# ASSESSMENT OF NATURAL RADIOACTIVITY LEVELS AND RADIATION HAZARD IN ANCIENT HARAR CITY, JUGAL, SOIL, ETHIOPIA

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*Abstract*. The purpose of this work is to the assess the natural radioactivity levels and radiation hazard of ancient Harar city, Jugal. The soil samples were taken from ten different sites around the Jugal. Estimation of background radiation was carried out using HPGe gamma-ray spectrometry. The specific activity of $^{40}$K, $^{238}$U and $^{232}$Th in soil samples were obtained in the range between 254.64±11.614 Bq/kg to 330.83 ±14,73 Bq/kg, 12.84±0.952 Bq/kg to 30.22±2.135 Bq/kg and 34.25±2.47 Bq/kg to 124.91±8.622 Bq/kg, with the average value of 261.63±11.00 Bq/kg, 17.26±2.07 Bq/kg and 77.92±5.80 Bq/kg respectively.. The radium equivalent activity, absorbed dose rate, annual effective dose estimated ranged from 83.22 Bq/kg to 234.32 Bq/kg, 38.165 to 103.202 nGy/h, 0.187 to 0.506 mSv/y, respectively. The internal and external hazard indices, gamma index estimated ranged from 0.292 to 0.817 and 0.261 to 0.714, 0.225 to 0.633, respectively. The annual gonad dose equivalent 269.57 μSv/y to 719.38 μSv/y, respectively. The results of soil examined in the ancient city show no significant radiological hazard arise from the soil.

*Key words*: specific radioactivity, radionuclides, groundwater, well, radium equivalent, HPGe gamma-ray spectroscopy, radiological hazard.

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## INTRODUCTION

The natural radioactive material that are originated in the environment is the major source of radiation. They exposed all living organisms, which is due to the activity concentration of natural radionuclides: $^{238}$U series, $^{232}$Th decay series and the isotope of potassium ($^{40}$K) [17] that are found in the surface of Earth, in the rock, soil, building material, water, air, food and inside human body [28]. Natural radionuclide present in the soil contributes a measurable quantity of background radiation exposure to the population. Natural radioactivity from extra-terrestrial [5] sources and radioactive elements in the Earth's crust is a source of continuous radiation exposure to human beings [18]. The natural radioactivity present in the

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environment is the main source of radiation exposure for humans and constitutes the background radiation [9, 17].

Soil is the main basis for natural radionuclide, and it is the source of natural radiation hazard to population. The occurrence of natural radionuclide in soil results in internal and external radiation exposure to humans. The most commonly encountered radionuclides that irradiate the human body through external exposure (primarily by gamma radiation) are $^{238}$U and $^{232}$Th, and their subsequent radioactive decay products, and $^{40}$K [1, 15, 16, 22].

Natural radioactivity present in the environment comes mainly from primitive radionuclides in soils, and rocks which have long half-lives and therefore continue to stay for several years [19]. Since, 10% of the average annual radiation dose to human body comes from all other types of ionizing radiation [39].

Gamma radiation emitted from naturally occurring radionuclides represents the terrestrial background radiation and is the main sources of irradiation on the human body. The predominant part of the radio activity of soil and sediment devices from the decay of the primordial radionuclides $^{238}$U, $^{235}$U, $^{232}$Th, and their numerous decay products. $^{87}$Rb and $^{40}$K and also significant amount of manmade radio nuclides $^{137}$Cs and $^{90}$Sr may also be present in the soil.

Gamma radiations emitted as a result of radioactivity of the naturally occurring radioactive elements such as $^{40}$K and the primordial radionuclides series of $^{232}$Th and $^{238}$U in soil [35] (often called terrestrial gamma background radiations) make the biggest contribution to the total natural background radiation dose [43].

Therefore, the main purpose of this work is to assess the natural radioactivity levels and radiation hazard in ancient Harar city, around Jugal. The historic town of Harar is located in the eastern part of the country on a plateau with deep gorges surrounded by deserts and savannah. Jugal is the historic town that has retained its traditions, urban fabric and rich Harari Muslim cultural heritage to the present time. The city is surrounded by the wall which separated the ancient holy Islamic city from the modern Harar city. The walls were built between the 13$^{th}$ and 16$^{th}$ centuries [7, 39].

