Natural Radioactivity in Selected Soil Samples from the Archaeological of Ur City in Dhi-Qar Province, Iraq

Iman Tarik Al-Alawy^{1*} and Monar Deya Salim²

¹AL-Mustansiriyah University, College of Science, Department of physics, Baghdad, Iraq

²Dhi -Qar University, College of Science, Department of physics, Dhi -Qar, Iraq

*Email: drimantarik@yahoo.com

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ABSTRACT. The specific activity of natural radionuclides in 24 soil samples collected from antiquities area of Ur city in Dhi-Qar province (31.0459863N, 46.2534257E) in southern Iraq have been studied and evaluated. Experimental results were obtained by using a Gamma ray spectrometer analysis system consists of a scintillation detector Sodium Iodide activated by Thallium NaI(Tl) of (3"×3") crystal dimension at the laboratory of radiation detection and measurement in Science Collage, University of Kufa. The spectrometer has been calibrated for energy by acquiring a spectrum from four standard sources of gamma radiations supplied by spectrum techniques (LLC). The measuring time of all soil samples is 18000 seconds; it was found that, the soil specific activity ranges from 29.93 ± 2.97 to 9.99 ± 2.56 Bq/kg for ²³⁸U, from 25.66 ± 2.55 to 7.77 ± 2.24 Bq/kg for ²³²Th and from 397.38 ± 12.06 to 215.75 ± 8.91 Bq/kg for ⁴⁰K, with mean values of 17.9 ± 3.02 Bq/kg, 13.66 ± 2.41 Bq/kg and 314.62 ± 11.3 Bq/kg, respectively. The results have been compared with the acceptable data of the worldwide literatures. In order to evaluate the radiological hazard of the natural radioactivity, the radium equivalent activity (R_{eq}), the gamma absorbed dose rate (AD), the annual effective dose rate and the both (external and internal) hazard index have been calculated and compared with the acceptable values of the worldwide average (UNSCEAR 2000).

1. INTRODUCTION

People are exposed to ionizing radiation from the radionuclides that are present in different types of natural sources. Human exposure to ionizing radiation is one of the scientific subjects that attract most public attention [1]. Science natural radiation is responsible for most of the total radiation exposure of the human population, knowledge of the dose received from natural radioactivity, is very important in the discussion of effect on health [2]. Natural Occurring Radionuclides Materials (NORM) is known to be present in rocks and soil [3]. The natural radionuclides of concern are mainly of ²³⁸U, ²³²Th or its progenies and ⁴⁰K [4, 5]. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world [6, 7, 8, 9]. The present Sumerian Ur city, which occupied a prominent center in the history of humanity and had a role in stabilizing the concepts of civil result of the interaction between nature and man have been mentioned in the Old Testament where some believe that there is a relationship between Ur city and Abraham (AS). The Ur city, at a distance of 17Km to the southern west of Nasiriyah city, located at 380Km to the southern east of Baghdad, and 200Km to the north of Basrah and the Arabian Gulf. It is located 6Km to the east of the line of traffic highway between Kuwait-Iraq-Jordan. Because the Ur city settlement since (4000BC). It is believed that the people of this role are the Sumerians first to put the principles of irrigation, developed agriculture and used metals, especially copper. The second round, which passed by Ur city is the role of frozen victory in (2900BC). This role is called the pre-strains role or the role of the flood, the achievements of this role, the emergence of writing, and the sculpture on the seals. And the role the other, which passed by Ur city is the role of age dawn of dynasties (2800-2400BC) is considered the golden age, reaching Ur city the height of its greatness in all areas in terms of

architecture, where they found architectural elements such as the bow and trimmings, it appears evident in the Temple (Bear-Lal-Mach) and the Royal Tomb of King Sholeki.

2. MATERIALS AND METHODS

In this study, we choose to investigate the level of specific activities of ^{238}U , ^{232}Th and ^{40}K in 24 soil samples collected from 8 places in the archaeological city of Ur, as shown in fig. 1 [10]. Soil samples were collected from 8 locations in the antiquities area of Ur city in Dhi-Qar governorate in the south of Iraq. Sampling locations are marked in fig. 1.

