Measurement Natural Radioactivity in Soil Samples from Important historical locals in Alnajaf Alashraf city, Iraq.

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ABSTRACT

Al-Najaf Al-Ashraf city one of the most important, oldest, historical and religious cities in Iraqi country which includes hundreds of holy shrines and historical mosques which over the built hundreds of years. The natural radiation of forty two samples of soil which collected randomly in June 2013 from different religious and historical places were measured using 3''×3'' Na(Tl) detection. The mean values activity concentrations of 238U, 232Th and 40K was (23.59±4.37, 12.10±0.54 and 60.68±2.30) Bq kg⁻¹ respectively, specific activity for all soil sample were in the worldwide average. The average values of the Radium equivalent activity and annual effective dose were (22.455 Bq/kg and 25.375 μSv/y) less than the world average. The heist external and internal hazard and gamma activity concentration index were (0.274, 0.412 and 0.705) lower than unity.

Keywords

Gamma ray spectrometry; Na(Tl) detector; Raeq activities and annual effective dose.
INTRODUCTION

Al Najaf city has been enjoyed in religious and cultural prestige, they are a natural extension of Kufa capital of the Islamic caliphate days of Imam Ali bin Abi Talib (peace be upon him), in al Najaf city many shrines and most important mosque.

Shrine of Imam Ali -also known as Alhaidariya holy - in the center of Najaf it refers as the advantage Islamic architectural style construction, and is considered one of the most important mosques and religious shrines in Iraq because it includes the remains of the Imam Ali bin Abi Talib who was martyred in 40 AH (661 AD).

Shrine of the Prophets Hood and Saleh is located in the Najaf cemetery major behind the city wall the old hand and the Valley of Peace shrine of prophets Hood and Saleh on the campus of one meant visitor to prayer and blessing, also Camille ibn Ziyad Nakha’i Shrine who realized the life of the Messenger of Allah 18 years old, and Imam Ali as well as the shrine of Muslim Ibn Aqil “al Hussein ambassador” and al Sahaabi Maltam al Tamar shrine and the house of Imam Ali which known as Bait Fatima and al sahlaa mosque and many other religious and historical places.

In this research the natural radioactivity level of elected soils of these shrines, religious places and historical holy has been studied because of religious and historical importance in addition to the large number of visitors It is important to identify the concentration of radioactive isotopes in soil because it constitutes a path for radioactivity to humans and is an indicator of radioactive accumulation in the environment [1,2].

Natural sources of radiation are cosmic radiation and terrestrial radiation arising from the decay of naturally occurring radioactive substance. If the half life of a radionuclide found in geological strata approximates the estimated age of the earth, then the radionuclide is primordial. It was presumably present from the time of the earth’s beginning. Inventories of primordial radionuclides are essential parts of the natural background level of radioactivity in the environment. The concentrations of the natural radio nuclides $^{238}$U, $^{232}$Th, their daughter products and $^{40}$K present in the soil and rocks which in turn depend upon the local geology of each region in the world are causes of variation of doses. Some areas are high natural background areas because in these areas levels of uranium and its decay products in rock and soil. High background radiation areas are due to local geology, location altitude and geochemical effects that cause enhanced levels of terrestrial radiation [3-9].

EXPERIMENTAL PROCEDURES:

The soil samples measured at (0-5) cm depth level were collected from sampling points located at religious and historical places. The location of the samples is shown on table (1) and figure (1) after collection. Samples are crushed into fine powder by grinder. Fine quality of the sample is obtained using fine sieve. Before measurement samples are dried in an oven at a temperature of 60 C° for 72 h, each sample is packed and sealed in an airtight PVC container and kept for about (4) weeks period to allow radioactive equilibrium among the daughters products. An average (1) kg of soil is used per sample.

To measured the specific activity we used NaI(Tl) a system which consist of a scintillation detector NaI(Tl) of (3’*3’) crystal dimension, supplied by (Alpha Spectra,Inc), coupled with a multi-channel analyzer (MCA) (ORTEC – Digi Base) with range of 4096 channel joined with ADC (Analog to Digital Convertor) unit, through interface. The spectral data was converted directly to the PC of the laboratory introduced by using (MAESTRO-32) software. The detector was enclosed in a graded lead shield. The gamma spectra of the collected samples were measured and the activities of $^{238}$U, $^{232}$Th series, $^{232}$Th series and $^{40}$K in each sample were determined by measuring the characteristic gamma-peaks of their daughters. The line at 1764 keV of $^{214}$Bi was used to determine $^{238}$U series activity, and the line at 2614 keV of $^{228}$Th for $^{232}$Th series. Also the peak at 1460 keV was used for $^{40}$K activity [10,11].