## MATERIALS AND METHODS

Harar city is astronomically it is located at 42.4° – 42.22 °E longitude and 9.15 – 9.27 °N latitude [40]. It has an average altitude of 1780 meter above sea level and average temperature of 22.65 °C.

According to [6], there are four types of soils found around Harar city. These are brownish soil, sand, gogoba (local name), kuyesa (local name). The relative

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density of gogoba, kuyesa, sandy and brownish are 2.62, 2.55, 2.61 and 2.62, respectively.

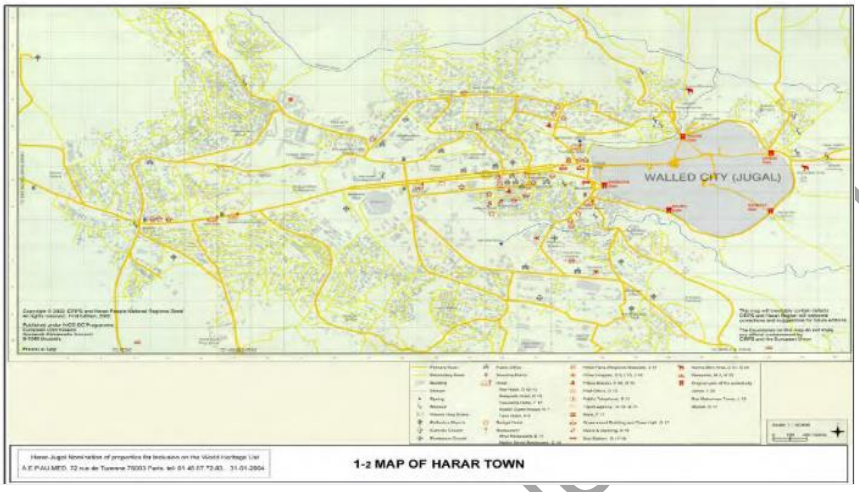


Fig. 1. Map of Harar sources [6, 29].

## SAMPLE PREPARATIONS

A total of ten soil samples were collected from different locations in the city of ancient Harar city around the Jugal. Each sample was taken from a depth of 0 – 20 cm at the chosen point [25], and the Global Positioning System (GPS) was used for tracking the data to record. The locations of four soil samples were chosen randomly from the city. All samples were collected from the surface and were marked by the number S1, S2, S3, S4, S5, S6, S7, S8, S9 and S10. The astronomical location of the sites is shown in Table 1.

*Table 1*

Coded samples and astronomical location of samples soil

| Sample code | Longitude E (°) | Latitude N (°) |
|---|---|---|
| S1 | 42.1370 | 9.3072 |
| S2 | 42.1356 | 9.3085 |
| S3 | 42.1337 | 9.3098 |
| S4 | 42.1365 | 9.3139 |
| S5 | 42.1357 | 9.3098 |
| S6 | 42.1357 | 9.3125 |
| S7 | 42.1357 | 9.3345 |
| S8 | 42.1345 | 9.3125 |
| S9 | 42.1335 | 9.3242 |

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| | | |
|---|---|---|
| S10 | 42.1257 | 9.3146 |

The samples were mixed and sieved with 2 mm mesh, then placed in an oven at 110 °C for 24 hours for the purpose of drying. Then the samples of 400 – 600 g weight were packed in Marinelli beaker, sealed with plastic tape to prevent the escape of airborne radionuclides and left for at least 4 weeks to ensure radioactive equilibrium between radon and its decay products [36].

The final sample preparation and all the gamma-ray spectrometry measurements were performed in consultation with the experts of radiation detection laboratory of Ethiopian Radiation Protection Authority.

## GAMMA-RAY SPECTROMETRY

The gamma spectrometric measurement was carried out using HPGe Gamma-ray spectrometric system at Ethiopian Radiation Protection Authority. The gamma spectrometry system was used for measuring gamma rays emitted from soil samples. The employed gamma spectrometer was an n-type coaxial Camberra high-purity germanium detector with a crystal diameter of 72.5 mm and a thickness of 72.5 mm (Prague, Czech Republic). Its detector has a relative efficiency of 70%, and energy resolution of 1.90 keV (FWHM) at 1333 keV and 1.05 keV (FWHM) at 122 keV a peak-to-Compton ratio of 70:1. This spectrometer is surrounded by a copper shield of 6 mm in thickness and a lead shield of 4 cm in thickness to reduce the background of gamma radiation to minimum [7].

