

Assessment of natural radioactivity in building material of the ancient city of Tell- Al Hiba in Thi-Qar southern Iraq

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Abstract

Tell al-Hiba is one of the most important ancient Sumerian sites, dating back to 1950 BC. This site is famous for having temples and places where priests lived, in addition to being an important economic site in that period. In the present study, the natural radioactivity concentrations of radionuclides were measured for building materials samples, collected from this site. Ten samples were collected from different kinds of building materials used in the building of houses and places of worship. A 3"×3" NaI (Tl) gamma-ray spectroscopy system was used. The results of the measurements showed that the specific activity of ^{238}U , ^{232}Th , ^{226}Ra , and ^{40}K is ranged (from 9.57- 36.57Bq/kg) with an average of (20.1Bq/kg), (8.31- 21.49Bq/kg) with an average of (15.99Bq/kg), (8.19 - 29.18Bq/kg) with an average value of (17.71Bq/kg), and (100.37 - 288.9 Bq/kg) with an average value (195.37Bq/kg) respectively. The Radium Equivalent Activity, Internal and External Hazard Index, Absorbed Gamma Dose, Representative Level Index, Annual Effective Dose, and Excess Lifetime Cancer Risk were calculated to evaluate the radiological hazards of radioactivity in building material samples. There are variations in radioactivity concentration values of these elements depending on the samples, but the results indicate that all samples of building materials for the ancient site of Tell Al-Hiba were less than the maximum allowed values recommended by international Organizations and agencies such as the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and International Commission on Radiological Protection (ICRP).

Keywords: Natural Radioactivity, Building Materials, Gamma Absorbed Dose, gamma spectroscopy system, Sumerian city.

1. Introduction

Unquestionably, the Sumerian civilization's archaeological sites (4100–1750 BC) rank among the most significant in the entire world. Located 375 km south of the Baghdad Governorate in the Thi-Qar Governorate. One of the earliest groups to inhabit Mesopotamia was the Sumerians, who lived in the Sumer area of southern Iraq [1]. High-energy cosmic rays and radioactive nuclides present in the earth's crust, which are present in water, air, and soil, are the two main sources of radiation to which humans are naturally exposed. Radioactive elements have existed since the creation of the earth and have a half-life estimated at millions of years [2]. Besides the radioactive potassium element which is one of

the most important sources of human exposure, the earth's crust also contains trace amounts of uranium, thorium, and other radioactive elements [3]. The information needed to monitor any radiation that impacts the environment depends on knowing the concentration and dispersion of radionuclides. Many isotopes, natural and artificial, possess a characteristic known as radioactivity [4] It is the process by which radioactive nuclei of one element spontaneously change into the nuclei of atoms of other elements that are more stable by generating a particular type of radiation [5]. There are three types of radiation that are released, are alpha particle, beta particles, and gamma radiation [6].

There are two types of natural radioactive sources, extraterrestrial sources (cosmic radiation) and radioactive sources in the Earth's crust, which may come from naturally radioactive elements such as our surrounding materials [7], However, gamma radiation is one of the external sources of radiation to which the human body is exposed and is emitted from naturally occurring radionuclides such as potassium and radionuclides created from Uranium and Thorium chains [8].

Nuclear processes result in electromagnetic radiation known as gamma radiation, described as electromagnetic radiation with short wavelengths and high energy. When absorbed by live cells, this high energy can result in significant damage. Gamma-ray protection demands a substantial amount of mass due to the nature of its penetrating. Typically, the best materials to utilize for absorption are those with large atomic numbers and high densities [9]. Tell Al-Hiba, the administrative capital of the archaeological city of Lagash, is one of the most important archaeological sites that persists, despite the difficult environmental conditions in this area. Hence, this study has come to find out the natural radioactive content of the building materials used in this city, thus, knowing the radioactive environment of this area at that time.

2. Theoretical Consideration

2.1 Specific radioactivity 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

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

where (N) is the net area under the photo-peak of the gamma-ray energy approved for spectrometry after subtracting the background radiation, (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) the mass of the samples (kg).[10]

2.2 Radium Equivalent Activity (Ra_{eq})

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

$$Ra_{eq} \left(\frac{\text{Bq}}{\text{Kg}} \right) = A_{\text{Ra}} + 1.43 \times A_{\text{Th}} + 0.077 \times A_{\text{K}} \quad (2)$$

Where (A_{Ra} , A_{Th} , and A_K) are the Radioactivity concentrations of $^{226}_{88}Ra$, $^{232}_{90}Th$, and $^{40}_{19}K$, respectively, measured in Becquerel per kilogram [11].

