



Assessment of Natural Radioactivity in Building Material of the Ancient City of Tell - Zurghul in Thi-Qar Southern Iraq

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Abstract

In this study, the qualitative activity of natural radionuclides was measured for building materials samples, which were collected from an archaeological area in the Sumerian city called "Tell Zughrul". Ten samples were collected from different types of building materials used in building houses, places of worship, and livestock pens. A 3"x3" NaI (Tl) gamma-ray spectroscopy system was used. The results of the measurements showed that the specific activity of U-238, Th-232, Ra-226, and K-40 is ranged (from 10.14 - 23.63 Bq/kg) with an average of (16.96 Bq/kg), (9.28 - 18.27 Bq/kg) with an average of (15.11 Bq/kg), (6.96 - 20.8 Bq/kg) with an average value of (15.2 Bq/kg), and (127.93 - 274.4 Bq/kg) with an average value (209.102 Bq/kg) respectively. To assess the Radiological Hazards of the radioactivity in building material samples, Radium Equivalent Activity, Internal and External Hazard Index, Absorbed Gamma Dose, Representative level index, Annual Effective Dose, and Excess Lifetime Cancer Risk were determined. The results show that all samples of building materials for the ancient area of (Tell Zughrul), were less than the recommended max. Limit values provided by UNSCEAR and ICRP reports, although there are differences in the concentration values of these elements according to the samples.

1056

Key Words: Natural Radioactivity, Building Materials, Gamma-ray Absorbed Dose, NaI (Tl), Sumerian City.

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Introduction

The archaeological sites of the Sumerian civilization (4100-1750 BC) are unquestionably one of the most important archaeological sites in the world. Situated in the Thi-Qar governorate at a distance (375 km) south of the Baghdad governorate. The Sumerians were considered one of the first peoples who inhabited Mesopotamia and settled in the Sumer region in southern Iraq [1]. Humans are naturally exposed to two main sources of radiation, high-energy cosmic rays, and radioactive nuclides available in the earth's crust, which are found in water, air, and soil, radioactive elements, have existed since the creation of the earth and have half-life estimated at millions of years [2].

There are small amounts of uranium and thorium in the earth's crust, in addition to other radioactive

elements such as potassium, which is one of the most important sources of human exposure. [3] Knowing the concentration and distribution of radionuclides is very important in providing the necessary information to monitor any radioactivity that affects the environment. Many isotopes, both natural and artificial, have a property known as Radioactivity [4]. It is the process of spontaneous transformation of the radioactive nuclei of an element into the nuclei of atoms of the elements others are more stable by emitting a certain type of radiation.

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Studies show that there are three types of radiation



emitted: alpha, beta, and gamma [5] the radioactivity Sources two types, sources originating from outside the Earth (cosmic rays) and radioactive sources in the crust of the earth, and these may come from naturally occurring radioactive elements, such as the materials surrounding us [6] however, Gamma rays are emitted from naturally occurring radionuclides such as potassium and product radionuclides from the uranium and thorium chains and are one of the external sources of radiation to which the human body is exposed [7] Gamma rays are electromagnetic radiation produced by nuclear interactions. It is generally characterized as high-energy radiation and short wavelengths within the electromagnetic spectrum. This high energy can cause serious damage when absorbed by living cells. Due to the nature of its penetration depth, the shielding of gamma rays requires a large amount of mass. Usually, it is preferable to use materials with high atomic numbers and high density in order to be better absorption [8]. Tell-Zurghul, which is the remains of a sacred city that were inhabited by priests and Nobles from the Sumerians in the era 2700-2100 BC, is one of the most important archaeological sites that remain so far, despite the difficult environmental conditions in this region. From here, this study came to know the natural radioactive content of building materials used in this city, and thus knowledge of the Radioactive Environment of this region at that time.

Theoretical Consideration

1. Specific Activity Concentration

The specific radioactive activity concentration defined as the activity per unit mass of the radioactive substance and measured in units of curies per gram or Becquerel per kilogram, and can be calculated using the following equation,

$$A \text{ (Bq/Kg)} = \frac{N}{t \times I_{\gamma}(E_{\gamma}) \times \varepsilon(E_{\gamma}) \times m} \pm \frac{\sqrt{N}}{t \times I_{\gamma}(E_{\gamma}) \times \varepsilon(E_{\gamma}) \times m} \quad (1)$$

Where, (N) is the net area under the gamma-ray peak measured for the spectrum after subtraction of the radiation background in (Bq / Kg), (t) the measurement time(sec) and $I_{\gamma}(E_{\gamma})$ the gamma-ray intensity of the measured gamma energy and, ($\varepsilon(E_{\gamma})$) the efficiency of the detector at the measured gamma-ray energy and (m) sample weight (kg).[9]

2. Radium Equivalent Activity (Ra_{eq})

The value of the equivalent concentration of radium (Ra_{eq}) that is used to estimate the hazards associated with materials containing radium ($^{226}_{88}\text{Ra}$), thorium ($^{232}_{90}\text{Th}$), and potassium ($^{40}_{19}\text{K}$) radioactive, which is calculated assuming that there is a concentration of (370Bq/Kg) for radium in this material or (260Bq/Kg) for thorium or (4810Bq/Kg) for potassium, which produces the same Dosage for gamma-rays. The Radium equivalent activity concentration (Ra_{eq}) calculated using the following equation:

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

Since (A_{Ra} , A_{Th} , and A_K) are Radioactivity concentrations Radium, Thorium and Potassium, respectively, and it is measured in Becquerel per kilogram [9].