The background radiation level of the laboratory environment was later subtracted from the measured γ-ray spectra of each sample [7]. At the end of the measurement, the region of interest, which was deducted from the background reading, was computed with a specialized template involving the energy, percentage error, count, uncertainty [7].

## SPECIFIC ACTIVITY MEASUREMENT

The specific activity is defined as activity per unite mass of radioactive substance and the reported in units such as Curie per gram or Becquerel per kilogram (Bq/kg). The specific activity of radionuclide concentrations, mainly $^{40}$K, $^{238}$U and $^{232}$Th were calculated using Eq (1) mentioned in reference [39] reported in units such as curie per gram or becquerel per kilogram (Bq/kg).

$$A = \frac{\frac{N_s}{t_s} - \frac{N_b}{t_b}}{\varepsilon(E_i) I_\gamma M_s} \quad (1)$$

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where $N_s$ is the net counts of the radionuclide in the samples, $N_b$ is the net counts of radionuclide in the background, $I_\gamma$ is gamma emission probability (gamma yield), $\varepsilon(E_i)$ is the peak efficiency of the detector at energy $E_i$ , $t_s$ Is sample counting time, $t_b$ is the background measuring time and $M_s$ is the mass of the sample (kg).

## RADIUM EQUIVALENT ACTIVITY

Radium equivalent ($Ra_{eq}$) is a weighted sum of the activities of $^{40}$K, $^{238}$U and $^{232}$Th and it is based on the assumption that 370 Bq/kg of $^{238}$U, 259 Bq/kg of $^{232}$Th, and 4810 Bq/kg of $^{40}$K produce the same gamma radiation dose rate [3, 42]. To avoid radiation hazards, materials, $Ra_{eq}$ is greater than 370 Bq/kg should not be used. $Ra_{eq}$ is defined by Eq (2) [10, 34, 41]

$$Ra_{eq} = C_U + 1.43\ C_{Th} + 0.077\ C_K \quad (2)$$

where $C_K$, $C_U$ and $C_{Th}$ are the specific activity of $^{40}$K, $^{238}$U and $^{232}$Th respectively, in Bq/kg [34].

## THE ABSORBED DOSE RATE IN AIR

Radiation emitted by a radioactive substance is absorbed by any material [10, 13, 23]. It encounters has given the dose conversion factors for converting the specific activity of $^{40}$K.$^{238}$U and $^{232}$Th into dose [40] (nGy/h per Bq/kg) [23] as 0.0414, 0.461 and 0.623, respectively [41].

$$AD = 0.461\ C_{Ra} + 0.623\ C_{Th} + 0.0414\ C_K \quad (3)$$

where $AD$ is the absorbed dose rate, 0.461, 0.623 and 0.0414 nGy/h / Bq/kg are the conversion factors of $^{226}$Ra, $^{232}$Th and $^{40}$K, respectively.

## ANNUAL EFFECTIVE DOSES EQUIVALENT

Annual estimated effective doses equivalent received by human being was calculated by 0.7 Sv/Gy, which was used to convert the absorbed dose are to human it was reported in [39], the effective dose equivalent with an outdoor 20% and 80% for indoor [10, 23, 35, 40] calculated using Eqs 4 and 5.

$$AED_{outdoor}\,(mSv/y) = AD\ (nGy/h) \times 8760\ h \times 0.2 \times 0.7\ Sv/Gy \times 10^{-6} \quad (4)$$

$$AED_{indoor}\,(mSv/y) = AD\ (nGy/h) \times 8760\ h \times 0.8 \times 0.7\ Sv/Gy \times 10^{-6} \quad (5)$$

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## RADIATION HAZARD INDICES

Most of the radioactive materials decay naturally and produces external radiation field which expose human being. Doses by ingestion are mainly due to $^{40}$K and to the $^{238}$U and $^{232}$Th series radionuclides present in food and drinking water other than in soil [9, 10, 12].

### Internal hazard index

The internal hazard index ($H_{in}$) gives the internal exposure to carcinogenic radon and its short-lived progeny [9, 10, 12]. To account for this threat the maximum permissible concentration for $^{226}$Ra must be reduced to half of the normal limit (185 Bq/kg) and it was determined using this equation [2, 9, 10, 12].

$$H_{\text{in}} = \frac{C_{\text{U}}}{185} + \frac{C_{\text{Th}}}{250} + \frac{C_{\text{K}}}{4810} \leq 1 \quad (6)$$

where $C_U$, $C_{Th}$ and $C_K$, are the specific radioactivity in Bq/kg of $^{226}$Ra, $^{232}$Th and $^{40}$K respectively. The value of this index must be less than unity for the radiation hazard to be negligible; $H_{ex}$ equal to unity corresponds to the upper limit of $Ra_{eq}$ (370 Bq/kg) [29].

