19±0.20	46.22
Maximum			522.8±4.97	25.98±1.00	12.94±0.38	82.72
Average			419.83±4.10	17.65±0.80	8.28±0.33	61.82
Worldwide average			400	32	10	370

The result shows the minimum value of specific activity for ^{238}U was recorded at depth 50 cm while the maximum value was recorded at depth 20 cm. for the ^{232}Th,the maximum value was calculated at 20 cm whereas the maximum value was in 10 cm. As concerns ^{40}K radionuclide the minimum value ways recorded at depth 50 cm while the maximum value was observed in depth 10 cm. as shown in table (1)

The variations of the radios of specific activity at ^{238}U ,^{232}Th and ^{40}K may be due to the type of fertilizers are used or to other reasons such as (PH) the radium equivalent activities also calculated and listed in Table (1)

Ra_{eq} Values vary from 46.22 to 82.72 Bq/Kg with an average 61.82 Bq/Kg.

Figure (1), (2), and (3) show the values of specific activity for ^{238}U ,^{232}Th and ^{40}K .

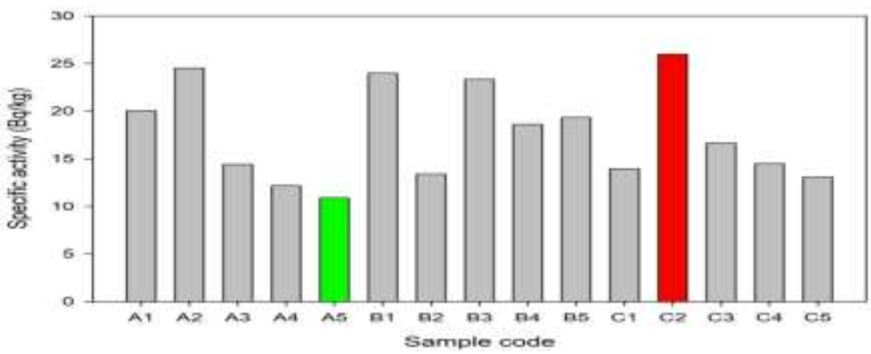

Figure (1): specific activity of Uranium in clay soil sample

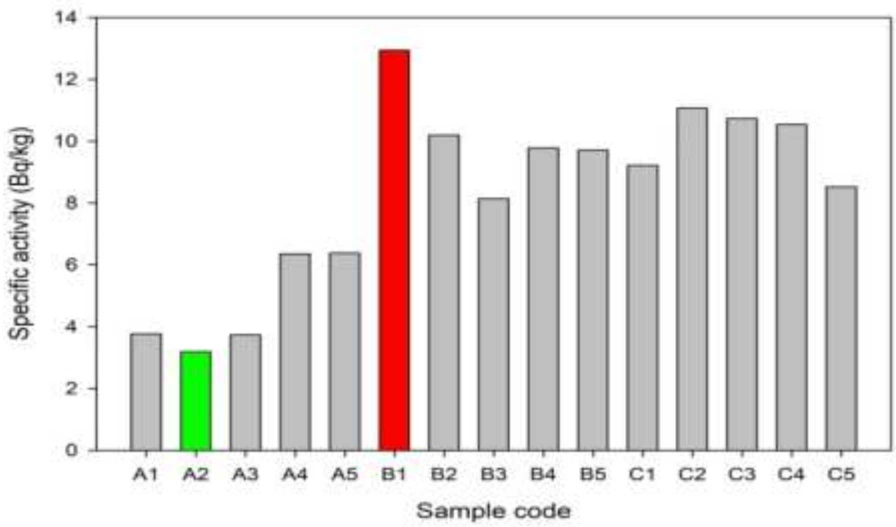

Figure (2): specific activity of thorium in clay soil sample

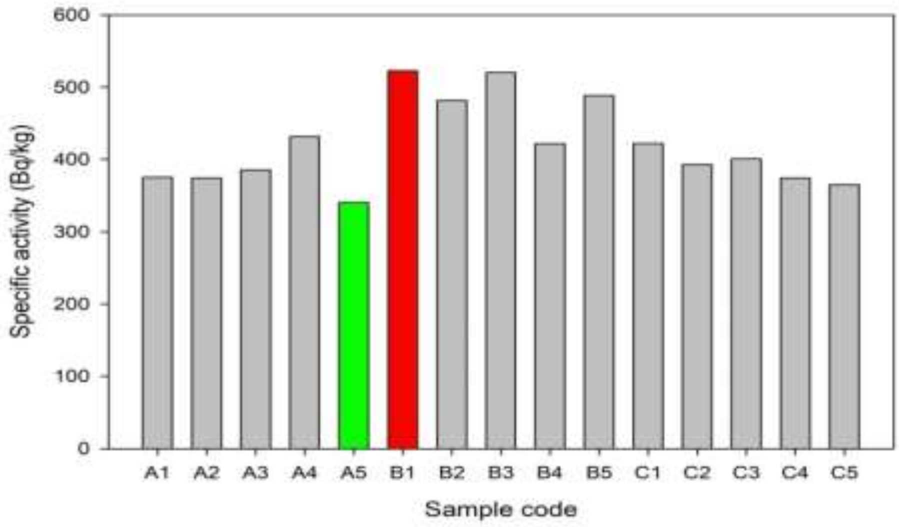

Figure (3): specific activity of ^{40}Kin clay soil sample

Virgin soil

The specific activity values of ^{238}U ,^{232}Th and ^{40}K radionuclides for 5 virgin soil samples are tabled in Table (2) as well as the Radium equivalent activity.

From this table we can find that the values of ^{238}U specific activity were ranged from 0.56±0.13 to 9.38±0.60 Bq/Kg with an average 4.72±0.39 Bq/Kg. while for ^{232}Th specific activity values the values have been found to lie in the range 4.42±0.23 to 10.89±0.39 Bq/Kg with an average 6.79±0.29 Bq/Kg. Specific activity values of ^{40}K sere range from 107.16±1.93 to 372.33±3.95 Bq/Kg with an average 244.25±2.09 Bq/Kg.