The specific activity of each radionuclide is calculated using the following equation

$$A = \frac{N_{net}}{\epsilon . I_{\gamma} . m . t} \pm \sqrt{\frac{N_{net}}{\epsilon . I_{\gamma} . m . t}} [Bq.kg^{-1}]$$

... ... ... (1)

Where $N_{net}$ is the net count (area under the specified energy peak after back ground subtraction ) in (c/s), $\epsilon$ is the efficiency of the detector, $I_{\gamma}$ is the transition probability of the emitted gamma ray, $t$ is the time (in sec) for spectrum collected and $m$ is the sample weight (in kg).

Radium equivalent activity ($Ra_{eq}$):

Distribution of $^{238}$U, $^{232}$Th and $^{40}$K in environment is not uniform, so that with respect to exposure to radiation, the radioactivity has been defined in terms of radium equivalent activity ($Ra_{eq}$) in Bq.kg-1 [12-14].

$$Ra_{eq} = A_{U} + 1.43A_{Th} + 0.077A_{K} \ldots (2)$$

Where $A_{U}$, $A_{Th}$ and $A_{K}$ are specific activity concentration in Bq.kg-1 of $^{238}$U, $^{232}$Th and $^{40}$K, respectively. The index is useful to compare the specific activity of materials containing different concentrations of $^{238}$U, $^{232}$Th and $^{40}$K.
Gamma Dose Rate (D)

The total dose rate $D$ in the air (out doors) due to uniform distribution of all the $^{226}$Ra, $^{232}$Th and $^{40}$K in the beach soil 1 m above the ground surface was estimated by [12-14]:

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

Where $D$ is the dose rate in (nGy.h$^{-1}$) and $A_U$, $A_{Th}$ and $A_K$ are the concentrations of uranium, thorium and potassium, respectively.

Annual Effective Dose Equivalent (AEDE)

In order to estimate the annual effective dose rate in air the conversion coefficient from absorbed dose in air to effective dose received by an adult had to be taken into consideration. This value is published in UNSCEAR (2000)[5] of (0.7 Sv/Gy). The outdoor occupancy factor which is about (0.2).

The annual effective dose equivalent was given by the following equation [12-16]:

$$AEDE (\mu Sv/\text{y}) = D(nGy/h) \times 8760(\text{h/y}) \times 0.2 \times 0.7(Sv/Gy) \times 10^{-3} \ ... \ (4)$$

Representative level index ($I_{\gamma_r}$)

In order to examine whether the sample meets limits of dose criteria, another radiation hazard index, representative level index $I_{\gamma_r}$, used to estimate the level of $\gamma$- radiation hazard associated with the radionuclides in specific investigated samples, is defined as the following equation [12-15]:

$$I_{\gamma_r} = A_U / 300 + A_{Th} / 200 + A_K / 3000 \ ... \ (5)$$

The index $I_{\gamma_r}$ was correlated with the annual dose due to the excess external gamma radiation caused by superficial material. Values of index $I \leq 1$ correspond to 0.3 mSv/y, while $I \leq 3$ correspond to 1 mSv/y. Thus, the activity concentration index should be used only as a screening tool for identifying materials which might be of concern to be used as covering material. According to this dose criterion, materials with $I \leq 3$ should be avoided [15].

External hazard index ($H_{ex}$)

The external hazard index ($H_{ex}$) was given by the following equation [12-15]

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \ ... \ (6)$$

Internal hazard index ($H_{in}$)

The internal exposure to $^{222}$Rn and its radioactive progeny is controlled by the internal hazard index ($H_{in}$) is given by [13, 14]

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \ ... \ (7)$$

For the safe use of a material in the construction of dwellings, index ($H_{in}$) should be less than unity and the maximum value of ($H_{in}$) to be less than unity.