### External Hazard Index

The external hazard index ($H_{ex}$) can be calculated by Eq. 7 [2, 10, 12].

$$H_{\text{ex}} = \frac{C_{\text{U}}}{370} + \frac{C_{\text{Th}}}{259} + \frac{C_{\text{K}}}{4810} \leq 1 \quad (7)$$

where $C_U$, $C_{Th}$ and $C_K$, are the specific radioactivity in Bq/kg of $^{238}$U, $^{232}$Th and $^{40}$K respectively. The value of this index must be less than unity for the radiation hazard to be negligible.

## ANNUAL GONAD EQUIVALENT DOSE (AGED)

The gonads, the bone marrow and the bone surface cells are considered as organs of interest by [41] because of their sensitivity to radiation. An increase in AGED has been known to affect the bone marrow, causing destruction of the red blood cells that are then replaced by white blood cells [8, 24]. It was calculated using the Eq (8).

$$AGED(\text{mSv/y}) = 3.09C_{\text{U}} + 4.18C_{\text{Th}} + 0.3144C_{\text{K}} \quad (8)$$

where $C_U$, $C_{Th}$ and $C_K$, are the $^{238}$U, $^{232}$Th and $^{40}$K specific activity (Bq/kg) in the soil samples, respectively.

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GAMMA INDEX

Gamma index ($I_\gamma$) was calculated using Eq (9). This is used to estimate the radiation hazard associated with the natural radionuclide in specific investigated samples [9]. Values of $I_\gamma$ correspond to an annual effective dose of less than or equal to 1 mSv [39].

$$I_\gamma = \frac{C_U}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \leq 1 \quad (9)$$

where $C_K$, $C_U$ and $C_{Th}$ are the $^{40}K$, $^{238}U$ and $^{232}Th$ specific activity (Bq/kg) in the soil samples, respectively.

## RESULTS AND DISCUSSION

The study was aimed at determination of assessment of natural radioactivity levels and radiation hazard in ancient Harar city, Jugal, has been determined using gamma-ray spectrometry. From soil samples different quantities were determined: the intensity, the activity concentration, the radium equivalent activity, the absorbed dose rate, the annual effective dose rate, gamma index, internal and external hazard indices, etc.

SPECIFIC ACTIVITY

The specific activity of $^{40}K$, $^{238}U$ and $^{232}Th$ were obtained from ten different soil samples from ancient Harar city, Jugal. The specific activity of K, U and Th range from 247.65±10.25 Bq/kg to 330.83±14.12 Bq/kg, 12.85+0.93 Bq/kg to 30.22±2.54 Bq/kg and 34.25±2.47 Bq/kg to 124.91±8.62 Bq/kg, respectively. The results shown in Table 2 revealed that the higher specific activity of $^{40}K$ was found in S2, the soil was brownish type, and the minimum was obtained in S5, sand soil type. Similarly, the highest and the lowest specific activity of $^{238}U$ were measured in S2 and S3 and the types of soils are different. The specific activity of $^{232}Th$ is the peak value 124.91±8.62 Bq/kg in S2 is site with the soil types of brownish, whereas, the minimum value 34.25±2.47 Bq/kg was obtained in S1 site with the soil types of brownish and the average specific activity value 77.918±5.799 Bq/kg.

The results shown in Table 2 are comparable to the worldwide average recommended by [41], which are 400, 30 and 35 Bq/kg for $^{40}K$, $^{238}U$ and $^{232}Th$ respectively [41]. It was found that all values of $^{232}Th$ specific activities are higher than the worldwide average whereas those of $^{238}U$ and $^{40}K$ are less than it.

The specific activity of $^{40}K$, $^{238}U$, and $^{232}Th$ radionuclides obtained in Harar were compared with the results of similar studies conducted in other countries of the world. A comparison of specific activity of radionuclides across the world is shown

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in Table 3. This helps predicting the source of contamination from one locality or country to the other. Table 3 revealed that the specific concentration of $^{232}$Th measured in the ancient Harar city, Jugal, is greater than the country shown in the Table 3. The specific activity for $^{40}$K, $^{238}$U and $^{232}$Th in soils of the study are generally low as compared to the values reported by [35].

