

Natural radioactivity in soil at regions around the uranium mine in Abu-Skhair Najaf Province, Iraq

**¹Hussain H. Al. Gazaly, ¹Mahdi A. Bahr al-Ulum, ¹Ali A. Al. Hamidawi
and ²Abdolzahra M. Al. Abbasi**

*¹Physics Department, Science College, Kufa University, Iraq
²Remote Sensing Center, Kufa University, Iraq*

ABSTRACT

The specific activity of ²³⁸U, ²³²Th and ⁴⁰K was determined in 60 soil samples were collected from sites around the uranium mine in the Abu - Skhair in Najaf province, Iraq. The samples were determined using NaI(Tl) gamma ray spectrometry system. Mean values for ²³⁸U, ²³²Th and ⁴⁰K specific activity were found to be (77.33±5.85) Bq/ kg, (9.36 ± 0.97) Bq/kg, and (426.31 ± 21.35) Bq/kg respectively. The study also examined radium equivalent R_{eq} associated with the natural radionuclides for 60 soil samples with an average of 123.54 ± 8.88 Bq/kg. A comparison of the measured values with the corresponding worldwide average values shows that the most specific activity of ²³⁸U and ⁴⁰K radionuclides in the studied samples are higher than world average activity values.

Key words: Abu-Skhair mine, Najaf province, Uranium and gamma ray spectroscopy.

INTRODUCTION

Radionuclides have been present always in every environment of the earth's surface. Only nuclides with half-lives comparable to the age of the earth or their corresponding decay products, existing in terrestrial materials, can still be found today on earth, e.g. ⁴⁰K, and the radionuclides from the ²³⁸U and ²³²Th series [1-3]. External exposure to radiation arises from natural and man-made radioactivity. Natural background radiation exposure is mainly caused by primordial radioactivity and cosmic radiation. Primordial radioactivity is mainly due to the presence of ²³⁸U, ²³²Th series radionuclides and ⁴⁰K in the earth's crust. Cosmic radiation comes through the earth's atmosphere, from the sun and galaxies. The primary cosmic radiation interacts with the atmospheric matter and produces Cosmo genic radionuclides also known as secondary cosmic radiation [4,5]. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend mainly on the geological and geographical conditions, and appear at different levels in the soils of each region in the world [6]. The other source of the radioactivity is from man-made radionuclides released into the atmosphere as a consequence of nuclear weapon testing, or nuclear power plant accidents. Many artificial radionuclides (¹³⁴Cs, ¹³⁷Cs, ¹³¹I, ⁹⁰Sr, etc) were released as a result of these events[7].

Area of study

The study focuses on an area located in the southwest of the Najaf province in Iraq. It's approximately 18 km from the city of Najaf and 2 km west of Al-Heera town within the district of Abu Skhair. The area of the study is called Hor Al-Jebssa, it is in between Al-Zejrey and Alsaneen villages. The area is located within the southern part of the sea of Najaf. The surface of the area is a semi plane of new depositional material which contains a mixture of soft sands and clays along with vegetal organic materials resulted from remnants of grass, plants and trees. This deposit has black color in some areas specifically in Hor Al-Jebssa. Such black deposits have been seen in other locations mixed with gypsum materials [8-10].

MATERIALS AND METHODS

Sample Preparation Techniques

60 soil samples were collected in an area of approximately 196250 m² from sites located around the uranium mine. We used a systematic circular sampling system involving 12 radial line, each line has a 30 degree difference from the previous line, and each of them consists 5 samples, the distance between sampling locations was 50 m, and each sample has a depth of 60 cm from the ground surface, as seen in Fig.1. In the laboratory, the soil samples were sieved by a 0.8mm mesh to remove larger objects and then ground using a mortar and pestle to fine powder in order to have the same matrix as the reference sample. After that, the samples were dried in an oven in order to reach constant weights. Then the samples were packed in 1 L polyethylene plastic Marinelli beakers of constant volume, so that there was geometric homogeneity around the detector. Then the respective net weights were measured and recorded with a high sensitive digital weighing balance with a percent of $\pm 0.01\%$. After that, the plastic Marinelli beakers were sealed with tape and stored for about one month before counting to allow secular equilibrium to be attained between ²²²Rn and its parent ²²⁶Ra in uranium chain[11].