2.3 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 $^{226}_{88}Ra$, $^{232}_{90}Th$, and $^{40}_{19}K$, respectively, in (Bq/Kg) [12].

2.4 Absorbed Gamma Dose (D_γ)

The amount of energy that a body exposed to gamma radiation absorbs per unit of mass is known as the absorbed dose of gamma-ray. All forms of radiation and energies for all substances and bodies are referred to by this phrase. For naturally occurring radionuclides, the rate of doses absorbed as a result of radiation is uniformly distributed. It is calculated in accordance with the recommendation of ICRP [13]. In a unit of (nGy/h) using the following equation:

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

The conversion factors used to calculate the absorbed dose of gamma rays for each activity concentration (Bq/Kg) correspond to (0,462 nGy/h for $^{226}_{88}Ra$, (0.604 nGy/h for $^{232}_{90}Th$, and 0,417 nGy/h for $^{40}_{19}K$ [14].

2.5 Representative level index (I_{yr})

The amount of radiological hazard posed by gamma rays linked with natural radionuclides in the analyzed samples has been estimated using the Representative level index (I_{yr}). The Organization for Economic Co-operation and Development (OECD) index is represented by a factor. The OECD group's equation, which is shown below, can be used to calculate the representative level index.

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

Where A_{Ra} , A_{Th} , and A_K are radioactivity concentrations of $^{226}_{88}Ra$, $^{232}_{90}Th$, and $^{40}_{19}K$, respectively, measured in Becquerel per kilogram Bq/Kg [15].

2.6 Annual Effective Dose (AED)

In order to calculate the annual effective dose, the conversion factor of the absorbed dose to the annual effective dose received from adults is usually taken into account. The current study was based on the conversion factor (0.7 Sv/Gy) proposed by (UNSCEAR 2000). It also took into account that people spend about 80% of their time indoors and about 20% outdoor. The internal and external annual effective dose can be calculated from the following equations [16-19]:

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

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

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

2.7 Excess Lifetime Cancer Risk (ELCR)

The Excess Lifetime Cancer Risk (ELCR) formula used to estimate the risk from gamma radiation brought on by the naturally occurring radionuclides in the samples under investigation. It provides the likelihood that a worker exposed to a group of people present in this location will contract the disease, which translates to the proportion of persons who will develop cancer because of the annual effective dose received [11].

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

$$\text{ELCR (outdoor)} = \text{AED(outdoor)} + \text{DL} + \text{RF} \quad (10)$$

$$\text{ELCR (total)} = \text{ELCR (Indoor)} + \text{ELCR (outdoor)} \quad (11)$$

Whereas (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 [13].

3. Materials and Methods

3.1 Area of the study

Tall Al-Hiba ($31^{\circ}42'13.20''N, 46^{\circ}40'80.74''E$) is the real, location of the city of Lagash, which is 22 km east of Shatrah city, and 8 km south of Al-Dawaya city. Tell Al-Hiba is an important Sumerian site located in southern Iraq within the borders of the Thi-Qar governorate. In the middle of a fertile area, punctuated by irrigation canals, making it a city enjoying economic and commercial prosperity, economic and social stability inevitably leads to preparation for the emergence of a great dynasty that has ruled for six generations without interruption for more than a century. Fig. (1) Shows the location of the area of the study.

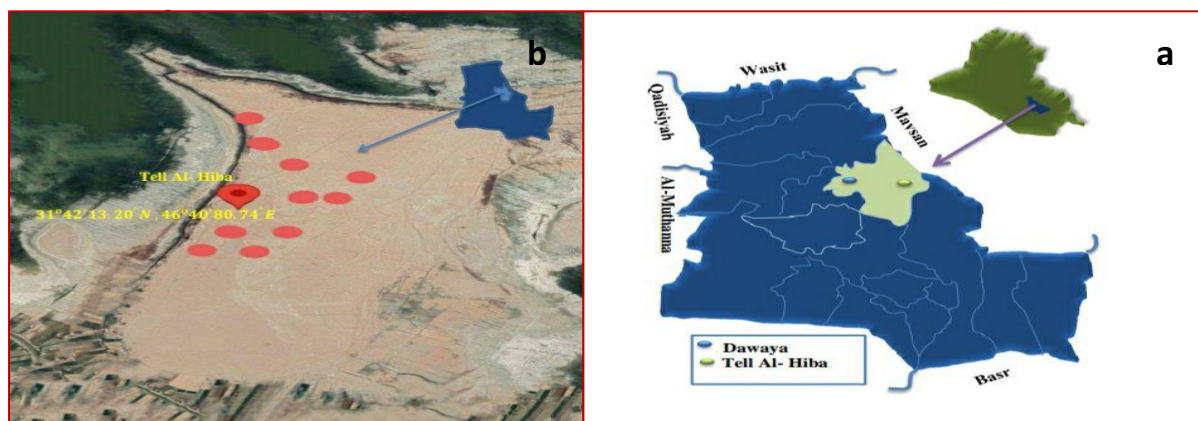


Figure 1. (a) Map of Thi-Qar governorate showing the area covered by the study. (b) An image of the archaeological site of Tell Al-Hiba [Google Earth].