RESULTS AND DISCUSSIONS:

The specific activity values of $^{238}$U, $^{232}$Th and $^{40}$K radionuclides for 42 soil sample are tabulated in table (1). They have been found to lie in the range of (5.89±2.23; S3 to 36.20±5.52; S29) Bq/kg with an average of 23.59±4.37 Bq/kg, from (0.72±0.16; S28 to 31.68±1.04; S16) Bq/kg with an average 12.10±0.54 Bq/kg and (20.89±1.38; S6 to 90.63±2.87; S18) Bq/kg with an average 60.68±2.30 Bq/kg for $^{238}$U, $^{232}$Th and $^{40}$K respectively. The result shows that all values of $^{238}$U, $^{232}$Th and $^{40}$K specific activity for all soil sample are in the worldwide average (35Bq/kg for $^{238}$U, 30 Bq/kg for $^{232}$Th and 400 Bq/kg for $^{40}$K) [17, 18].
| Sample No. | Samples Name                                           | \(^{238}\)U | \(^{232}\)Th | \(^{40}\)K |
|------------|-------------------------------------------------------|-------------|-------------|----------|
| S1         | the Imam Ali holy shrine (1)                          | 11.82±2.5205| 14.74±0.5661| 41.73±1.5554|
| S2         | the Imam Ali holy shrine (2)                          | 25.23±3.8935| 11.06±0.5185| 61.83±2.0017|
| S3         | the Imam Ali holy shrine (3)                          | 5.89±2.2274 | 19.31±0.8110| 74.74±2.6052|
| S4         | the Imam Ali holy shrine (4)                          | 26.94±4.7623| 16.66±0.7532| 41.41±1.9392|
| S5         | Maqam Hood and Saleh                                  | 21.89±4.2927| 2.15±0.2703 | 82.55±2.7379|
| S6         | Maqam Imam al hoja                                    | 27.78±4.8362| 26.87±0.9567| 20.89±1.3772|
| S7         | Beer alawee Mosque                                    | 19.36±4.0375| 8.69±0.5439 | 79.01±2.6786|
| S8         | Maqam Ibrahim al gamr                                | 35.36±5.4559| 23.43±0.8944| 27.70±1.4359|
| S9         | Fatima shrine (1)                                     | 17.68±3.8579| 29.50±1.0023| 30.88±1.6745|
| S10        | Fatima shrine (2)                                     | 19.36±4.0375| 20.03±0.8259| 60.84±2.3506|
| S11        | Fatima shrine (3)                                     | 21.89±4.2927| 22.27±0.8710| 88.09±2.8283|
| S12        | Fatima shrine (4)                                     | 17.68±3.8579| 26.57±0.9512| 80.82±2.7092|
| S13        | SAFEE alsafa shrine (1)                               | 12.63±3.2605| 22.14±0.8683| 72.65±2.5685 |
| S14        | SAFEE alsafa shrine (2)                               | 26.94±4.7623| 7.83±0.5165 | 48.13±2.0906|
| S15        | SAFEE alsafa shrine (3)                               | 24.41±4.5336| 25.75±0.9365| 57.21±2.2794|
| S16        | Maqam roqea bint al Hassan                            | 16.84±3.7650| 31.68±1.0387| 22.70±1.4359|
| S17        | Camille ibn Ziyad shrine (1)                          | 15.15±3.5718| 22.99±0.8846| 42.68±1.9688|
| S18        | Camille ibn Ziyad shrine (2)                          | 30.31±5.0512| 30.52±1.0195| 90.63±2.8688|
| S19        | Camille ibn Ziyad shrine (3)                          | 17.68±3.8579| 22.24±0.8704| 86.58±2.4953|
| S20        | Hussienieh Al rasol ala’adm-Mashkhab                   | 21.05±4.2094| 23.47±0.8940| 38.32±1.8655|
| S21        | Hussienieh Mashkhab-Mashkhab                          | 16.00±3.6696| 30.31±1.0161| 65.57±2.4401|
| S22        | mosque Al rasol ala’adm-Mashkhab                      | 15.15±3.5718| 15.33±0.7225| 77.19±2.6476|
| S23        | Sons of Imam Kadhim shrine                            | 30.31±5.0512| 11.38±0.6225| 86.82±2.8078|
| S24        | Nabi Younis shrine                                    | 15.15±3.5718| 22.14±0.8683| 69.02±2.5035|
| S25        | Maitham Younis shrine                                 | 27.78±4.8362| 0.78±0.1633 | 59.84±2.3312|
| S26        | Faithful home (1)                                     | 31.15±5.1209| 1.09±0.1927 | 41.59±1.9435|
| S27        | Faithful home (2)                                     | 28.62±4.9089| 1.46±0.2233 | 69.47±2.5117|
| S28        | Faithful home (3)                                     | 20.20±4.1243| 0.72±0.1561 | 89.27±2.8472|
| S29        | Faithful home (4)                                     | 36.20±5.5205| 1.02±0.1866 | 86.00±2.7946|
| S30        | Alsahlaa mosque (1)                                   | 31.15±5.1209| 1.12±0.1957 | 54.31±2.2207|
| S31        | Alsahlaa mosque (2)                                   | 31.15±5.1209| 1.12±0.1957 | 54.31±2.2207|
| S32        | Alsahlaa mosque (3)                                   | 31.15±5.1209| 1.12±0.1957 | 54.31±2.2207|
| S33        | Alsahlaa mosque (4)                                   | 22.73±4.3745| 0.82±0.1669 | 54.49±2.2244|
| S34        | Muslim Ibn Aqil shrine (1)                            | 28.62±4.9089| 0.89±0.1737 | 63.48±2.4    |
| S35        | Muslim Ibn Aqil shrine (2)                            | 29.47±4.9806| 1.46±0.2233 | 84.73±2.7738|
The radium equivalent activities was calculated and listed in table (2). Ra\textsubscript{eq} values vary from (5.143;S40 to 50.872;S18) Bq/kg with average value of (22.455) Bq/kg. It can be seen that the Ra\textsubscript{eq} values for all samples are lower than the recommended value 370 Bq/kg\[17,18].