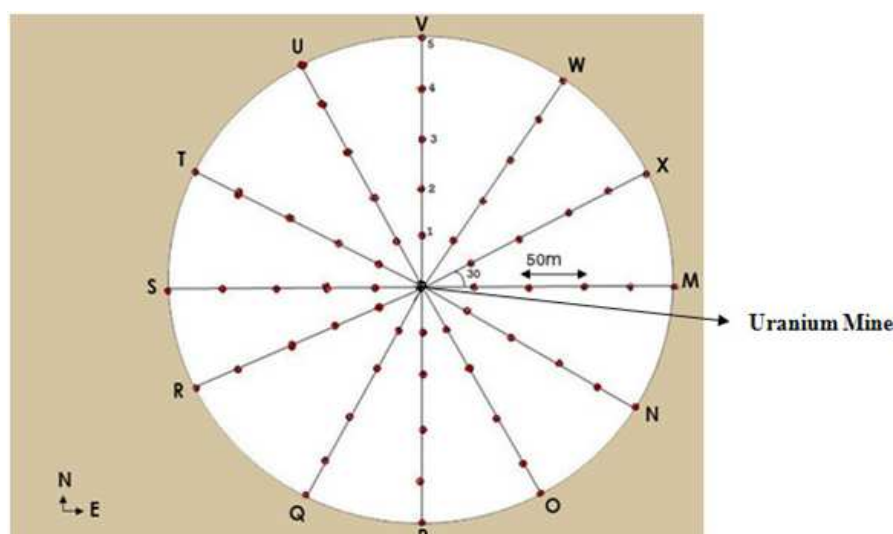


Fig. 1 : Schematic diagram of systematic circular sampling system

Analysis using Gamma Spectrometry

Radiation levels were measured using a gamma spectrometer which includes gamma multichannel analyzer equipped with NaI(Tl) detector of (3"×3") crystal dimension. The gamma spectra were analyzed using the ORTEC Maestro 32 data acquisition and analysis system. The detector had coaxial closed-facing geometry with the following specifications: The calculated resolution is 7.9% for energy of 661.66 keV of ¹³⁷Cs standard source. Relative efficiency at 1.33 MeV ⁶⁰Co was 2.2% and at 1.274 MeV ²²Na was 2.4%. The detector was shielded by a cylindrical lead shield in order to achieve the lowest back ground level. An energy calibration for this detector was performed with a set of standard γ -ray 1- μ Ci active ¹³⁷Cs, ⁶⁰Co, ⁵⁴Mn, and ²²Na sources. In this study, the activity concentration of ⁴⁰K was determined directly from the peak areas at 1460 keV. The activity concentrations of ²³⁸U and ²³²Th were calculated assuming secular equilibrium with their decay products. The gamma transition lines of ²¹⁴Pb (1765 keV) were used to calculate activity concentration of radioisotope in the ²³⁸U-series. The activity concentrations of radioisotope in the ²³²Th-series were determined using gamma transition lines of ²⁰⁸Tl (2614 keV). The counting time for each sample was at 18000 s. Specific Activities are calculated as follows[12,13]:

$$A = N / (t \times \epsilon \times I_{\gamma} \times m) \quad (1)$$

where N is the net peak area at energy E of radionuclide ²³⁸U, ²³²Th and ⁴⁰K respectively, ϵ is the detection efficiency, t is the counting time, I_{γ} is the gamma emission probability, and m is the mass in kg of the measured sample.

Specific Activity were given as Bq/kg of dry weight and the uncertainty values presented as \pm are the total uncertainties, and were calculated taking into account counting and efficiency calibration errors. The average specific activity of ²³⁸U, ²³²Th and ⁴⁰K is (33 , 45 and 412 Bq/Kg) respectively [14].