3.2 Sample Preparation

Ten samples of various building materials were gathered. The samples were transferred to the lab of the Environmental and Radiation Pollution Research Unit, Department of Physics, College of Science, University of Thi-Qar, after being stored in airtight plastic boxes. The samples were processed and dried for 24 hours at $110^{\circ}C$ in an electric oven to remove any remaining moisture. The dried materials were ground using a

laboratory mill to a granular size of less than 1000 μm . Before measurement, the samples were stored in a hermetically sealed container for a month to achieve the proper radioactive equilibrium. Table (1) contain the collected samples and their types.

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

No	Sample ID	Type of Sample
1	HS1	Brick
2	HS2	Bitumen
3	HS3	Stone
4	HS4	Clay brick
5	HS5	Bitumen
6	HS6	Clay brick
7	HS7	Brick
8	HS8	Brick
9	HS9	Stone
10	HS10	Clay brick

3.3 Sample Measurements

A (76mm \times 76mm) NaI (TI) Teledyne Isotope gamma spectrometry system was used. This system is characterized 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 (ThO₂) and the other is a standard radioactive point source. Fig. (2) Represents a diagram of the gamma spectroscopy system used in the present study. The natural radionuclides targeted in the present study are $^{226}_{88}\text{Ra}$, $^{232}_{90}\text{Th}$, and $^{40}_{19}\text{K}$, 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 [10], 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
$^{232}_{90}\text{Th}$	$^{208}_{83}\text{Tl}$	583.0	84
$^{226}_{88}\text{Ra}$	$^{214}_{83}\text{Bi}$	609.3	46
$^{40}_{19}\text{K}$	Naturel	1460.8	10.6

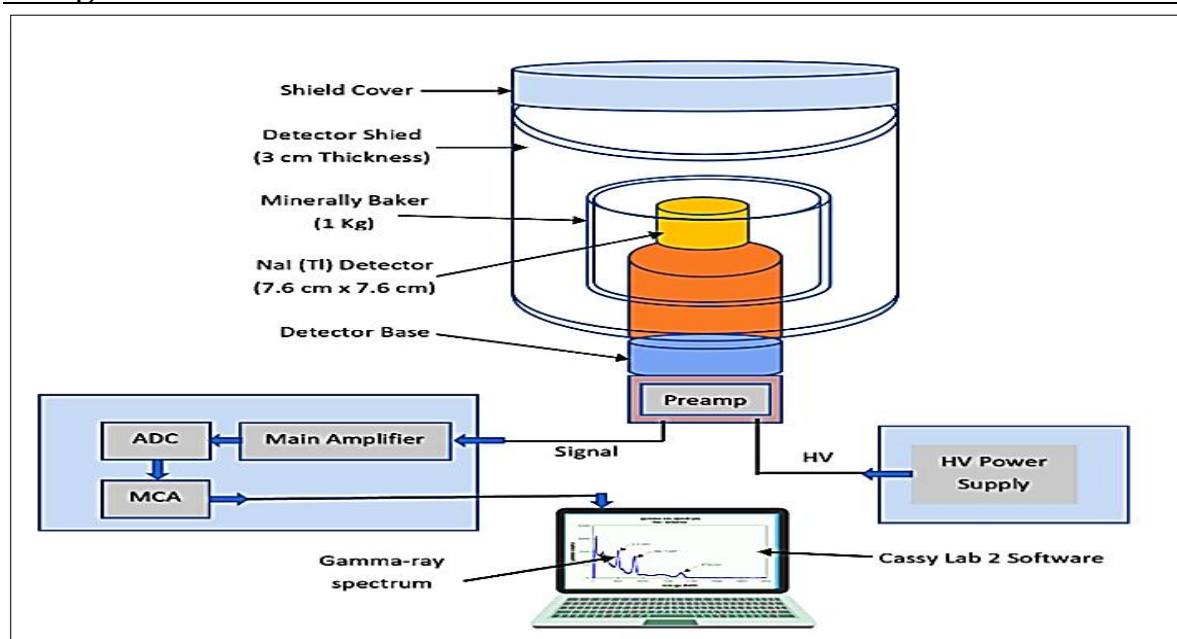


Figure. 2 A diagram of gamma spectroscopy system used in present study.