3. Internal (H_{in}) and External (H_{ex}) Hazard Index

External risk factors are used to determine external hazards caused by gamma radiation, which estimate the expected gamma dose a person may receive through direct external exposure to materials containing gamma radiation, while, Internal risk factors determine the dose limits an individual receives in a work environment involving natural radiation activity, which is exposed by swallowing or inhaling gamma-ray emitters. It is possible to calculate the internal (H_{in}) and external (H_{ex}) hazard index using the following formulas:

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3)$$

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (4)$$

Where (A_{Ra} , A_{Th} , and A_K) are radioactivity concentrations of Radium, Thorium and Potassium, respectively, in (Bq/Kg) [10].

4. Absorbed Gamma Dose (D_{γ})

The absorbed gamma-ray dose is the energy absorbed per unit mass of a body exposed to gamma radiation. This term is used for all types of radiation and energies for all bodies and substances. The rate of doses absorbed because of radiation is uniformly distributed for naturally occurring radionuclides. It is calculated based on the recommendations of ICRP [11] in a unit of (nGy/h) using the following equation:

$$D_{\gamma} \text{ (ICRP)} = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (5)$$

The conversion factors used to calculate the absorbed dose of gamma rays for each activity concentration (Bq/Kg) correspond to (0,462nGy/h for radium $^{226}_{88}\text{Ra}$), (0,604nGy/h for thorium ($^{232}_{90}\text{Th}$),



and (0,417nGy/h for potassium (^{40}K) [11].

5. Representative Level Index (I_{yr})

The Representative level index (I_{yr}) has been used to estimate the level of radiological danger of gamma rays associated with natural radionuclides in the measured samples. A factor represents the (Organization for Economic Co-operation and Development OECD) index. The representative level index can be calculated from the following equation derived by the OECD group.

$$I_{yr} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \quad (6)$$

Where A_{Ra} , A_{Th} , and A_K are radioactivity concentrations of Ra-226, Th-232, and K-40, respectively, measured in Becquerel per kilogram Bq/Kg [12].

6. Annul Effective Dose (AED)

To calculate the annual effective dose, the conversion factor from the absorbed dose to the effective dose and the internal survival factor must be taken into account, and to calculate the effective dose of the gamma-ray emitting element (UNSCEAR 2000). The conversion constant of (0.7 Sv/Gy) was adopted as a conversion factor from the dose absorbed in the air to the annual effective dose received by adults, as well as the percentage of time of 80% that spent inside the laboratory or residence and 20% is the percentage of time spent abroad. From these data, it was found that the annual effective dose is calculated as follows [13]:

$$\text{Indoor AED (mSv/y)} = AD(\text{nGy/h}) \times 10^{-6} \times 8760 \text{ h/y} \times 0.8 \times 0.7 \text{ Sv/Gy} \quad (7)$$

$$\text{Outdoor AED (mSv/y)} = AD(\text{nGy/h}) \times 10^{-6} \times 8760 \text{ h/y} \times 0.2 \times 0.7 \text{ Sv/G} \quad (8)$$

Where the number (8760) is the number of hours in one year.

7. Excess Lifetime Cancer Risk (ELCR)

Excess Lifetime Cancer Risk (ELCR) it is a factor used to calculate the risk arising from gamma radiation associated with the natural radionuclides in the studied samples. It gives the probability that the worker in this environment will be infected by a group of people present in this place, meaning that it gives the percentage of those who will develop cancer as a result of the annual effective dose received. [14].

$$\text{ELCR} = \text{AED} + \text{DL} + \text{RF} \quad (9)$$

Whereas (AED) the annual effective dose (DL) is the life expectancy of a person and is approximately equal to 70 years, and (RF) is the risk factor for infection per Sievert and is equal to (0.05) for the general population according to ICRP [11].

Materials and Methods

1. Area of the Study

Tell Zurghul, (31°22'36.0"N, 46°29'43.0"E), It was called (Nina) or (Szara) or (Uru-Ku-ug) meaning the holy city and it was a center for the worship of the god (Natcha). located about (31 km) east of Shatrah city, and about (16.17 km) south of Al-Dawaya city. Fig. (1) Show the location of area of the study.