Gamma Dose Rate (D), Annual Effective Dose Equivalent (AEDE), Representative level index (I\textsubscript{γr}), External hazard index (H\textsubscript{ex}) and Internal hazard index (H\textsubscript{in}) are calculated and listed in table (2) the Gamma Dose Rate (D) range from (10.590;S41 to 37.041;S18) nGy/h with average 20.691 nGy/h, the (AEDE) range are from (12.988;S41 to 45.427;S18) (μSv/y) with average 25.375(μSv/y) all the soil samples have the annual effective dose less than the world average 460 (μSv/y)\[17,18], Representative level index (I\textsubscript{γr}) range from (0.073;S40 to 0.705;S18) with average 0.311 , External hazard index (H\textsubscript{ex}) range from (0.027;S40 to 0.274;S18) with average 0.120 and Internal hazard index (H\textsubscript{in}) range from (0.041;S40 to 0.412;S18) with average 0.181. External and internal hazard and gamma activity concentration were lower than unity according to the Radiation Protection 112 [17].

Table (2): Radium equivalent (Bq.kg\textsuperscript{-1}), Dose rate (nGy/h), AEDE (μSv/y), the internal and external hazard indexes representative level index for all samples.

| Sample No. | Ra\textsubscript{eq} | D(nGy/h) | AEDE (μSv/y) | H\textsubscript{in}≤1 | H\textsubscript{ex}≤1 | I\textsubscript{γr}≤1 |
|------------|-----------------|----------|---------------|-------------------|-------------------|-------------------|
| S1         | 24.654          | 16.600   | 20.359        | 0.199             | 0.132             | 0.340             |
| S2         | 20.915          | 20.754   | 25.453        | 0.169             | 0.112             | 0.291             |
| S3         | 33.969          | 18.514   | 22.706        | 0.274             | 0.182             | 0.469             |
| S4         | 27.614          | 24.310   | 29.813        | 0.222             | 0.148             | 0.378             |
| S5         | 9.759           | 14.316   | 17.558        | 0.078             | 0.052             | 0.142             |
| S6         | 40.665          | 30.551   | 37.467        | 0.328             | 0.218             | 0.554             |
| S7         | 18.858          | 17.415   | 21.358        | 0.152             | 0.101             | 0.265             |
| S8         | 35.551          | 31.587   | 38.738        | 0.287             | 0.191             | 0.486             |
| S9         | 44.769          | 28.403   | 34.833        | 0.362             | 0.241             | 0.614             |
| S10        | 34.114          | 24.142   | 29.608        | 0.274             | 0.182             | 0.468             |
| S11        | 39.427          | 27.880   | 34.192        | 0.317             | 0.211             | 0.544             |
| S12        | 44.781          | 28.611   | 35.089        | 0.361             | 0.240             | 0.618             |
| S13        | 37.415          | 23.172   | 28.418        | 0.303             | 0.202             | 0.519             |