As the distribution of natural radionuclides in samples is not uniform, a common radiological hazard index has been introduced in order to compare the specific activities of materials containing different amounts of ²²⁶Ra, ²³²Th and

^{40}K . This index used to obtain the sum of those activities, is called Radium equivalent activity (Ra_{eq}) and is given by the relation below [15,16]:

$$\text{Ra}_{\text{eq}}(\text{Bq/Kg}) = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}} \quad (2)$$

where A_{Ra} , A_{Th} and A_{K} are respectively the specific activity of the radionuclides ^{226}Ra , ^{232}Th and ^{40}K respectively. The above formula is based on the assumption that 370 Bq/Kg of ^{226}Ra , 259 Bq/Kg of ^{232}Th and 4810 Bq/Kg of ^{40}K produce the same gamma dose rate. The maximum value of Ra_{eq} must be less than 370 Bq/Kg [14,15,17].

RESULTS AND DISCUSSION

The values have been found to lie in the range of $(19.28 \pm 3.91$ to $1118.16 \pm 23.13)$ Bq/kg with an average of 77.33 ± 5.85 Bq/kg, from $(3.03 \pm 0.73$ to $13.41 \pm 0.89)$ Bq/kg with an average value of 9.36 ± 0.97 Bq/kg and $(184.04 \pm 9.23$ to $655.25 \pm 34.99)$ Bq/kg with an average value of 426.31 ± 21.35 Bq/kg for ^{238}U , ^{232}Th and ^{40}K respectively. The calculated Ra_{eq} values for all samples shows that values oscillates between $(46.39 \pm 6.98$ to $1172.95 \pm 26.87)$ Bq/kg with an average of 123.54 ± 8.88 Bq/kg. The values are presented in table [1]:

Table 1 : Specific activity of ^{238}U , ^{232}Th , ^{40}K and Ra_{eq} of soil samples

Sample Code	Specific Activity (Bq/kg)			Ra_{eq} (Bq/kg)
	A_{U}	A_{Th}	A_{K}	
M1	64.04±3.76	13.41±0.89	460.86±22.98	118.7±6.80
M2	41.53±4.19	12.7±0.86	473.58±22.82	96.16±7.18
M3	28.97±4.6	5.21±0.94	425.83±19.16	69.21±7.42
M4	56.26±5.64	12.93±0.93	452.25±23.12	109.57±8.75
M5	33.42±6.05	8.3±1.03	419.49±20.96	77.59±9.14
N1	265.45±8.43	9.45±1.09	501.4±23.1	317.57±11.77
N2	19.28±3.91	6.46±0.9	271.56±12.86	49.43±6.19
N3	42.92±4.23	12.59±0.99	470.07±23.84	97.12±7.48
N4	38.1±4.87	9.88±0.94	451.85±20.96	87.02±7.83
N5	52.14±3.59	10.81±0.84	464.01±25.12	103.33±6.73
O1	541.95±13.81	8.07±1.55	565.57±30.02	597.04±18.34
O2	37.66±5.74	12.28±0.95	438.93±23.4	89.02±8.90
O3	33.36±4.69	5.94±0.76	215.96±11.4	58.49±6.65
O4	47.4±4.31	10.13±0.69	424.23±18.22	94.55±6.7
O5	48.3±4.48	9.79±0.8	507.47±25.08	101.38±7.56
P1	50.98±6.36	6.68±1.05	328.36±16.05	85.82±9.1
P2	36.89±5.37	12.04±0.95	461.76±22.86	89.66±8.49
P3	30.07±4.68	7.85±1.17	337.16±17.42	67.26±7.69
P4	46.9±5.45	8.81±1.07	469.51±24.83	95.65±8.89
P5	33.79±5.49	10.84±1.23	379.98±20.57	78.55±8.83
Q1	111.27±8.33	10.32±1.05	517.24±26.68	165.86±11.89
Q2	39.47±6.41	11.05±1.23	488.04±24.38	92.85±10.05
Q3	42.76±5.01	5.98±1.23	437.05±21.6	84.96±8.43
Q4	38±5.4	10.04±1.1	443.8±23.42	86.53±8.78
Q5	38.46±5.79	7.79±1.11	341.92±18.08	75.93±8.77
R1	127.48±8.17	8.33±1.06	479.19±24.81	176.29±11.6
R2	44.48±5.98	12.52±0.92	431.31±22.29	95.6±9.01
R3	42.17±5.24	8.04±0.86	223.49±11.46	70.88±7.35
R4	27.43±4.55	7.91±0.9	441.29±20.36	72.72±7.41
R5	40.65±4.89	10.43±0.83	433.4±22.1	88.94±7.78
S1	50.96±6.44	8.38±1.31	457.77±24.54	98.19±10.20
S2	56.52±6.34	8.56±1.08	463.01±23.29	104.41±9.68
S3	38.34±6.4	9.95±1.21	471.8±23.7	88.9±9.96
S4	59.31±4.91	14.29±0.76	425.06±21.96	112.47±7.69
S5	35.4±5.38	9.25±0.92	411.68±21.27	80.33±8.33
T1	36.32±5.2	9.34±1.02	427.87±22.54	82.62±8.39
T2	40.98±5.78	10.66±0.79	491.86±24.76	94.1±8.82
T3	35.77±5.5	7.1±1.01	424.32±21.84	78.6±8.63
T4	45.95±5.82	11.33±0.79	394.23±19.52	92.51±8.45
T5	47.11±5.14	8.26±0.84	363.48±18.43	86.91±7.76
U1	125.13±7.06	8.32±1.09	457.48±22.53	172.25±10.35
U2	49.93±4.06	8.52±0.93	333.49±16.97	87.79±6.7
U3	42.42±2.29	7.4±1.13	463.94±22.4	88.73±5.63
U4	191.48±10.44	5.89±1.27	551.34±28.65	242.36±14.46
U5	1118.16±23.13	3.03±0.73	655.25±34.99	1172.95±26.87
V1	77.15±5.66	10.74±0.91	425.1±20.78	125.24±8.56
V2	34.07±5.02	8.71±0.54	248.36±11.48	65.65±6.68