Fig. 1 The sampling locations of 24 soil samples in the antiquities area of Ur city in Dhi-Qar governorate in the south of Iraq [10]/

The samples were ground into a fine powder with a particle size less than $100\mu\text{m}$ and then dried in a temperature-controlled furnace at 110°C for 24h to remove moisture. Each sample stored in a sealed polyethylene marinelli beaker for 30days to achieve the secular equilibrium. Marinelli beaker was used as sampling and measuring container. Before use the containers were washed with hydrochloric acid and rinsed with distilled water [11]. The activities of natural radionuclides ^{238}U , ^{232}Th and ^{40}K in the samples was determined by gamma rays spectrometry NaI(Tl) of (3"×3") crystal dimensions, supplied by (Alpha Spectra, Inc.-12I12/3), coupled with a multi-channel analyzer (MCA) (ORTEC-Digit Base) with range of 4096 channel joined with analog to digital converter (ADC) unit, through interface. The spectroscopic measurements and analysis are performed via the (MAESTRO-32) software into the PC of the laboratory. The specific activity concentration of ^{238}U was determined using the gamma-lines 1765keV of ^{214}Bi . The corresponding results of ^{232}Th were determined using the gamma-ray lines 2614keV of ^{208}Tl . The activities of ^{40}K were determined from its 1460keV γ -line. Counting time interval was 18,000 seconds. The background spectrum was recorded immediately after or before the sample counting. The specific activity concentration of ^{238}U was determined using the gamma-lines 1765keV of ^{214}Bi . The corresponding results of ^{232}Th were determined using the gamma-ray lines 2614keV of ^{208}Tl . The activities of ^{40}K were determined from its 1460KeV γ -line. Counting time interval was 18,000 seconds. The background spectrum was recorded immediately after or before the sample counting. The distribution of ^{238}U , ^{232}Th and ^{40}K in soil is not uniform. Uniformity with respect to exposure to radiation has been defined in terms of radium equivalent activity Ra_{eq} in Bq/kg to compare the specific activity of materials containing different amounts of ^{238}U , ^{232}Th and ^{40}K . It is calculated using the following relation [12, 13]:

$$Ra_{eq} = A_U + 1.43 A_{Th} + 0.07 A_K \quad (1)$$

Where A_U , A_{Th} and A_K are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K in Bq/kg, respectively. While defining Ra_{eq} activity according to equation (1), it has been assumed that 370Bq/Kg for ^{238}U or 259Bq/kg for ^{232}Th or 4810Bq/kg for ^{40}K produce the same gamma dose rate [14]. The external gamma absorbed dose rate in the air at 1m above ground level was calculated from the measured activities of ^{238}U , ^{232}Th and ^{40}K in soil assuming that other radionuclides, such as ^{137}Cs , ^{113}Cd and ^{235}U series can be neglected as they contribute very little to the total dose from environmental background [15,16]. The calculations were performed according to the following equation [2]:

$$D = 0.462 A_U + 0.604 A_{Th} + 0.042 A_K \quad (2)$$

Where D is the dose rate in nGy/h. The external hazard index, H_{ex} , is defined as [12]:

$$H_{ex} = A_U/370 + A_{Th}/259 + A_K/4810 \quad (3)$$

The value of this index must be less than unity in order to keep the radiation hazard insignificant. The maximum value of H_{ex} equal to unity corresponds to the upper limit of radium equivalent activity 370Bq/kg. To estimate the annual effective dose, the following must be taken into account: (a) The conversion coefficient from absorbed dose in air to effective dose and (b) The indoor occupancy factor. Using the dose rate data obtained from the concentration values of natural radionuclides in soil, adopting the conversion factor of 0.7 Sv/Gy [2] from absorbed dose rate in air to effective dose received by adults and considering that people in Hat Yai district, on the average, spent 20% of their time outdoors, the annual effective doses are calculated [2]:

$$\text{Outdoor annual effective dose (Sv)} = D \times 24 \times 365 \times 0.7 \times 0.20 \quad (4)$$

$$\text{Indoor annual effective dose (Sv)} = D \times 24 \times 365 \times 0.7 \times 0.80 \quad (5)$$

