

Radiological Assessment of Natural Radioactivity Levels in Selected Ceramic Tile Brands used in Kenya

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ABSTRACT

The assessment of the radiation levels of the natural radionuclides ^{232}Th , ^{238}U and ^{40}K in ceramic tiles used in Kenya has been studied. The radiometric analysis was done using a high-efficiency gamma-ray spectrometer. A total of thirty-seven (37) samples of ceramic tiles were pulverized, weighed accurately, and packaged in 250ml standard plastic containers, well labelled and stored for 28 days to attain secular equilibrium. Analysis of specific gamma energies from activities of ^{232}Th , ^{238}U and ^{40}K reported activity of $53.73 \pm 2.34 \text{ Bqkg}^{-1}$, $43.17 \pm 3.40 \text{ Bqkg}^{-1}$, and $525.99 \pm 36.10 \text{ Bqkg}^{-1}$ respectively. The variations in the radiation activity concentration is attributed to the composition of geological raw material used for tiles manufacturing. The average radium equivalent, absorbed dose, indoor and outdoor annual effective dose, and hazard indices (internal and external) obtained were 159.59 BqKg^{-1} , 75.55 nGyh^{-1} , 0.28, 0.19, 0.54 and 0.43 respectively. The results obtained from the study showed that the use of ceramic tiles in Kenya does not pose any significant harmful radiation effects to users.

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1. INTRODUCTION

All living organisms are exposed to ionising radiation of both natural and/or artificial origin. The natural sources of ionizing include cosmic, terrestrial and radon which are major contributors to cumulative radiation exposure. Since ceramic tile raw materials originate from terrestrial sources, they contain natural radionuclides ^{232}Th , ^{238}U , and ^{40}K which are major sources of natural radiations[1]. Studies on natural radioactivity assert that radionuclides whose half-life are in the order of the age of the earth are still present at significant levels in some geological profiles and lithological units[2]. Radiometric surveys reveal that over 81% of the annual radiation dose received by the human population comes from natural radiation sources with 18% of the total annual effective doses emanating from building materials such as rocks and soils[3, 4]. Studies further associate higher risks of radiation exposure from construction materials such as tiles, building blocks, sand, and cement for the human population which spend about 80% of their time indoors [5, 6]. The non-uniform distribution of terrestrial radionuclides in various environmental media makes it prudent to establish levels of radiation to help minimize human exposure to hazardous radiation as much as possible. Determination

of potential dangers of radiation is an integral issue in human and environmental protection. According to local standards and the European Commission, the dangers of exposure to harmful radiation should be minimized as much as possible [7]. Relatively longer indoor occupancy timelines coupled with a variety of materials used in construction may cumulatively enhance indoor radiation levels [1].

Building materials such terrazzo, and ceramic tiles are manufactured from soils and igneous rocks. Though radiological surveys report elevated levels of radioactivity in granitic igneous rocks over other forms of rocks like sedimentary and metamorphic, granitic rocks like quartz, kaolin and feldspar have been preferred for use in the manufacture of tiles due to their desirable characteristics; hardness and ability to resist abrasion hence giving them a brilliant finish for ornamental and interior works [8]. Ceramic tiles manufactured from rocks with elevated levels of terrestrial radionuclides are likely to have elevated radiations doses [9]. The exposure of these radiations may be as a result of inhalation and swallowing of the radionuclides or external exposure when gamma radiations from ^{232}Th , ^{238}U , and ^{40}K radionuclides present in building materials irradiate the human skin [10]. Though radiation-related anomalies are stochastic, consistent research shows that natural radiation doses exceeding 1mSv per year may result in harmful cellular effects like mutation, apoptosis, and deoxyribonucleic acid(DNA) damage whose repair mechanisms may be prone to errors [11]. Therefore, radiation exposure should be maintained as low as possible to enhance the safety of living organisms. This work, therefore, sought to determine the activity concentration and other radiological parameters of the natural radionuclides in selected ceramic tiles used in Kenya using gamma ray spectroscopy to establish their safety to users.