This result shows that the minimum value of ^{238}U was found in depth 30 cm while maximum value was found in depth 50 cm.

Table 2: Specific activity and radium equivalent dose for virgin soil.

No	Depth (cm)	Sample Code	Specific activity in (Bq/kg) ^{40}K	^{238}U	^{232}Th	Ra_{eq} (Bq/kg)
1	10	G1	260.46±3.25	3.73±0.37	7.72±0.33	34.83
2	20	G2	332.85±3.71	5.73±0.47	10.89±0.39	46.94
3	30	G3	337.67±3.64	5.59±0.45	9.03±0.35	44.51
4	40	G4	372.33±3.95	3.08±0.34	10.10±0.38	46.20
5	50	G5	361.29±3.90	9.38±0.60	7.52±0.33	47.95
Minimum			260.46±3.25	3.08±0.34	7.52±0.33	34.83
Maximum			372.33±3.95	9.38±0.60	10.89±0.39	47.95
Average			332.92±209	5.50±0.39	9.05±0.29	44.08
Worldwide average			400	32	30	370

But for ^{232}Th the minimum value was recorded in 50 cm and maximum value was recorded in depth at 20 cm, the ^{40}K values was minimum at depth 20 cm and the maximum was measured at depth 40 cm. as shown in Table (2).

Radium equivalent activity values for virgin soil samples are range from 18.51 to 47.95 Bq/Kg with an average 33.23 Bq/Kg. The clay soil have five values greater than the world wide average this increase in clay soil may be due to a high nutrient supplying capacity and very low internal drainage. One also can note, that minimum value of ^{238}U are measured at depth 50 cm in agricultural soil other than virgin soil which measured at depth 30 cm. this may be due to continuous fertilization in agricultural soils. The same as well as for ^{40}K. While for ^{232}Th radionuclide we find the maximum value are found in upper surface in both agricultural and virgin soils. This may be due to slowly react of thorium with water so it accumulate at upper surface of soil.