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}Tl)	583	84	0.0719	0.1198
$^{226}_{88}Ra$ (^{214}Bi)	609.3	46	0.0671	0.1069
$^{238}_{92}U$ (^{214}Bi)	1764.4	15	0.0121	0.295
$^{40}_{19}K$ (Naturel)	1460.8	10.6	0.0164	11.070

4. Results and Discussion

From the results of measurements and equation (1), the natural radioactive elements concentrations of Tell Al-Hiba site building materials samples were calculated for $^{238}_{92}U$, $^{232}_{90}Th$, $^{226}_{88}Ra$, and $^{40}_{19}K$ as shown in table (4). The results showed that the concentration of $^{238}_{92}U$ for all samples ranged ($9.57 \pm 0.29 - 36.57 \pm 0.58$) Bq/Kg at an average of (20.1 ± 0.41) Bq/Kg, as for $^{232}_{90}Th$ concentrations, they ranged ($8.31 \pm 0.05 - 21.49 \pm 0.074$) Bq/Kg, with an average of (15.99 ± 0.063) Bq/Kg, As for $^{226}_{88}Ra$ concentrations, they ranged ($8.19 \pm 0.062 - 29.18 \pm 0.12$) Bq/Kg and with an average of (17.71 ± 0.093) Bq/Kg, Finally, the results of $^{40}_{19}K$ concentrations appeared Between ($100.37 \pm 1.04 - 288.9 \pm 1.6$) Bq/Kg, with an average of (195.37 ± 1.3) Bq/Kg. Fig. (3) Shows the behavior of radioactivity concentrations of Uranium $^{238}_{92}U$, Thorium $^{232}_{90}Th$, Radium $^{226}_{88}Ra$, and Potassium $^{40}_{19}K$ for Tell Al-Hiba building materials samples. All measurements were within the internationally permissible range and according to the recommendations of organizations such as (ICRP) and (UNSCEAR).

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

Sample ID	$^{238}_{92}U$ (Bq/Kg)	$^{232}_{90}Th$ (Bq/Kg)	$^{226}_{88}Ra$ (Bq/Kg)	$^{40}_{19}K$ (Bq/Kg)
HS1	17.35	18.65	16.28	194.5
HS2	11.23	8.31	9.95	107.93
HS3	29.73	17.1	26.77	197.07
HS4	17.94	14.83	16.74	223.18
HS5	9.57	10.41	8.19	100.37
HS6	22.95	18.84	18.06	288.9
HS7	17.59	15.7	14.54	212.35
HS8	18.69	16.65	18.13	206.37
HS9	36.57	21.49	29.18	189.67
HS10	19.42	17.96	19.23	233.31
Average	20.1	15.99	17.71	195.37
STDEV	8.05	3.98	6.49	55.85
Wor.ave.[20]	35	30	35	400

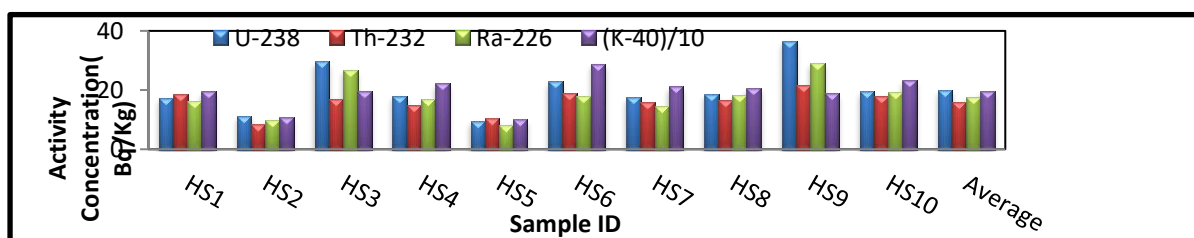


Figure.3. Radioactivity concentrations of $^{238}_{92}U$, $^{232}_{90}Th$, $^{226}_{88}Ra$, and $^{40}_{19}K$

For all samples.