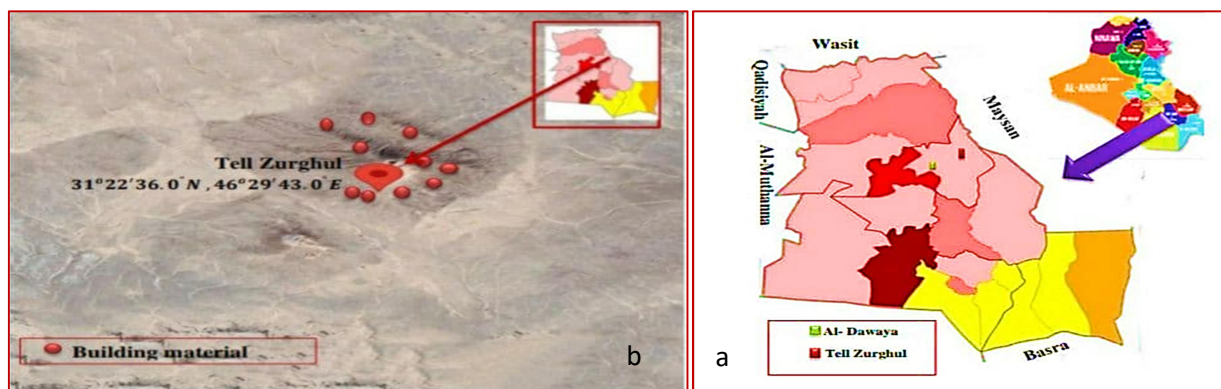


Fig. 1. (a) Map of Thi-Qar governorate showing the area covered by the study. (b) A picture of the archaeological site of Tell Zurghul [Google Earth]

2. Sample Preparation

Ten samples of building materials were collect. The samples were stored in airtight plastic boxes and

then transferred to the laboratory of the Environmental and Radiation Pollution Research Unit, Department of Physics, College of Science, University of Thi-Qar. To get rid of moisture, the



samples were processed and dried in an electric oven at a temperature of 80 °C for Twenty-Four hours. After drying, the samples were ground using a micro plant grinder into a granular size of 1000 microns and becomes ready for measurement. The samples were tightly sealed and stored for one month to get an appropriate radioactive equilibrium before measurement. Table (1) contain the collected samples and their types.

Table 1. The collected samples from the area of the study (Tell Zurghul site) and their types

No	Sample ID	Type of Sample
1	ZS1	Bricks
2	ZS2	Bricks
3	ZS3	Stone
4	ZS4	Clay bricks
5	ZS5	Clay bricks
6	ZS6	Stone
7	ZS7	Bricks
8	ZS8	Clay bricks
9	ZS9	Clay bricks
10	ZS10	Stone

3. Sample Measurements

A (76mm×76mm) NaI (Tl) Teledyne Isotope

gamma spectrometry system was used. This system is characterize by a good energy resolution, close to (7.5%) at an energy line of (661.7Kev) which is emitted from the radioactive($^{137}_{55}\text{Cs}$). The system was calibrated using two types of standard gamma-ray sources, first, is a natural source represented by a standard radioactive Thorium oxide (ThO_2) and the other is a standard radioactive point sources. Fig. (2) Represents a diagram of the gamma spectroscopy system used in the present study. The natural radionuclides targeted in the present study are U-238, Th-232, Ra-226, and K-40, table (2) contains these nuclides, and the energy of gamma ray photons approved for their measurements. The minimum detectable activity (MDA) of the system was calculated according to reference [15], as shown in table (3).

Table 2. The natural radionuclides targeted in the present study, and the energy of gamma ray photons approved for their measurements

Parent	Daughter	Energy (KeV)	Intensity %
$^{238}_{92}\text{U}$	$^{214}_{83}\text{Bi}$	1764.4	15
$^{232}_{90}\text{Th}$	$^{208}_{81}\text{Tl}$	583.0	84
$^{226}_{88}\text{Ra}$	$^{214}_{83}\text{Bi}$	609.3	46
$^{40}_{19}\text{K}$	Naturel	1460.8	10.6