3. RESULTS AND DISCUSSION

Fig. 2 shows the distribution of specific activity concentrations of ^{40}K , ^{238}U and ^{232}Th radionuclides for 24 samples of soils collected from 8 different locations in Ur city region, taken from three different depths 0-5, 5-10 and 10-15 cm. For ^{40}K , the values have been found to vary between 215.75 ± 8.91 Bq/kg in sample A3 and 397.38 ± 12.02 Bq/kg in sample A1 at depth 5-10cm with an average value 314.62 ± 11.3 Bq/kg. For ^{238}U vary from 9.99 ± 2.56 Bq/kg in sample A3 to 29.93 ± 2.98 Bq/kg in sample A7 at depth 5-10cm with an average of 17.9 ± 3.02 Bq/kg. While for ^{232}Th it varies from 7.77 ± 2.24 Bq/kg in sample A6 at depth 10-15cm to 25.66 ± 2.55 Bq/kg in sample A1 at depth 5-10cm with an average of 13.66 ± 2.41 Bq/kg. Hence, at depth 0-5cm, the minimum values for ^{40}K , ^{238}U and ^{232}Th radionuclides are 245.57 ± 9.35 Bq/kg in sample A6, 14.2 ± 3.07 Bq/kg in sample A7 and 10.74 ± 2.84 Bq/kg in sample A7 respectively, while the maximum values are 393.35 ± 12.1 Bq/kg recorded in sample A3, 21.97 ± 2.96 Bq/kg in sample A8 and 24.26 ± 2.87 Bq/kg in sample A1, with an average of 318.2 ± 11.05 Bq/kg, 17.33 ± 3.04 Bq/kg and 13.83 ± 2.33 Bq/kg respectively. For depth 5-10cm, the minimum value for ^{40}K , ^{238}U and ^{232}Th radionuclides is 257.29 ± 10.67 Bq/kg in sample A3, 9.99 ± 2.56 Bq/kg in sample A3 and 8.33 ± 1.65 Bq/kg in sample A3 respectively, while the maximum value is 397.38 ± 12.02 Bq/kg in sample A1, 29.93 ± 2.98 Bq/kg in sample A7 and 25.66 ± 2.55 Bq/kg sample A1, with an average values 326.71 ± 11.16 Bq/kg, 19.66 ± 2.94 Bq/kg and 15.54 ± 2.44 Bq/kg respectively. For depth 10-15cm, the minimum value for ^{40}K , ^{238}U and ^{232}Th radionuclides is 215.75 ± 8.91 Bq/kg in sample A3, 11.79 ± 2.82 Bq/kg in sample A3 and 7.77 ± 2.24 Bq/kg recorded in sample A6 respectively, while the maximum value is 394.62 ± 11.24 Bq/kg in sample A5, 23.73 ± 2.89 Bq/kg recorded in sample A7 and 15.38 ± 2.48 Bq/kg recorded in sample A7, with an average values 298.95 ± 10.8 Bq/kg, 16.71 ± 3.09 Bq/kg and 11.7 ± 2.45 Bq/kg respectively. Generally, the specific activity concentration values in all soil samples of Ur city region are less than the worldwide average 35Bq/kg for ^{238}U , 30Bq/kg for ^{232}Th and 400Bq/kg for ^{40}K [2].

Fig. 3 shows the Radium equivalent Ra_{eq} activities for the soil samples versus soil depth of the archeological UR city. Ra_{eq} values vary from 41.72 ± 5.74 Bq/kg in sample A8 to 92.29 ± 7.47 Bq/kg in sample A8 with an average value of 61.67 ± 7.03 Bq/kg. For depth 0-5cm, the

minimum value is $52.25 \pm 6.07 \text{ Bq/kg}$ recorded in sample A2 while the maximum value is $81.56 \pm 9.17 \text{ Bq/kg}$ in sample A1 with an average of $61.62 \pm 7.23 \text{ Bq/kg}$. For depth 5-10cm, the minimum value is $41.72 \pm 5.74 \text{ Bq/kg}$ recorded in sample A3 while the maximum value is $92.29 \pm 7.47 \text{ Bq/kg}$ in sample A1 with an average of $66.93 \pm 7.3 \text{ Bq/kg}$. For depth 0-15cm, the minimum value is $42.37 \pm 6.65 \text{ Bq/kg}$ recorded in sample A3 while the maximum value is $64.63 \pm 7.57 \text{ Bq/kg}$ in sample A1 with an average of $56.21 \pm 7.4 \text{ Bq/kg}$. It can be seen that the R_{eq} values for all soil samples are lower than the recommended permissible value 370 Bq/kg [2]. The activity levels due to terrestrial background radiation are related to the type of rock from which the soil originates. No correlation is found between activity and depth. The measured values of radioactivity show that it is randomly distributed in different depths of the soil.