Tables (5) and (6) includes the results of the calculations of absorbed gamma doses estimated in units of ($nGy \cdot h^{-1}$), 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 (14.12-

34.37)nGy. h^{-1} , with an average of (25.99 nGy. h^{-1}) . Fig. (4) Showing absorbed gamma dose as a function of sample ID. Using equation (2), the Radium equivalent radioactivity concentration ($^{226}_{88}Raeq$) values were calculated, which ranged between, (30.14-74.52) Bq/K g with an average of (55.62 Bq/Kg), Fig. (5) showing the ($^{226}_{88}Raeq$) as a function of sample ID. The OECD representative level index also calculated using the equation (6), which symbolized by the symbol (I_{yr}), where the values ranged between (0.221-0.536) Bq/Kg to an average (0.408Bq/Kg). Fig. (6) Showing Gamma-Ray representative level Index to the samples. Moreover, calculated the external hazard (H_{ex}) and internal hazard (H_{in}) indexes for the measured samples, and their values ranged between values were (0.081-0.201), (0.105-0.28) Bq/kg, averaging (0.150), (0.198) Bq/kg, respectively. Fig. (7) Showing external and internal hazard indexes. The table also includes the indoor and outdoor annual effective dose, which was calculated through equations (7) and (8), approved by (ICRP), where the Annual Effective Dose (AED) (indoor) values ranged between (0.069-0.168) mSv. y^{-1} with an average of (0.127 mSv. y^{-1}) . As for the Annual Effective Dose (AED) outdoor, ranged between (0.017-0.042 mSv. y^{-1}), with an average of (0.032 mSv. y^{-1}) . Fig. (8) Shows the annual effective dose (AED) (indoor and outdoor) of the samples, the table also included calculating the total lifetime excess cancer risk (ELCR) which was calculated from the relation (9), where its value ranged between (0.301×10^{-3} - 0.735×10^{-3}), with an average of (0.555×10^{-3}) Fig.(9) shows the variation in the total (ELCR).

Table 5. Calculated absorbed dose according to the recommendation of ICRP60, Equivalent concentrations of Radium, Representative level Index, in addition, the internal and external hazard indexes.

Sample ID	D_{ICRP} (nGy/h)	$^{226}_{88}Raeq$ (Bq/Kg)	I_{yr} (Bq/kg)	H_{in} (Bq/kg)	H_{ex} (Bq/kg)
HS1	26.89	57.93	0.201	0.156	0.215
HS2	14.12	30.14	0.108	0.081	0.100
HS3	30.91	66.39	0.252	0.179	0.177
HS4	25.99	55.13	0.194	0.149	0.171
HS5	14.26	30.8	0.105	0.083	0.206
HS6	31.77	67.25	0.23	0.182	0.142
HS7	25.06	53.34	0.183	0.144	0.205
HS8	27.04	57.83	0.205	0.156	0.231
HS9	34.37	74.52	0.28	0.201	0.182
HS10	29.46	62.88	0.222	0.169	0.208
Aver.	25.99	55.62	0.198	0.150	0.184
Word. Ave. [20]	55	350	≤ 1	≤ 1	≤ 1

Table 6. Represents annual effective dose (indoor and outdoor) and Excess Lifetime Cancer Risk (ELCR).

sample ID	AED (mSv/y)		ELCR $\times 10^{-3}$ Indoor	ELCR $\times 10^{-3}$ Outdoor	ELCR $\times 10^{-3}$ Total
	In*	Out*			
HS1	0.131	0.033	0.459	0.116	0.574
HS2	0.069	0.017	0.242	0.059	0.301
HS3	0.151	0.038	0.529	0.133	0.662
HS4	0.127	0.032	0.445	0.112	0.557
HS5	0.069	0.017	0.242	0.059	0.301
HS6	0.155	0.039	0.543	0.137	0.679
HS7	0.123	0.031	0.431	0.109	0.539
HS8	0.132	0.033	0.462	0.116	0.578
HS9	0.168	0.042	0.588	0.147	0.735
HS10	0.144	0.036	0.504	0.126	0.63
Aver.	0.1269	0.0318	0.444	0.111	0.555
STDEV	0.034	0.009	0.117	0.029	0.147
Word. Ave. [20]	≤ 1			0.29	