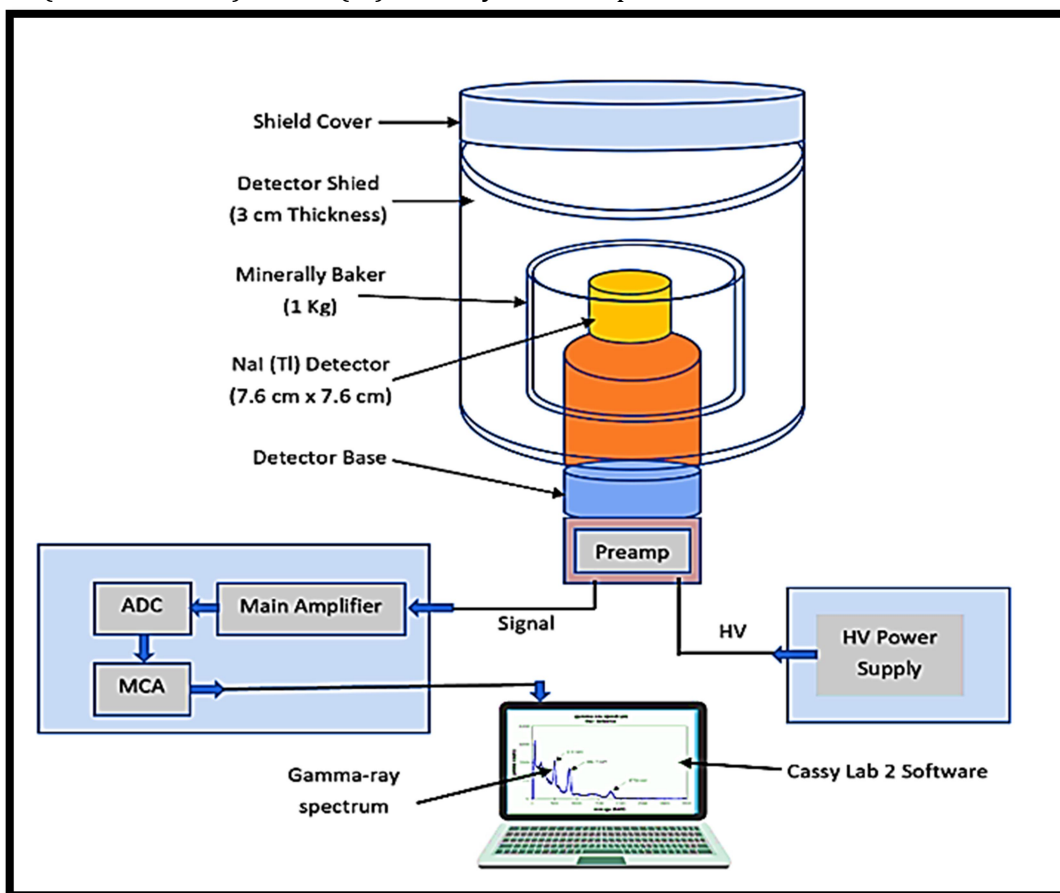


Fig. 2. A diagram of gamma spectroscopy system used in present study

Table 3. Calculated minimum detectable activity (MDA) for the radionuclides targeted in the present study

Nuclide	$E_{\gamma}(KeV)$	$I_{\gamma}\%$	$\epsilon(E_{\gamma})$	MDA (Bq/Kg)
$^{232}_{90}Th(^{208}_{81}Tl)$	583	84	0.0719	0.1198
$^{226}_{88}Ra(^{214}_{83}Bi)$	609.3	46	0.0671	0.1069
$^{238}_{92}U(^{214}_{83}Bi)$	1764.4	15	0.0121	0.295
$^{40}_{19}K(Naturel)$	1460.8	10.6	0.0164	11.070

Results and Discussion

From the results of measurements and equation (1), the natural radioactive elements concentrations of Tell-Zurghul site building materials samples was calculated for U238, Th232, Ra226, and K40 as shown in table (4). The results showed that the concentration of $^{238}_{92}U$ for all samples ranged between (10.14±0.38 - 23.63±0.58) Bq/Kg at an average of (16.96± 0.48) Bq/Kg, as for, $^{232}_{90}Th$ concentrations, they ranged between (9.28±0.20–18.27±0.29) Bq/Kg, with an average of(15.11±0.26)Bq/Kg, As for radium $^{226}_{88}Ra$ concentrations, they ranged between (6.96±0.08–

20.8±0.14) Bq/Kg and with an average of (15.2±0.12) Bq/Kg, Finally, the results of potassium $^{40}_{19}K$ concentrations appeared Between (127.93±1.46-205.21±1.86) Bq/Kg, with an average of (209.102±1.87)Bq/Kg. Fig. (3) Concentrations of radioactivity for each of uranium $^{238}_{92}U$, thorium $^{232}_{90}Th$, radium $^{226}_{88}Ra$, and potassium $^{40}_{19}K$ for building materials samples for Tell-Zurghul. All measurements were within the internationally permissible range and according to the recommendations of organizations such as (ICRP).