V3	38.6 ± 5.4	8.57 ± 1.01	442.37 ± 21.66	84.92 ± 8.51
V4	24.42 ± 4.71	12.35 ± 0.78	468.03 ± 22.56	78.12 ± 7.56
V5	29.54 ± 4.95	10.02 ± 0.84	444.33 ± 21.88	78.08 ± 7.84
W1	57.4 ± 5.5	9.01 ± 0.91	386.12 ± 18.81	100.02 ± 8.25
W2	44.24 ± 4.37	11.78 ± 0.68	389.68 ± 19.19	91.09 ± 6.82
W3	27.77 ± 4.14	6.03 ± 1.19	366.44 ± 16.91	64.61 ± 7.14
W4	27.74 ± 4.71	11.23 ± 0.77	454.49 ± 21.91	78.8 ± 7.5
W5	21.45 ± 5.08	7.53 ± 0.83	184.04 ± 9.23	46.39 ± 6.98
X1	36.82 ± 6.08	6.8 ± 1.12	441.4 ± 23.21	80.53 ± 9.47
X2	27.78 ± 4.85	10.54 ± 1.11	440.33 ± 20.89	76.76 ± 8.05
X3	38.61 ± 5.22	11.35 ± 0.91	444.93 ± 22.15	89.1 ± 8.23
X4	40.16 ± 6.45	12.7 ± 0.93	446.69 ± 21.72	92.72 ± 9.45
X5	38.43 ± 5.31	7.29 ± 1.13	447.28 ± 21.18	83.3 ± 8.56
Min.	19.28 ± 3.91	3.03 ± 0.73	184.04 ± 9.23	46.39 ± 6.98
Max.	1118.16 ± 23.13	14.29 ± 0.76	655.25 ± 34.99	1172.95 ± 26.87
Mean	77.33 ± 5.85	9.36 ± 0.97	426.31 ± 21.35	123.54 ± 8.88

It was found that all values of ^{238}U specific activities were higher than the worldwide average with the exception of ten samples (M3, N2, P3, R4, V4, V5, W3, W4, W5, X2), where the maximum specific activity value was calculated in U5 sample and the minimum value is in N2, as shown in Fig.2 . The results for all values of ^{232}Th specific activities are less than the worldwide average (45 Bq/kg). For ^{40}K , it is clear that the specific activities, with the exception of 16 samples, are found to be higher than the worldwide average, where the maximum specific activity value is calculated in U5 sample and the minimum value is in W5, as seen in Fig.(2) . It is observed that the values of Ra_{eq} in 58 samples were lower than the acceptable safe limit of 370Bq/kg , as shown in Table (1) and Fig.2 indicate that there are two values (O1, U5) greater than the worldwide average. As a rule, the matter whose Ra_{eq} exceeds 370 Bq/kg is discouraged[14,17].