In* =indoor, Out* =outdoor

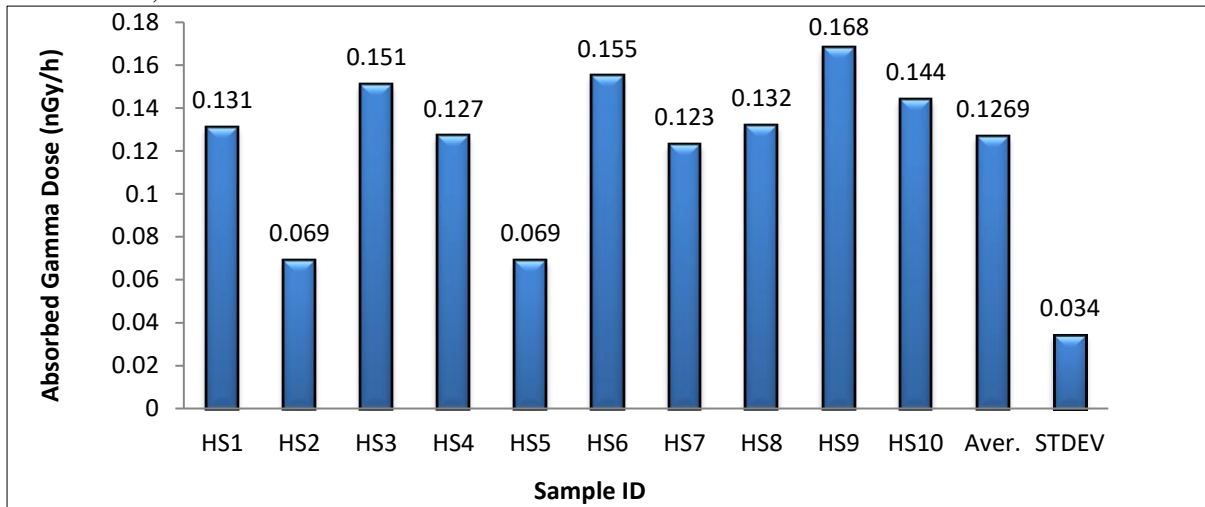


Figure 4. Absorbed Gamma Dose, for all samples.

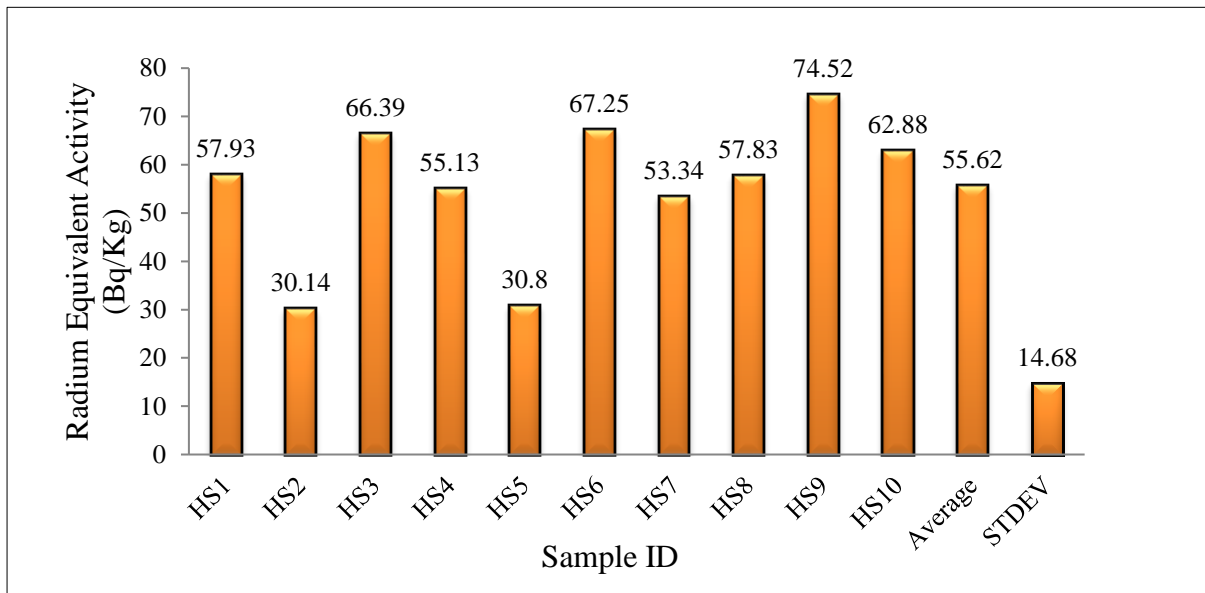


Figure 5. Equivalent radioactivity concentration of Radium-226 for all sample.