2. RESEARCH METHODS

2.1 Sampling and sample preparation

The systematic random sampling method was used to sample thirty-seven (37) samples from various local ceramic tiles outlets. Sixteen (16) tile samples were manufactured locally while twenty-one (21) were imported from Tanzania, Uganda, China, and India. The tile samples were milled into a fine powder to maximize filling capacity and sieved using a 1.0mm sieve for homogeneity. Separately, the samples were dried in a temperature-controlled furnace at 120^oc for 24 hours to get rid of any moisture for direct radionuclide identity. To maintain counting geometry, a mass of 203 g±0.05% for each sample equivalent to the mass of the reference sample was accurately transferred to an airtight, radon impermeable standard container and stored for 28 days for secular equilibrium to be attained before actual data taking.

2.2 Gamma-ray spectroscopy

Thallium-doped sodium iodide detector model PMT 905-4 - (NaI (TI), with a counting crystal measuring 3 × 3 cm and sufficiently shielded in a 10 cm thick shield lined with cadmium and copper of thickness 1.5 and 0.8 mm respectively was used in this work. The detector energy was calibrated using a multi-nuclide radiation source consisting of Americium (^{241}Am), Cesium (^{137}Cs) and Cobalt (^{60}Co) certified by the International Atomic Energy Agency(IAEA) [12]. This calibration certificate was preferred due to its energy range of 60 keV to 1400 keV covering the energy range of radionuclides of interest. The spectrometer photopeak counting efficiency (CE) was determined using equation 1.

$$\eta = \frac{N}{A \cdot \rho \cdot m \cdot t} \quad (1)$$

Where; η is the photopeak counting efficiency at a specific gamma line, N -is the net activity count at the full energy peak, A , ρ , m and t are the activities of ^{238}U , ^{232}Th and ^{40}K of the standard sample in Bacquels, standard gamma decay probabilities [13], mass (kg) of the sample and the counting live time (s) respectively. To optimize the quality of the data, background radiation measurements were determined using deionized water under similar experimental conditions as those of the samples. The net count rate (C_n) for each sample was therefore obtained as the difference between the gross count rate given by the tile sample spectrum and that from the background spectrum as shown in equation 2.

$$C_n = C_g - C_b \quad (2)$$

Where C_n is the net count rate from the sample, C_g is the gross count rate and C_b is the count due to background emissions.

2.3 Calculation of radiometric parameters.

Different Radiometric parameters were calculated as follows:

The activity concentration in BqKg^{-1} was calculated using equation 3 [14].

$$C = \frac{N_p}{p \cdot \eta \cdot m} \quad (3)$$

Where, N_p is the net count rate, p is the gamma-ray yield or emission probability, η (E) is the absolute counting efficiency of the detector while m is the sample mass in (kg).

Radium equivalent is used to assess the radiation hazard as a single quantity for activities of ^{226}Ra , ^{232}Th , and ^{40}K was calculated using equation 4 [15].

$$Ra_{eq} = C_{Ra} + 1.423C_{Th} + 0.077C_K \quad (4)$$

Where, C_{Ra} , C_{Th} and C_K are the average activity concentration of ^{238}U , ^{232}Th and ^{40}K in tile powder samples respectively given in BqKg^{-1} .

The absorbed gamma radiation dose rates were calculated from the corresponding activity concentration of ^{226}Ra (^{238}Th), ^{232}Th and ^{40}K using equation 5 [3].

$$D = 0.427C_{Ra} + 0.662C_{Th} + 0.043C_K \quad (5)$$

Where, C_{Ra} , C_{Th} and C_K are the average activity concentration of ^{238}U , ^{232}Th and ^{40}K respectively.

The annual effective dose rate (AEDR) was calculated using equations 6 and 7 for the indoor and outdoor annual effective doses rates respectively. Depending on the time spent indoors or outdoors, an occupancy factor of 0.6 indoors and 0.4 outdoors was used [16].

$$E_{in} = D \times 8760 \text{hy}^{-1} \times 0.6 \times 0.7 \text{SvGy}^{-1} \times 10^{-6} \quad (6)$$

$$E_{out} = D \times 8760 \text{hy}^{-1} \times 0.4 \times 0.7 \text{SvGy}^{-1} \times 10^{-6} \quad (7)$$

Where E_{in} and E_{out} are AEDR for indoor and outdoor environments respectively, $D(\text{nGyh}^{-1})$ is the absorbed dose in air, 8760hy^{-1} is the number of hours in one year, $0.7(\text{SvGy}^{-1})$ converts the absorbed dose in the air to annual effective dose [6].