*Table 2*

Coordinates and specific activity of $^{40}$K, $^{238}$U and $^{232}$Th of natural radionuclides in the soil samples from ancient Harar city, Jugal

| Sample code | Soil type | Specific activity (Bq/kg) | | |
|---|---|---|---|---|
| | | $^{40}$K | $^{238}$U | $^{232}$Th |
| S1 | Brownish | 270.89±12.28 | 13.38±1.01 | 34.25±2.47 |
| S2 | Brownish | 330.83±14.12 | 30.22±2.54 | 124.91±8.62 |
| S3 | Sand | 258.87±11.12 | 12.85+0.93 | 62.8±4.35 |
| S4 | Sand | 253.64±11.34 | 13.10±0.95 | 74.11±5.19 |
| S5 | Sand | 247.65±10.25 | 21.32±0.34 | 85.65±5.23 |
| S6 | Gogoba (local name) | 248.76±13.45 | 17.34±1.02 | 75.32±6.20 |
| S7 | Gogoba | 252.56±12.34 | 19.65±0.85 | 85.75±7.5 |
| S8 | Kuyesa (local name) | 254.43±11.76 | 15.43±0.78 | 75.52±4.5 |
| S9 | Kuyesa | 248.76±11.32 | 13.65±0.85 | 65.12±5.43 |
| S10 | Brownish | 249.87±11.05 | 15.67±0.54 | 95.75±8.5 |
| Max | | 330.83±14.12 | 30.22±2.54 | 124.91±8.62 |
| MIN | | 247.65±10.25 | 12.85+0.93 | 34.25±2.47 |
| Mean | | 261.63±11.904 | 17.26±2.066 | 77.92±5.799 |
| Std | | 25.265±1.17 | 5.384±3.87 | 23.508±1.94 |

*Table 3*

The specific activity values of $^{40}$K, $^{238}$U and $^{232}$Th of soil samples were obtained from literature review of different regions or countries on the world including the present study

| Region /Country | $^{40}$K (Bq/kg) | $^{238}$U (Bq/kg) | $^{232}$Th (Bq/kg) | Reference |
|---|---|---|---|---|
| Portugal | 840 | 49 | 51 | [41] |
| Iran | 640 | 28 | 22 | [41] |
| India | 400 | 29 | 64 | [41] |
| Denmark | 460 | 17 | 19 | [29] |
| Poland | 410 | 26 | 21 | [21] |
| Algeria | 370 | 30 | 25 | [41] |
| Nigeria | 210 | 18 | 22 | [32] |

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| | | | | |
|---|---|---|---|---|
| Egypt | 320 | 37 | 18 | [41] |
| Turkey | 744.8 | 48.4 | 20.5 | [33] |
| USA | 370 | 40 | 35 | [27] |
| Dire Dawa, Ethiopia | 716.59 | 19.97 | 56.38 | [9–10] |
| **Harar city, Jugal** | 278.81 | 17.36 | 74.02 | **Present study** |
| Worldwide average | 400 | 35 | 30 | [42] |

*Table 4*

Radium equivalent, absorbed dose rates, annual effective dose rates, hazard indices, gamma index and annual gonad equivalent dose for the soil samples in Harar city