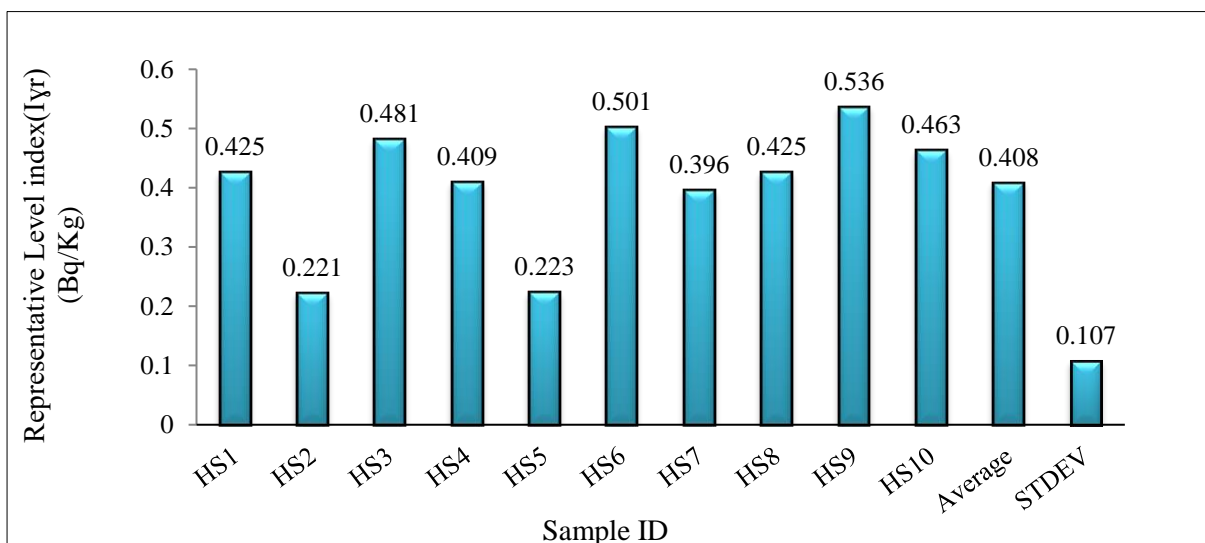


Figure 6. Gamma-Ray representative level Index for all Samples

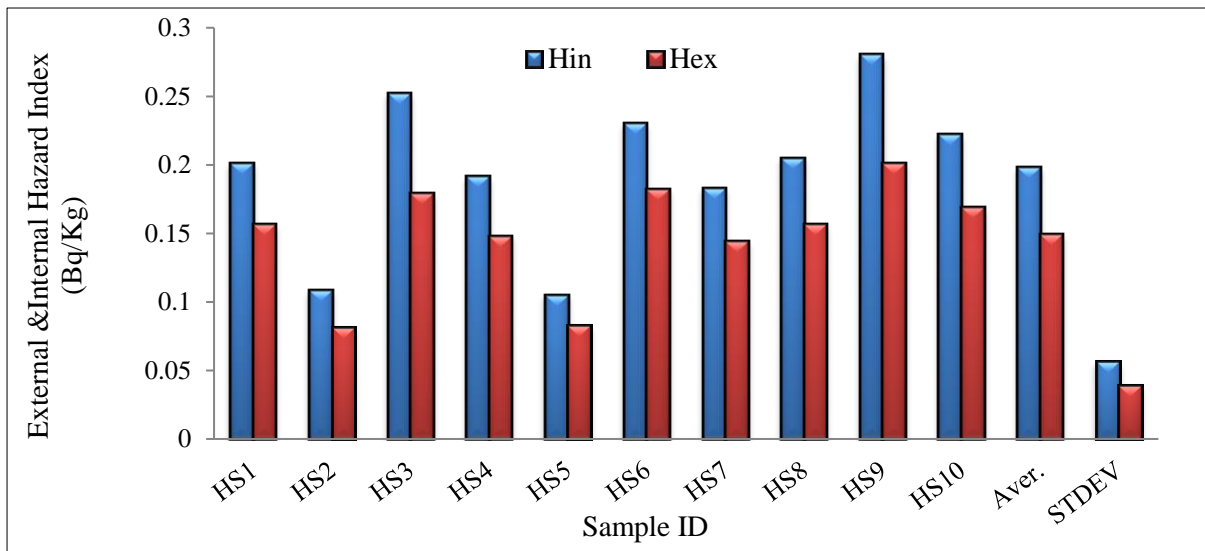


Figure 7. External (H_{ex}) and Internal (H_{in}) hazard indexes for all Samples.