Clay soil

Table (3) shows the values of Absorbed Dose , internal Hazard index , external Hazard index and activity concentration index for clay soil .

For Absorbed dose values was varied from 23.08 to 40.69 nGy/h with on average 30.66 nGy/h , maximum values was recorded at 10cm depth while minimum values at 50cm depth .

Internal and external Hazard indices are ranged between 0.15 to 0.29 with average 0.12 and 0.12 to 0.22 with an average value 0.16 respectively while the activity concentration index was found to be in the range 0.18 to 0.32 with an arrange 0.24 .

Table (3) Absorbed Dose and Hazard indices for clay soil

No.	Sample cod	Depth (cm)	AD (nGy/h)	H_{in}	H_{ex}	I_{γ}
1	A1	10	27.18	0.20	0.15	0.21
2	A2	20	28.87	0.22	0.16	0.22
3	A3	30	24.98	0.17	0.13	0.20
4	A4	40	27.46	0.18	0.15	0.22
5	A5	50	23.08	0.15	0.12	0.18
6	B1	10	40.69	0.29	0.22	0.32
7	B2	20	32.42	0.21	0.18	0.26
8	B3	30	37.38	0.27	0.20	0.29
9	B4	40	32.10	0.23	0.18	0.25
10	B5	50	35.16	0.24	0.19	0.28
11	C1	10	29.62	0.20	0.16	0.23
12	C2	20	35.08	0.26	0.19	0.27
13	C3	30	30.87	0.21	0.17	0.24
14	C4	40	28.68	0.20	0.16	0.23
15	C5	50	26.46	0.18	0.14	0.21
	Minimum		23.08	0.15	0.12	0.18
	Maximum		40.69	0.29	0.22	0.32
	Average		30.66	0.21	0.16	0.24

Virgin Soil

Table (4) show the values of Absorbed dose internal Hazard index , external Hazard index and activity concentration index for virgin soil sample which are measured in this study .

The value of absorbed dose are ranged from 9.01 to 23.94 (nGy/h) with an average 16.47 (nGy/h) the internal Hazard index values are very from 0.06 to 0.15 with mean value 0.10 while external Hazard index values are found to be in the range between 0.05 to 0.13 with an average 0.13 , finally the activity concentration index are fluctuate between 0.07 to 0.19 with am average 0.13 .
The maximum and minimum values are measured at 50cm and 40cm respectively

Table (4) Absorbed Dose and Hazard indices for virgin soil

No.	Sample cod	Depth (cm)	AD (nGy/h)	H_{in}	H_{ex}	I_{γ}
1	G1	10	17.25	0.10	0.09	0.14
2	G2	20	23.11	0.14	0.13	0.18
3	G3	30	22.12	0.14	0.12	0.18
4	G4	40	23.05	0.13	0.12	0.18
5	G5	50	23.94	0.15	0.13	0.19
	minimum		9.01	0.06	0.05	0.07
	maximum		23.94	0.15	0.13	0.19
	Average		16.47	0.10	0.09	0.13

Conclusion

The obtained results confirm some conclusions as below:

1-The present work has shown that the specific activity of ^{238}U and ^{232}Th were within permissible limit in all soli types, while some values of ^{40}K were greater than worldwide average specially in clay.

2-All hazard indices are in permissible limits.

3-The clay soil has the maximum values of specific activity for ^{238}U, ^{232}Th and ^{40}K.

4-The virgin soli have the minimum values of specific activity for ^{238}U, ^{232}Th and ^{40}K.

5-The specific activity of ^{238}U decrease with depth in clay soil.

Recommendations

The obtained results confirm some conclusions as below:

1-We recommend reducing the usage of chemical fertilizers as far as possible in the farm land.

2-We recommend strongly to Cultivation of plants that can absorb heavy elements.

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