| Site | $Ra_{eq}$ (Bq/kg) | $D$ (nGy/h) | $AED_{\text{indoor}}$ (mSv/y) | $AED_{\text{outdoor}}$ (mSv/y) | $I_\gamma$ | $H_{\text{in}}$ | $H_{\text{ex}}$ | $AGED$ (μSv/y) |
|---|---|---|---|---|---|---|---|---|
| S1 | 83.216 | 38.165 | 0.187 | 0.046 | 0.298 | 0.261 | 0.225 | 269.569 |
| S2 | 234.315 | 103.203 | 0.506 | 0.125 | 0.817 | 0.714 | 0.632 | 719.384 |
| S3 | 122.587 | 54.663 | 0.268 | 0.066 | 0.435 | 0.366 | 0.331 | 383.496 |
| S4 | 138.608 | 61.391 | 0.301 | 0.075 | 0.490 | 0.410 | 0.374 | 429.902 |
| S5 | 162.869 | 71.910 | 0.353 | 0.087 | 0.568 | 0.497 | 0.440 | 501.658 |
| S6 | 144.202 | 63.878 | 0.313 | 0.078 | 0.506 | 0.436 | 0.389 | 446.529 |
| S7 | 161.720 | 71.403 | 0.350 | 0.087 | 0.566 | 0.490 | 0.437 | 498.457 |
| S8 | 143.014 | 63.352 | 0.311 | 0.077 | 0.504 | 0.428 | 0.386 | 443.243 |
| S9 | 125.926 | 56.012 | 0.275 | 0.068 | 0.445 | 0.377 | 0.340 | 392.490 |
| S10 | 171.832 | 75.492 | 0.370 | 0.092 | 0.604 | 0.506 | 0.464 | 527.114 |
| Max | 234.315 | 103.202 | 0.506 | 0.125 | 0.817 | 0.714 | 0.633 | 719.384 |
| Min | 83.2160 | 38.1647 | 0.187 | 0.046 | 0.298 | 0.261 | 0.225 | 269.569 |
| Average | 148.829 | 65.947 | 0.324 | 0.080 | 0.523 | 0.449 | 0.402 | 461.184 |
| Std | 39.269 | 16.912 | 0.083 | 0.020 | 0.135 | 0.119 | 0.106 | 116.917 |
| World wide average | ≤370 | 78 | 0.48 | | ≤1 | ≤1 | ≤1 | 300 |

The radium equivalent activity ($Ra_{eq}$) in the ancient city soil samples shown in Table 4. ranges from 83.216 Bq/kg (S1) to 234.315 Bq/kg (S2) with mean value of 148.83 Bq/kg which is less than the safe limit (370 Bq/kg) recommended by [31, 41].

The absorbed dose rate ($D$) and the annual effective dose rate of the soil samples collected from the ancient city of Harar are shown in Table 4, 3rd and 4th columns. The International Commission on Radiological Protection (ICRP) has recommended the annual effective dose equivalent limit of 1 mSv/y for the populations and 20 mSv/y for the radiation workers mentioned in reference [20].

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From Table 4, 3rd column, the absorbed dose rate indoor ranged from 38.165 nGy/h to 103.203 nGy/h with an average value of 65.947 nGy/h. Table 4 shows that the absorbed dose rate in site S2 was higher than the value shown in reference [36].

The annual effective doses rate of indoor and outdoor from 0.187 mSv/y to 0.506 mSv/y and 0.046 mSv/y to 0.125 mSv/y with an average value of 0.324 mSv/y and 0.080 mSv/y, respectively. The results revealed that the annual effective dose rate in S2 site is greater than average world value of effective dose for the indoor is 0.48 mSv/y. The internal and external hazard values of indices for the soil samples were range from 0.261 to 0.764 and from 0.255 to 0.633, with the average value of 0.449 and 0.402 respectively. Table 4 shows that for both internal and external hazard indices values were lower than the international limit at each soil collection site mentioned in reference [41]. Therefore, according to the report of European Commission in Radiation Protection, the area under study is safe for life and the residences do not expose on significant radiological hazard according to [16].

The radiation gamma index for the soil samples ranges from 0.298 to 0.817 with the average value of 0.523 which is less than the international limit mentioned in reference [41]. The result revealed that the soil in the city was safe.

Moreover, Table 4 shows the annual gonadal dose equivalent. The obtained results range from 269.569 μSv/y to 719.384 μSv/y with the average value of 461.184 μSv/y. The mean value of the annual gonad dose is higher than the average international limit but less than the mean value of soil samples reported in reference [9] and greater than the mean value of soil samples reported in reference [26].

Moreover, the hazard indices less than unity indicate no significant radiological hazard for the soil samples in the study area. The average value of the AGED was found to be 461.184 μSv/y, which is much higher than the world average value of 298 μSv/y reported in reference [31].

## CONCLUSIONS

The main purpose of this work is the assessment of natural radioactivity levels $^{40}K$, $^{238}U$ and $^{232}Th$ and radiation hazard in ancient Harar city, Jugal, soil samples, Ethiopia. The specific activity of soils were determined using gamma-ray spectrometry. The specific activity of $^{232}Th$ measured in soil was the highest measured value compared to other countries, as shown in Table 3. The radium equivalent specific activity concentration was measured, and it was less than the worldwide average limit. Effective doses rate and annual gonadal dose equivalent were collected from the soil samples of the Harar city. The results obtained were in the limit of worldwide recommended values.

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