| S14        | 15.699          | 18.759   | 23.006        | 0.125             | 0.083             | 0.215             |
| S15        | 42.007          | 29.931   | 36.707        | 0.338             | 0.225             | 0.576             |
| S16        | 47.581          | 29.135   | 35.731        | 0.384             | 0.256             | 0.649             |
| S17        | 36.385          | 23.525   | 28.852        | 0.294             | 0.196             | 0.501             |
| S18        | 50.872          | 37.041   | 45.427        | 0.412             | 0.274             | 0.705             |
|      |      |      |      |      |      |      |
|------|------|------|------|------|------|------|
| S19  | 37.287 | 25.221 | 30.931 | 0.302 | 0.201 | 0.517 |
| S20  | 37.076 | 26.170 | 32.095 | 0.299 | 0.199 | 0.507 |
| S21  | 48.710 | 29.717 | 36.444 | 0.394 | 0.262 | 0.672 |
| S22  | 28.358 | 19.936 | 24.450 | 0.229 | 0.152 | 0.394 |
| S23  | 23.561 | 24.205 | 29.685 | 0.189 | 0.126 | 0.329 |
| S24  | 37.480 | 24.094 | 29.549 | 0.302 | 0.201 | 0.517 |
| S25  | 6.316  | 14.955 | 18.340 | 0.050 | 0.033 | 0.090 |
| S26  | 5.339  | 15.811 | 19.390 | 0.042 | 0.027 | 0.074 |
| S27  | 7.920  | 16.179 | 19.842 | 0.063 | 0.042 | 0.114 |
| S28  | 8.343  | 12.940 | 15.869 | 0.066 | 0.044 | 0.122 |
| S29  | 8.762  | 19.832 | 24.322 | 0.069 | 0.046 | 0.126 |
| S30  | 6.164  | 16.380 | 20.088 | 0.049 | 0.032 | 0.089 |
| S31  | 5.638  | 16.491 | 20.225 | 0.045 | 0.029 | 0.081 |
| S32  | 6.190  | 11.340 | 13.908 | 0.049 | 0.032 | 0.089 |
| S33  | 5.871  | 12.590 | 15.440 | 0.046 | 0.030 | 0.084 |
| S34  | 6.722  | 15.538 | 19.056 | 0.053 | 0.035 | 0.096 |
| S35  | 9.176  | 17.195 | 21.087 | 0.073 | 0.048 | 0.132 |
| S36  | 6.387  | 14.889 | 18.259 | 0.050 | 0.033 | 0.089 |
| S37  | 6.975  | 17.669 | 21.670 | 0.055 | 0.036 | 0.097 |
| S38  | 8.515  | 16.576 | 20.328 | 0.068 | 0.045 | 0.124 |
| S39  | 7.130  | 12.623 | 15.480 | 0.057 | 0.038 | 0.104 |
| S40  | 5.143  | 15.805 | 19.383 | 0.041 | 0.027 | 0.073 |
| S41  | 6.866  | 10.590 | 12.988 | 0.054 | 0.036 | 0.098 |
| S42  | 8.127  | 17.614 | 21.602 | 0.064 | 0.042 | 0.115 |
| Average | 22.455 | 20.691 | 25.375 | 0.181 | 0.120 | 0.311 |
| Max. | 50.872 | 37.041 | 45.427 | 0.412 | 0.274 | 0.705 |
| Min. | 5.143  | 10.590 | 12.988 | 0.041 | 0.027 | 0.073 |

**Figure 1:** Map of Al-Najaf city
CONCLUSIONS:

The activity concentrations of $^{238}$U, $^{232}$Th and $^{40}$K for (42) soil samples from different religious and historical places was determined. The activity concentrations were measured using Na(Tl) detection. The mean values activity concentrations of $^{238}$U, $^{232}$Th and $^{40}$K was (23.59±4.37, 12.10±0.54 and 60.68±2.30) Bq kg$^{-1}$ respectively. The values of the Radium equivalent activity and annual effective dose was less than the world average. External and internal hazard and gamma activity concentration (representative level index) indexes were lower than unity.

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