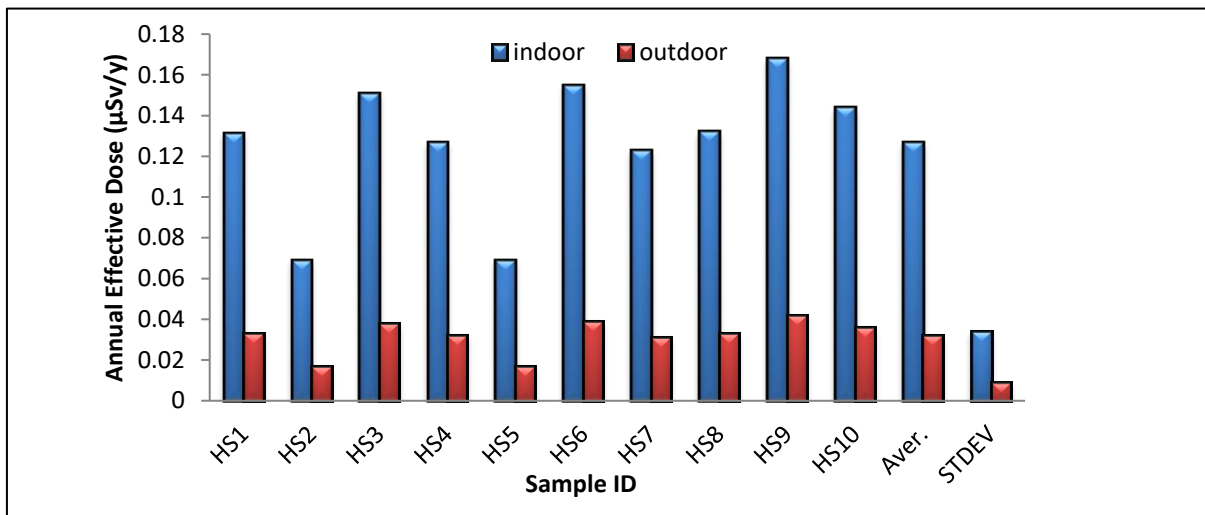


Figure 8. Annual equivalent dose (Indoor & Outdoor) for all samples.

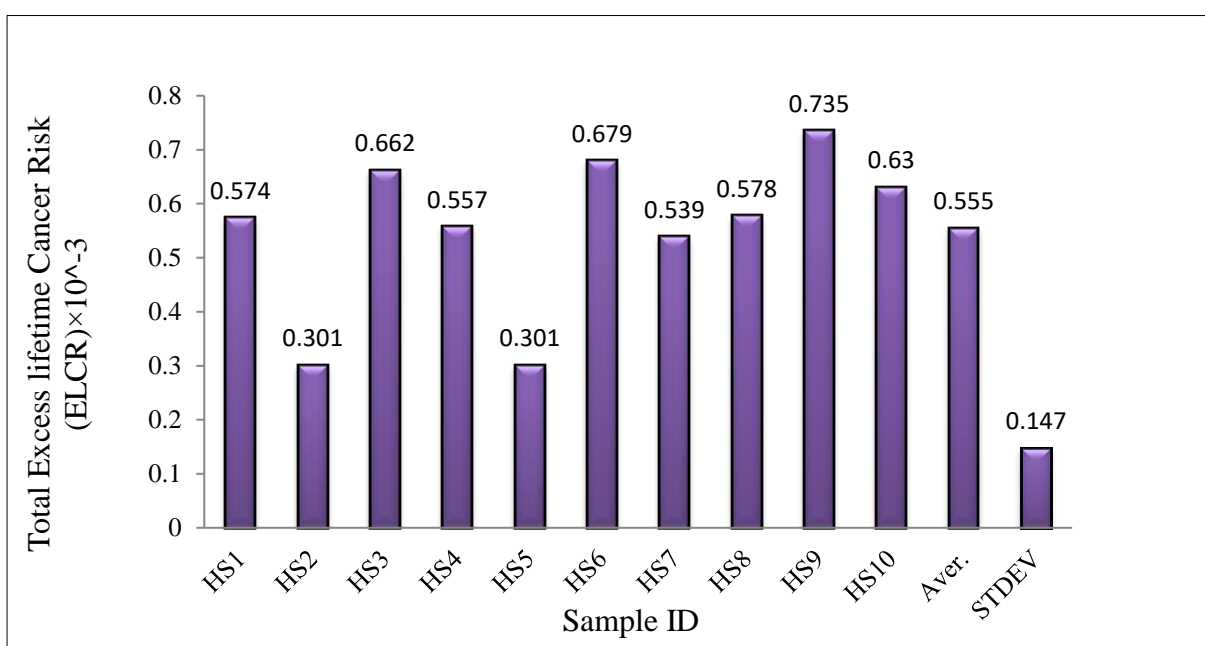


Figure 9. Total Excess Lifetime Cancer Risk (ELCR) for all samples.

5. Conclusion

According to the results of the current study, the levels of natural radioactivity in the construction materials used in the pre-third millennium BC archaeological site of Tell Al-Hiba are within the acceptable ranges as recommended by international organizations and agencies like the (ICRP) and (UNSCEAR). The results indicate that the environment in which this site was built is similar to the environment today. This indicates that the natural radiation contained at this site has not changed.

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