The external and internal hazard indices were determined using the equations 8 and 9, respectively.

$$H_{EX} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (8)$$

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (9)$$

Where, C_{Ra} , C_{Th} and C_K are the mean activity concentrations in BqKg^{-1} of ^{226}Ra , ^{232}Th and ^{40}K respectively. The value of these indices should be below a unit for the radiation to be termed safe to humans [15, 17].

3. RESULTS AND DISCUSSIONS

In this study, various radiometric parameters including activity concentration, radium equivalent, absorbed dose, annual effective dose and hazard indices were evaluated and results are discussed in the following subsections.

3.1 Activity concentration (C)

The activity concentration for the natural radionuclides ^{238}U , ^{232}Th and ^{40}K was calculated using equation 3 for all the 37 ceramic tile samples and the numerical results are presented in table 1 and graphically in figure 1a and 1b.

Table 1: Activity concentration of ^{238}U , ^{232}Th and ^{40}K .

Sample code	Activity concentration in BqKg^{-1}			
	^{232}Th	^{238}U	^{40}K	
1	CV001W	116.21±3.23	60.8±6.52	667.18±49.59
2	CV002W	93.53±3.07	39.93±5.91	623.95±47.27
3	CV003W	112.42±2.98	60.34±5.77	727.12±45.73
4	CB001W	101.96±2.66	71.23±3.20	124.5±28.83
5	CB002W	63.07±1.54	45.42±2.61	415.92±31.14

6	IX001W	98.06±3.10	30.39±2.24	61.25±31.22
7	IX002W	100.98±2.87	52.21±5.86	446.74±47.82
8	KS001W	45.97±2.28	48.21±4.74	880.58±45.32
9	KS002W	48.88±2.11	48.04±4.42	360.46±30.76
10	KSO01F	48.52±2.77	4.01±1.42	810.47±36.66
11	KS002F	46.49±1.59	47.74±2.64	786.17±35.64
12	KS003F	51.31±1.80	40.56±2.63	506.29±31.44
13	KS004F	55.53±1.82	34.68±2.30	534.3±32.09
14	KS005F	63.03±1.67	39.5±3.23	1533.28±31.95
15	KSJ001W	50.28±2.12	83.42±3.31	634.52±31.81
16	KSJ002W	36.78±1.47	62.11±2.67	432.5±31.98
17	KT001W	29.75±1.36	36.69±2.63	308.8±30.09
18	KT002W	40.32±2.42	25.78±5.27	667.63±42.92
19	KT003W	37.52±1.51	37.21±2.57	345.41±31.82
20	KT004F	52.99±2.19	0.65±0.12	365.28±30.39
21	KT005W	37.52±1.51	37.21±2.57	367.75±31.82
22	KBL001F	52.65±2.60	37.14±5.04	1051.81±45.42
23	KBL002F	49.60±2.57	57.39±3.85	1013.41±44.46
24	UG001F	78.58±3.20	85.66±3.16	544.18±35.67
25	UG002F	77.31±3.28	76.02±3.61	332.84±32.54
26	UG003F	34.00±2.48	110.21±3.40	532.63±34.58
27	UG004F	34.12±4.38	124.79±4.04	272.09±32.09
28	UG005F	5.71±1.20	78.53±7.07	347.83±33.79
29	TG001W	28.73±1.39	18.81±2.24	753.97±27.35
30	TG002W	37.32±2.17	34.28±3.35	271.29±41.33
31	TG001F	47.01±2.37	6.09±2.06	867.68±43.22
32	TG002F	36.80±2.21	6.28±1.67	757.98±44.17
33	TG003F	38.92±2.20	12.18±1.88	757.61±42.14
34	TS001F	41.92±2.03	1.02±0.92	59.23±11.94
35	TS002F	23.10±1.20	14.07±2.25	121.43±31.85
36	TS003F	31.34±1.38	20.06±2.22	103.36±31.32
37	TS004F	39.66±2.01	8.70±1.64	74.04±28.60

AVERAGE	53.73±1.53	43.17±2.19	525.99±6.01
World Average [18]	45	33	420

The activity concentration of the radionuclides in the analyzed samples vary from sample to sample with ²³⁸U having the lowest value of 0.65±0.12 BqKg⁻¹ and highest value of 124.79±4.04 BqKg⁻¹, ²³²Th has the lowest value of 5.71±1.2 BqKg⁻¹ and highest of 116.21±3.23 BqKg⁻¹ and ⁴⁰K has the lowest value of 59.23±11.94 BqKg⁻¹ and the highest of 1533.28±31.95 BqKg⁻¹.