Fig. 4 shows the distribution of absorbed dose rate (AD) for the soil samples versus soil depth of the archeological Ur city. The calculated results of the absorbed dose rate shows that the highest value is 43.78 nGy/h noted in sample A1 at depth 5-10cm, whereas the lowest value is 20.34 nGy/h noted in sample A3 at depth 5-10cm. The average value is 29.66 nGy/h . The measurements indicate that the absorbed dose rate values due to the depth of soil 0-5cm have a minimum value 25.24 nGy/h in sample A2 and a maximum value 38.72 nGy/h recorded in sample A1 with an average 29.77 nGy/h . For depth 5-10cm, the minimum value is 20.34 nGy/h recorded in sample A3 and the maximum value is 43.78 nGy/h in sample A1 with an average of 32.08 nGy/h . While for depth 10-15cm, the minimum value is 20.36 nGy/h in sample A3 and the maximum value is 31.38 nGy/h recorded in sample A1 with an average of 27.24 nGy/h . The values of absorbed dose rate for soil sample are below the worldwide average limit 55 nGy/h [17].

Fig. 5 shows indoor and outdoor annual effective doses for the soil samples versus soil depth of the archeological Ur city, respectively. The calculated values of annual effective doses shows that the maximum values of indoor and outdoor are 0.215 mSv/y and 0.054 mSv/y recorded in sample A1 at depth 5-10cm and the minimum values were 0.099 and 0.025 mSv/y recorded in sample A3 at depth 5-10cm. The average values of annual indoor and outdoor effective dose are 0.145 and 0.036 mSv/y respectively. For depth 0-5cm the minimum values of (indoor and outdoor) are 0.124 and 0.031 recorded in sample A2 while the maximum values are 0.19 and 0.047 recorded in sample A1 with an average values 0.145 and 0.036 respectively. For depth 5-10cm, the minimum values are 0.099 and 0.025 recorded in sample A3 while the maximum values are 0.215 and 0.054 recorded in sample A1 with an average values 0.157 and 0.039 respectively. For depth 10-15cm, the minimum values are 0.1 and 0.025 recorded in sample A3, while the maximum values are 0.154 and 0.038 recorded in sample A1 with an average values 0.133 and 0.033 respectively. All values of indoor and outdoor annual effective dose are less than worldwide average 1 mSv/y which is recommended by International Commission on Radiological Protection (ICRP 1993) [17].

The elemental concentrations of ^{238}U varies between 0.808 ppm in soil sample A3 and 2.423 ppm in soil sample A7 at depth 5-10cm, with an average value 1.449 ppm , whereas that of ^{232}Th varies between 1.914 ppm recorded in soil sample A6 at depth 10-15cm and 6.321 ppm recorded in soil sample A1 at depth 5-10cm, with an average value 3.366 ppm . For ^{40}K radionuclide, the elemental concentrations ranged from 0.689% recorded in sample A3 to 1.26% recorded in sample A5 at depth 10-15cm, with an average value 1.005% . For depth 0-5cm, the minimum values of the elemental concentrations of ^{40}K , ^{238}U , and ^{232}Th radionuclides are 0.784% recorded in sample A6, 1.15 ppm recorded in sample A7 and 2.645 ppm recorded in sample A7 respectively, while the maximum values are 1.256% recorded in sample A3, 1.778 ppm recorded in sample A8 and 5.975 ppm recorded in sample A1 respectively, with an average of 1.016% , 1.403 ppm and 3.408 ppm respectively. While for depth 5-10cm, the minimum values of are 0.822% recorded in sample A3, 0.808 ppm recorded in sample A3 and 2.052 ppm recorded in sample A3 respectively, while the maximum values are 1.126% recorded in sample A1, 2.423 ppm recorded in sample A7 and 6.321 ppm recorded in sample A1 respectively, with an average of 1.025% ,