Table 4. Radioactivity concentrations of $^{238}_{92}U$, $^{232}_{90}Th$, $^{226}_{88}Ra$, and $^{40}_{19}K$ For building materials samples for Tell-Zurghul

Sample ID	$^{238}_{92}U$ (Bq/Kg ± Err)	$^{232}_{90}Th$ (Bq/Kg ± Err)	$^{226}_{88}Ra$ (Bq/Kg ± Err)	$^{40}_{19}K$ (Bq/Kg ± Err)
ZS1	20.36 ± 0.53	17.34 ± 0.28	18.23 ± 0.13	241.2 ± 2.01
ZS2	10.14 ± 0.38	9.28 ± 0.20	6.96 ± 0.08	127.93 ± 1.46
ZS3	16.02 ± 0.48	13.0 ± 0.24	15.38 ± 0.12	210.05 ± 1.88
ZS4	16.67 ± 0.49	14.6 ± 0.26	13.06 ± 0.11	214.51 ± 0,90
ZS5	18.15 ± 0.52	18.27 ± 0.29	16.03 ± 0.13	237.77 ± 1.16
ZS6	12.19 ± 0.42	12.24 ± 0.24	11.7 ± 0.11	153.74 ± 1.60
ZS7	20.03 ± 0.53	18.27 ± 0.29	18.27 ± 0.13	173.84 ± 1.70
ZS8	23.63 ± 0.58	17.08 ± 0.28	20.8 ± 0.14	252.3 ± 2.02
ZS9	14.74 ± 0.46	16.08 ± 0.27	14.2 ± 0.12	205.21 ± 1.86
ZS10	17.69 ± 0.50	14.78 ± 0.26	17.3 ± 0.13	274.4 ± 2.14
Average	16.96 ± 0.48	15.11 ± 0.26	15.2 ± 0.12	209.102 ± 1.87
Word. Ave. [15]	35	30	35	400



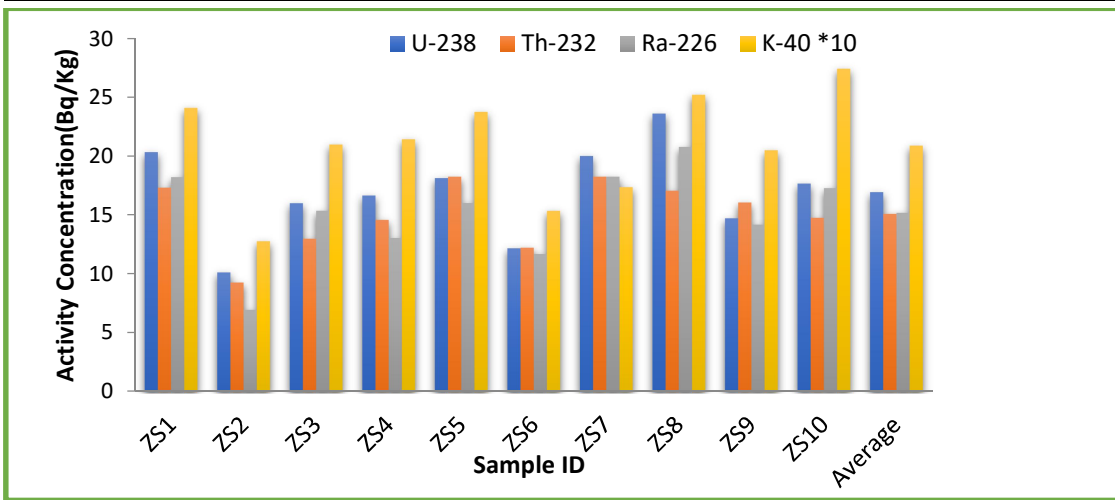


Fig. 3. Radioactivity concentrations of ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K For all samples

Table (5) includes the results of the calculations of absorbed gamma doses estimated in units of ($\text{nGy} \cdot \text{h}^{-1}$), which was calculated according to the equation (5) approved by the International Commission on Radiation Protection published 60(ICRP60) which is symbolized in the present study by the symbol D_{ICRP} , where its value ranged between ($14.16 - 30.45$) $\text{nGy} \cdot \text{h}^{-1}$, with an average of ($24.86 \text{ nGy} \cdot \text{h}^{-1}$). Fig. (4) showing absorbed gamma dose as a function of sample ID. Using equation (2), the radium equivalent ($^{226}\text{Ra}_{eq}$) values were calculated, which ranged between, ($30.08-64.66$) Bq/Kg with an average of (52.89 Bq/Kg), Fig. (5) showing the radium equivalent as a function of sample ID. The table also includes the internal and external annual effective dose, which was calculated through equations (7) and (8), approved by (ICRP), where the annual effective dose(ADE) (indoor) values ranged between ($0.069-0.149$) $\text{mSv} \cdot \text{y}^{-1}$ with an average of $0.122 \text{ mSv} \cdot \text{y}^{-1}$ As for the annual effective dose(ADE) (outdoor), it ranged between ($0.0173-0.0373$) $\text{mSv} \cdot \text{y}^{-1}$, with an average of $0.031 \text{ mSv} \cdot \text{y}^{-1}$. Fig. (6) Shows the