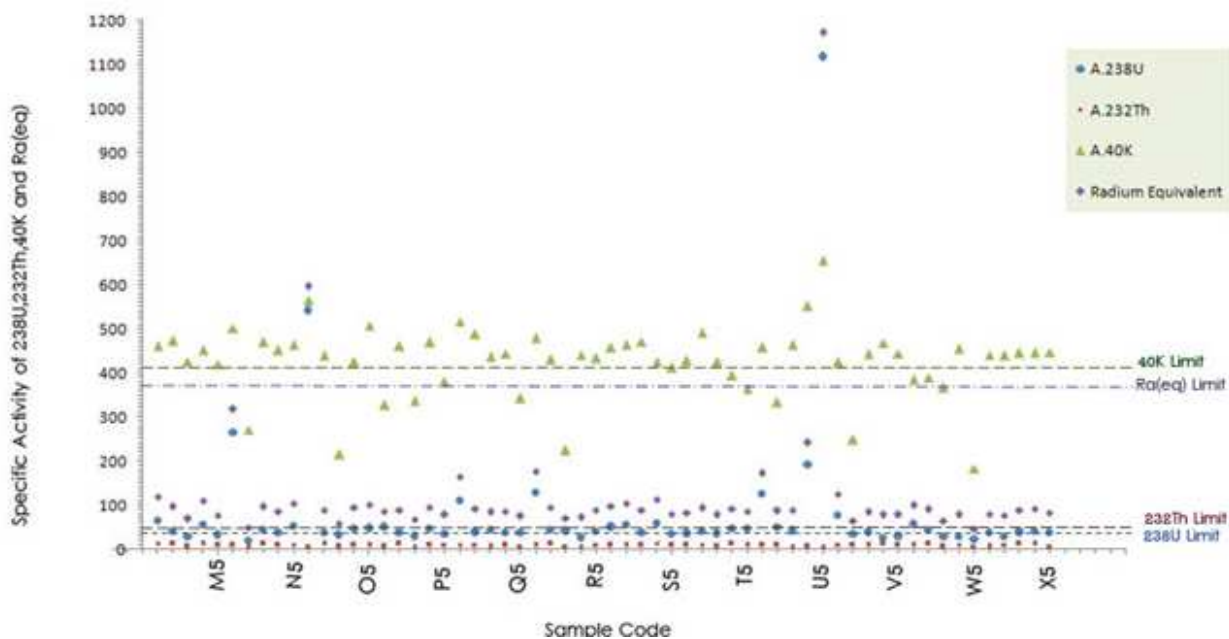


Fig.2 : Specific activity of ^{238}U , ^{232}Th , ^{40}K and Ra_{eq}

We conclude that the soil layer around the uranium mine has inhomogeneous uranium distribution and significant variation in various locations and average uranium activities greater than the worldwide average. This inhomogeneous uranium distribution can be mainly due to geological reasons such as the presence radioactive of anomalies related to the primary uranium that exist within the upper parts of the Euphrates Formation (Middle Miocene) in this location and also changes in groundwater velocities due to the influence of faults which led to enrichment of uranium in the water as a result of transportation from some localities and deposition in others. There are physical reasons as well, such as the human activities of drilling and exploration operations of the mine and the transfer of uranium extraction which led to increased pollution in this region and contributing to raise the level of radioactivity in the location around the mine and finally agriculture and plowing operations taking place in the territory of that region and irrigation through ground water containing high concentrations of uranium cause redistribution and spread the uranium on the soil surface. The thorium activities were within normal level in the studied area. The potassium radionuclide in soil samples with the exception of 16 samples were higher than the

range of the worldwide average, we can explain the high values of potassium by the fact that the land surrounding the uranium mine is an agricultural area and using fertilizer could increase the concentration of potassium. The results of our work that we found in the sites located around the uranium mine at Abu – Skhair district of Najaf province are comparable with other studies that determined the natural radiation levels (^{238}U , ^{232}Th , ^{40}K) in soil of other locations in the Najaf province, we found that our location have natural radiation levels higher than other regions also it has the highest levels of the worldwide average levels, thus we can identify it as a high background radiation area.

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