1.591ppm and 3.807ppm respectively. Also for the depth 10-15cm, the minimum values of ^{40}K , ^{238}U , and ^{232}Th radionuclides are 0.689% recorded in sample A3, 0.954ppm recorded in sample A3 and 1.914ppm recorded in sample A6 respectively, while the maximum values are 1.26% recorded in sample A5, 1.921ppm recorded in sample A7 and 3.788ppm recorded in sample A7 respectively, with an average of 0.954%, 1.352ppm and 2.881ppm respectively. The Maximum value 5.975ppm of ^{232}Th elemental concentrations was recorded in soil sample A1 at depth 0-5cm, while maximum value 6.321ppm of ^{232}Th was recorded in soil sample A1 at the depth 5-10cm and the maximum value 3.788ppm of ^{232}Th was found in soil sample A7 at depth 10-15cm. These values are less than the worldwide average 7.38ppm for ^{232}Th . All results of elemental concentrations are within the permissible limits 1.28% for ^{40}K , 2.83ppm for ^{238}U , and 7.38ppm for ^{232}Th [2].

Fig. 6 shows the distribution of external and internal hazard index for the soil samples with the soil depth of the archeological Ur city. The maximum values of external and internal hazard index are 0.249 and 0.316 recorded in sample A1 at depth 5-10cm and the minimum values of external and internal hazard index are 0.112 and 0.139 noted in sample A3 at depth 5-10cm, the average values of external and internal hazard index are 0.166 and 0.215 respectively. Therefore, for the depth 0-5cm, the minimum values of external and internal are 0.141 and 0.18 recorded in sample A2 while the maximum values are 0.22 and 0.27 recorded in sample A1 with an average of 0.166 and 0.212 respectively. Also, for depth 5-10cm, the minimum values are 0.112 and 0.139 recorded in sample A3 while the maximum values are 0.249 recorded in sample A1 and 0.318 recorded in sample A7 with an average of 0.18 and 0.233 respectively. While, for 10-15cm depth, the minimum values are 0.114 and 0.146 recorded in sample A3 while the maximum values are 0.147 recorded in sample A1 and 0.233 recorded in sample A7 with an average of 0.152 and 0.197 respectively. All soil samples of Ur city are not exceed the upper limit for (H_{ex} and H_{in}), which is unity [2].

Finally, fig. 7 shows a set of gamma-ray spectra of a typical samples from the soil under study for Ur city, which analyzed by NaI(Tl) detector.

All the results demonstrated by this study make the archaeological of Ur city in a safe environment from exposure of radiation, and there is no risk to workers or tourists coming to see the archaeological ruins. Despite the presence of the military base and airport of Imam Ali (AS) military, near the archeological city of Ur (about 1.8 km). The activity levels due to terrestrial background radiation are related to the type of rock from which the soil originates. As regard of activity with respect to depth, no correlation was found between activity and depth. The measured values of radioactivity show that it's randomly distributed in different depth of the soil.

4. CONCLUSIONS

The environmental monitoring of natural background radiation using Sodium Iodide NaI(Tl) detector revealed is a good technique for measuring the distribution of the natural radiation levels in soil and pottery samples. From the obtained result, one could see that the distribution was not uniform. The measured values of radioactivity show that it is randomly distributed in different depth of the soil. Also artificial radionuclide was not detected in any of the measured samples. The obtained results confirm some conclusions as below:

- 1- The present work has shown that the radioactivity concentration of ^{238}U , ^{232}Th and ^{40}K are generally acceptable and have consistent values with those reported by many other countries in the world for samples.
- 2- Since the radiation levels were within permissible limits, thus the radioactive hazard is low for human beings (employees and tourists) in these areas.
- 3- Specific activity of ^{40}K is much higher than that of ^{238}U and ^{232}Th .

- 4- The gamma absorbed dose rate of samples varies appreciably from one sample to another due to the variation of ^{238}U , ^{232}Th and ^{40}K contents.
- 5- The estimated average absorbed dose rates in air for the studied area are below or comparable with world average (55nGy/h). All average values of annual indoor and outdoor effective dose were less than the permissible limit.

Fig. 2 Variation of specific activity concentration of ^{238}U , ^{232}Th and ^{40}K versus soil depth for Ur city region.

Fig. 3 Variation of Radium equivalent activity versus soil depth for Ur city region.

Fig. 4 Variation of absorbed dose rate versus soil depth for Ur city region.

Fig. 5 Variation of annual effective doses (indoor and outdoor) versus soil depth for Ur city region.

Fig. 6 Variation of (external and internal) hazard index versus soil depth for Ur city region

Fig. 7 Gamma-ray spectrum of three typical soil samples from Ur city region.

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