annual effective dose (ADE) (indoor and outdoor) of the samples, the table also included calculating the coefficient of increasing the likelihood of developing cancer during the lifespan of the individual, ELCR (Excess Lifetime Cancer Risk) which was calculated from the relation (9), where its value ranged between ($0.061 \times 10^{-3} - 0.131 \times 10^{-3}$), with an average of (0.107×10^{-3}), Fig. (7) shows the variation in the coefficient of increased hazard of developing cancer (ELCR). The OECD representative level index was also calculated using the equation (6), which is symbolized by the symbol (I_{yr}), where the values ranged between ($0.224-0.477$) Bq/Kg with an average of 0.392 Bq/Kg. Fig. (8) Showing the levels of this indicator according to the samples. And also calculated the external hazard (H_{ex}) and internal hazard (H_{in}) coefficient for the measured samples, and their values ranged between ($0.081-0.175$) Bq/Kg and ($0.100-0.231$) Bq/Kg, and an average of 0.143 Bq/Kg and 0.184 Bq/Kg, respectively. Fig. (9) Showing external and internal hazard coefficient.

Table 5. Calculated absorbed dose according to the recommendations of the ICRP60 and the annual effective dose, as well as the coefficient of Estimating the risk of developing cancer, the estimated gamma Coefficient and the internal and external hazard coefficient

Sample ID	D_{ICRP} (nGy/h)	Ra_{eq} (Bq/Kg)	AED (mSv/y)		ELCR $\times 10^{-3}$	I_{yr} (Bq/kg)	H_{ex} (Bq/kg)	H_{in} (Bq/kg)
			In*	Out*				
ZS1	28.95	61.59	0.142	0.036	0.124	0.456	0.166	0.215
ZS2	14.16	30.08	0.069	0.017	0.061	0.224	0.081	0.100
ZS3	23.76	50.24	0.117	0.029	0.101	0.373	0.136	0.177
ZS4	23.83	50.54	0.117	0.029	0.102	0.377	0.136	0.171
ZS5	28.35	60.46	0.139	0.035	0.122	0.448	0.164	0.206
ZS6	19.21	41.04	0.094	0.024	0.082	0.303	0.111	0.142
ZS7	26.73	57.78	0.131	0.033	0.115	0.420	0.156	0.205
ZS8	30.45	64.66	0.149	0.037	0.131	0.477	0.175	0.231
ZS9	24.84	53.02	0.122	0.031	0.106	0.392	0.143	0.182
ZS10	28.36	59.56	0.139	0.035	0.121	0.446	0.161	0.208
Average	24.86	52.89	0.122	0.031	0.107	0.392	0.143	0.184
Word. Ave. [15]	55	350	≤ 1	≤ 1	0.29	≤ 1	≤ 1	≤ 1