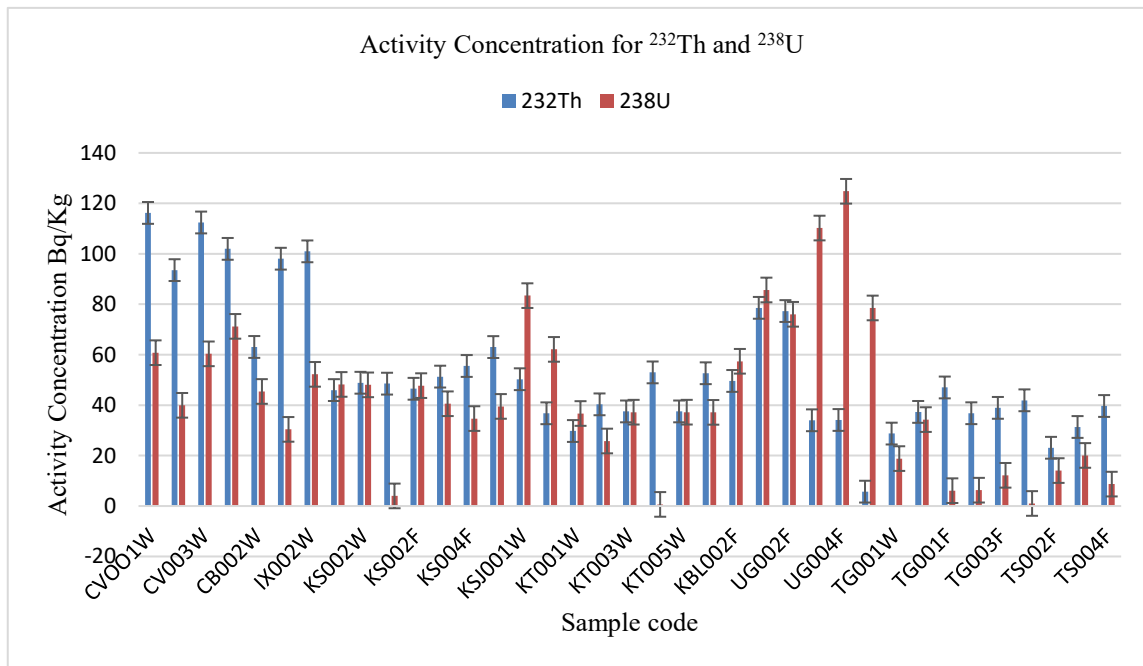


Figure 1a: Activity concentration in BqKg⁻¹ for ²³²Th and ²³⁸U.

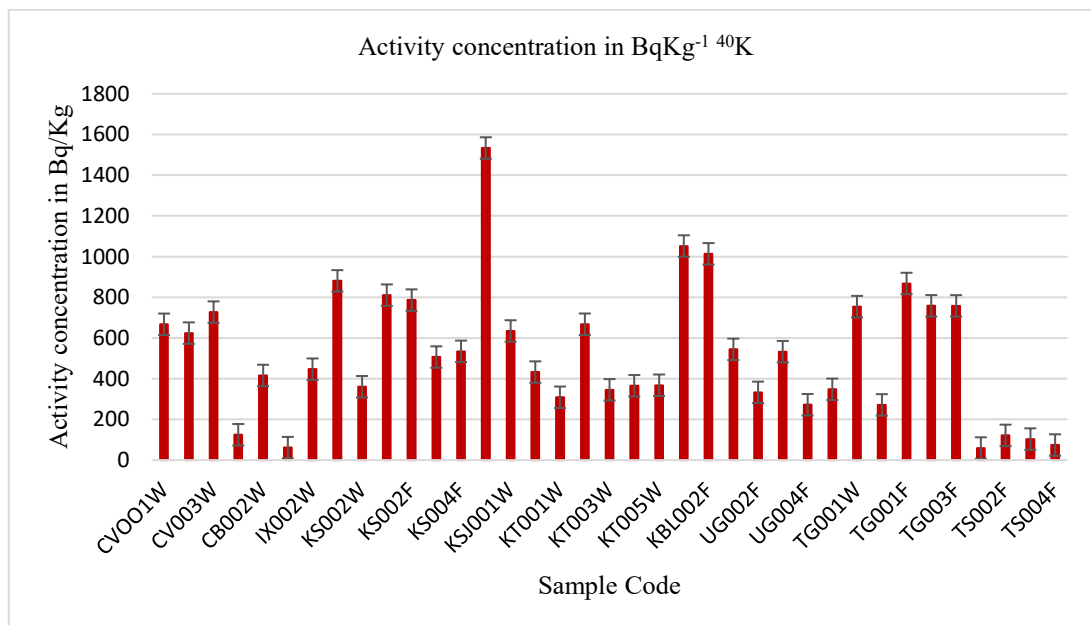


Figure 1b: Activity concentration in BqKg⁻¹ for ⁴⁰K.

The average activity concentration of the tile samples analyzed in this study are 53.73 ±2.34 BqKg⁻¹ for, 43.17±3.40 BqKg⁻¹ for and 525.99±36.10 BqKg⁻¹ for, ²³²Th, ²³⁸U and ⁴⁰K, respectively. These averages are slightly above the world averages 45 Bqkg⁻¹, 33 BqKg⁻¹ and 420 BqKg⁻¹ for ²³²Th, ²³⁸U and ⁴⁰K respectively

[18]. The variation of the activity concentration is observed also from the averages of the samples analyzed from different countries as indicated in table 2 for different radionuclides.

Table 2: Average activity concentration of the ²³⁸U, ²³²Th and ⁴⁰K for tiles from the different countries.

Country of origin	²³⁸ U	²³² Th	⁴⁰ K
Kenya	40.02±4.1	46.69±3.14	662.4±42
Tanzania	13.50±2.5	30.09±2.5	418.±35.7
Uganda	95.04±4.2	45.94±3.6	405.9±33
China	55.54±4.8	97.44±2.7	511.7±40
India	41.30±4.0	99.52±2.9	253.9±39

The findings shows that the region geology plays a great role in the kind and levels of radionuclides present in certain place [19, 20]. There is a considerable level of the primordial radioisotopes, ²³²Th, ²³⁸U and ⁴⁰K, in the raw materials from which the sample tiles are manufactured from. Studies show that radioactivity of a given soil relates to the rock type with soils originating from the igneous rocks giving higher radiations and sedimentary rocks lower values [3, 21, 22]. The values observed here are therefore characteristic of the meta-igneous rocks present in all regions within the Mozambique belt of which Kenya, Uganda, and Tanzania are part of. For instance the sixteen samples manufactured in Kenya, are from the coast and south eastern parts of the country which are rich in crystalline lime stones, quartzite, gneisses granulite's and potash soils which contain high levels of natural radioactivity [23]. The highest registered activity concentration value of ⁴⁰K was 1533 BqKg⁻¹ posted by a Kenyan tile sample (KS005F). This is attributed to the high level of potassium, carbonates and monazites in this region [24, 25]

Considering ²³²Th and ²³⁸U, tiles from Tanzania posted the lowest values for activity concentration. This agrees with a study done in Tanzania that listed the major soil types in the country as ferric, chromic and eutric cambisols which contain very little thorium and uranium [22]. According to Graef [21], potassium was found to be limited in surface soil and only presently abundant between 50 and 300 meters underground. This relates positively with the low activity concentration of the three radio nuclides in Tanzania tile samples.

3.2 Radium equivalent (R_{eq})

The radium equivalent R_{eq} is used to assess the radiological risk associated with exposure to ionizing radiation. Any building material with radium equivalent more than 370 BqKg⁻¹ is considered radio logically hazardous and therefore should not be used [26]. Equation 5 was used to estimate the radium equivalent, and the results presented in figure 2.