*In=indoor, out=outdoor

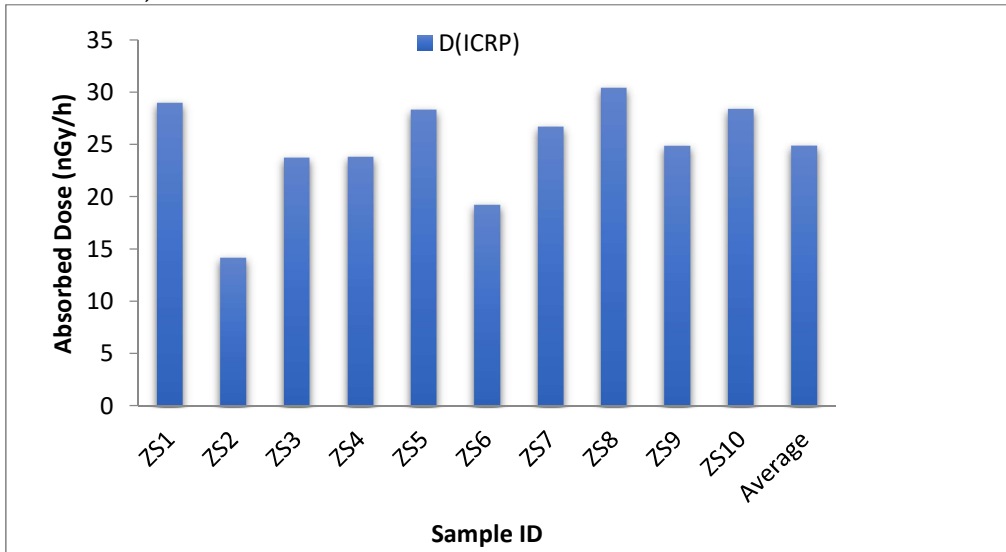


Fig. 4. Calculated absorbed Gamma Dose According to ICRP60, For all Samples

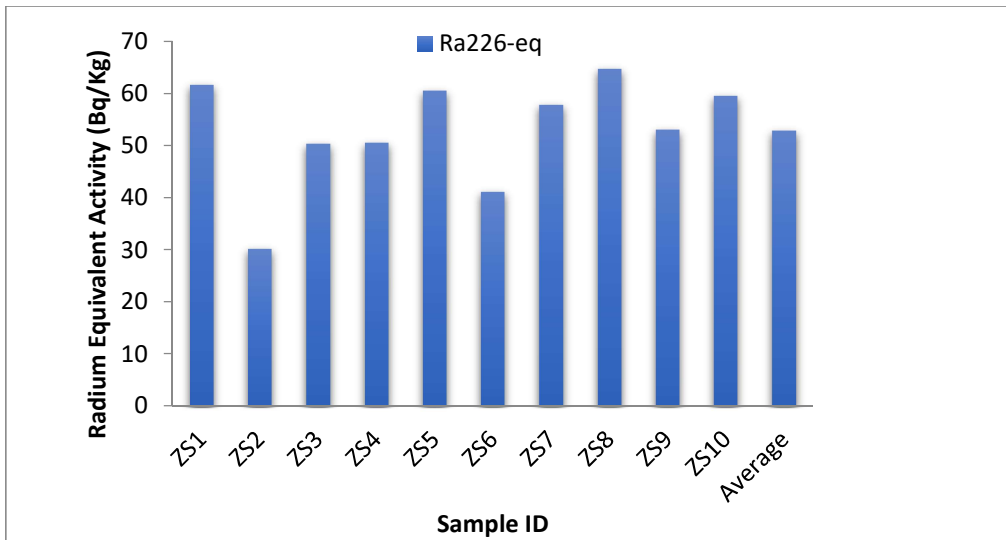


Fig. 5. Equivalent concentration of radium, for all samples

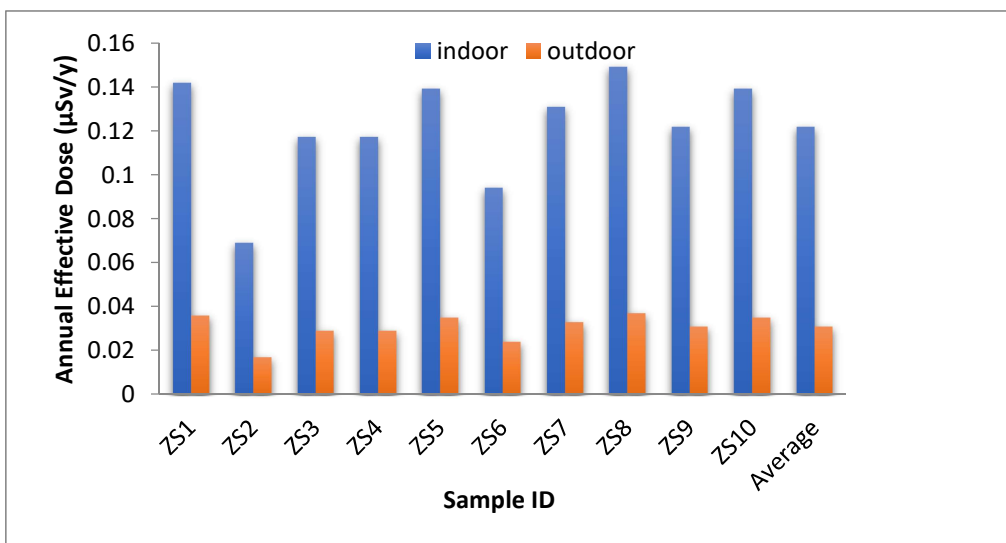


Fig. 6. Annual equivalent dose (Indoor& Outdoor) calculated for all samples

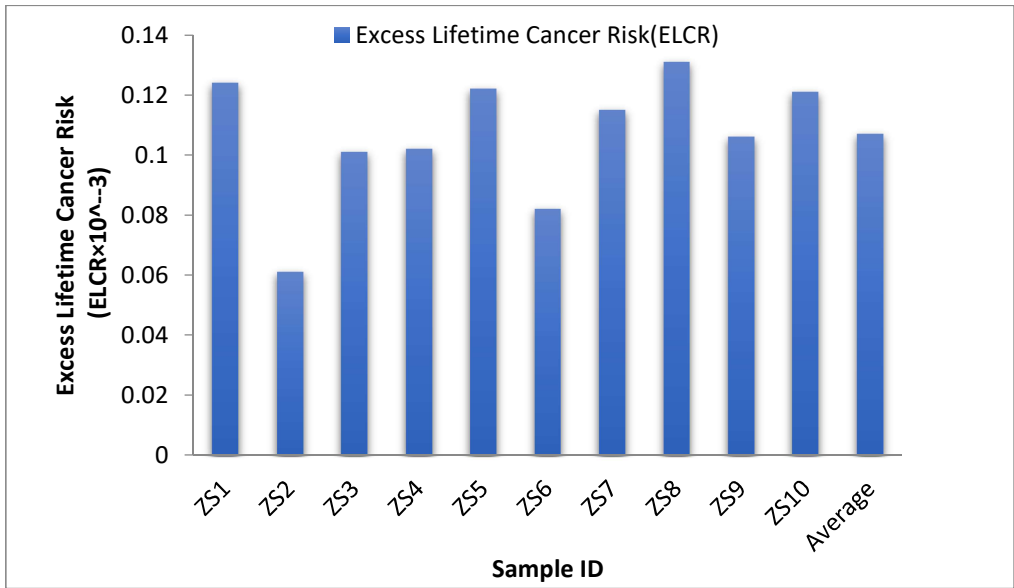


Fig. 7. The excess lifetime cancer risk (ELCR) calculated according to the Recommendations of (ICRP60) for all samples

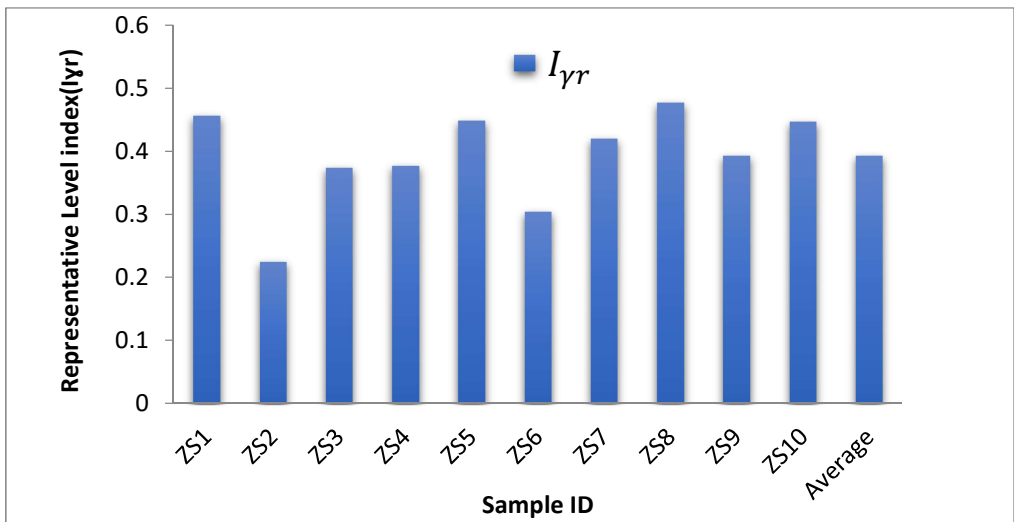


Fig. 8. Gamma-Ray representative level Index calculated according to the Recommendations (ICRP60) for all Samples

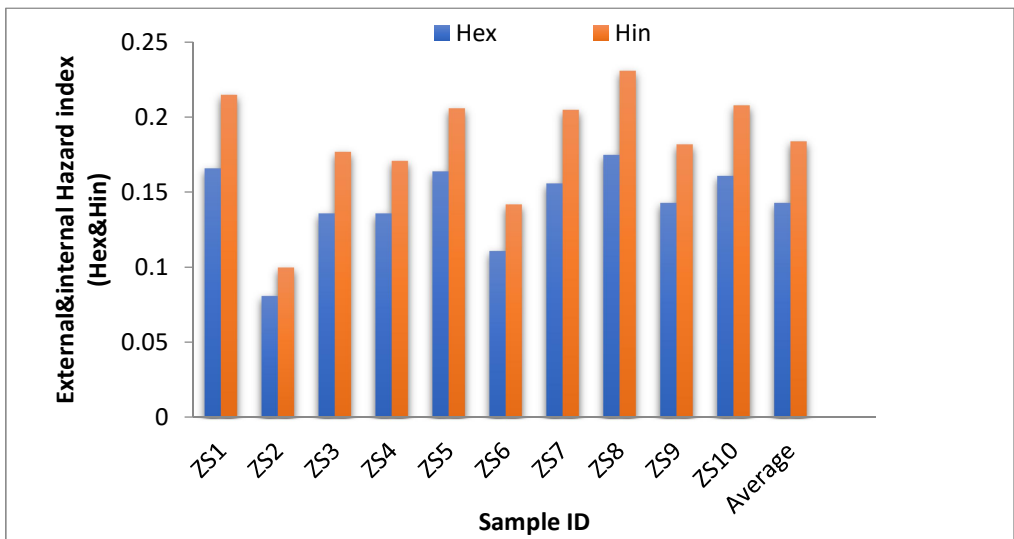


Fig. 9. External(Hex) and Internal (Hin) hazard coefficient for all Samples

Conclusion

The current study showed that the levels of natural radioactivity in the building materials used in the archaeological city of Tell-Zurghul that date back to the pre-third millennium BC are within the permissible limits according to the recommendations of international organizations and agencies, such as (ICRP), (UNCSEAR) and (EPA), which means that the environment that was these cities and sites are built are similar to the current environment. That means there is no change in the environment of natural radiation in these sites.

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