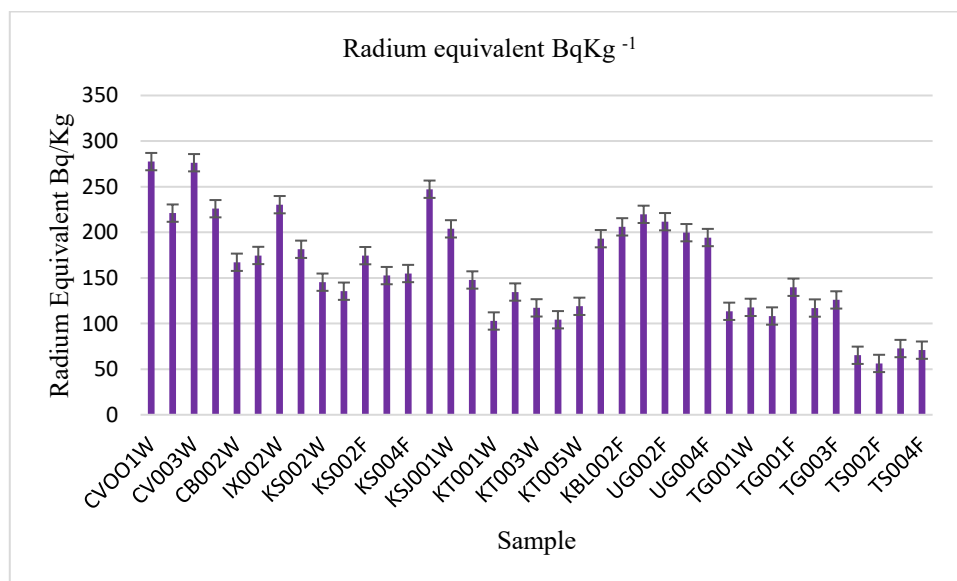


Figure 2: Radium equivalent for analyzed samples.

The average value of radium equivalent in this study was found to be 159.59 BqKg^{-1} which was lower than the safety limit of 370 BqKg^{-1} [26]. The highest value of radium equivalent ($277.53 \text{ Bq Kg}^{-1}$) was for a sample (CV001W) followed by sample CV003W with 276.31 BqKg^{-1} . Tiles from Tanzania posted radium equivalent values generally lower than all the other countries with the lowest value being 56.29 BqKg^{-1} for floor tile sample (TS002F). This observation was attributed to lower enrichments of ^{238}U , ^{232}Th and ^{40}K in the rock ore used during production. Nonhomogeneous distribution of radio nuclides in the environmental mediums could have led to the lower radium in this sample [21, 22]

3.3 Absorbed dose (D)

The absorbed dose was computed from the corresponding activity concentration of ^{232}Th , ^{238}U and ^{40}K using equation 5. The absorbed dose results are presented in figure 3. The mean value of the absorbed dose posted by this study was 75.55 nGy/h which was slightly above the world's average level of 60 nGy/h but lower than the permissible safety limit of $1500 \pm 75 \text{ nGy/h}$ [6].

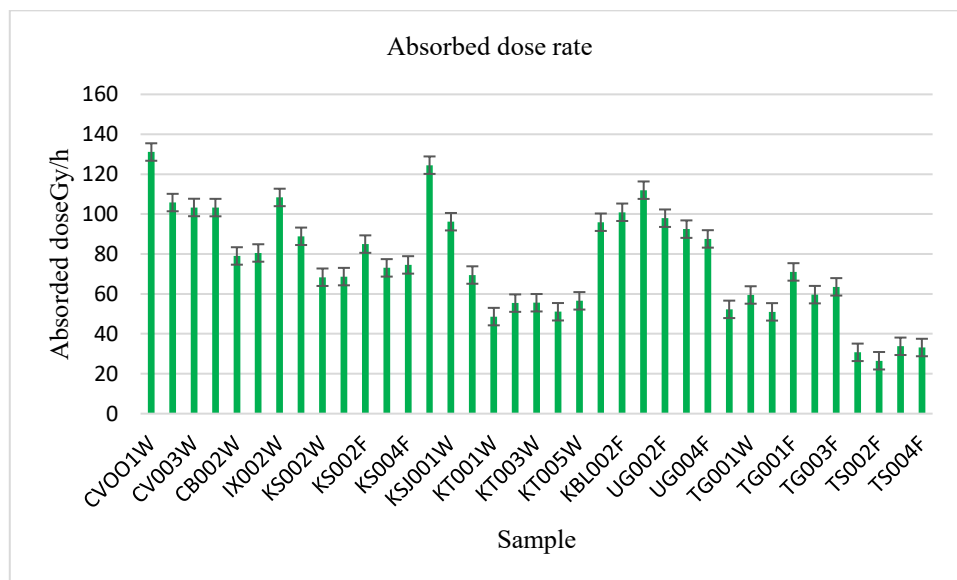


Figure 3: Absorbed dose for the samples analyzed.

3.4 Annual Effective Dose Rate (AEDR)

The radiation dose absorbed by an individual in one year was estimated from the absorbed dose (D) considering a 60% indoor occupancy time and a conversion factor of 0.7 [27]. Equations 6 and 7 were used for the estimate calculations for both indoor and outdoor and resulted presented in figure 4.

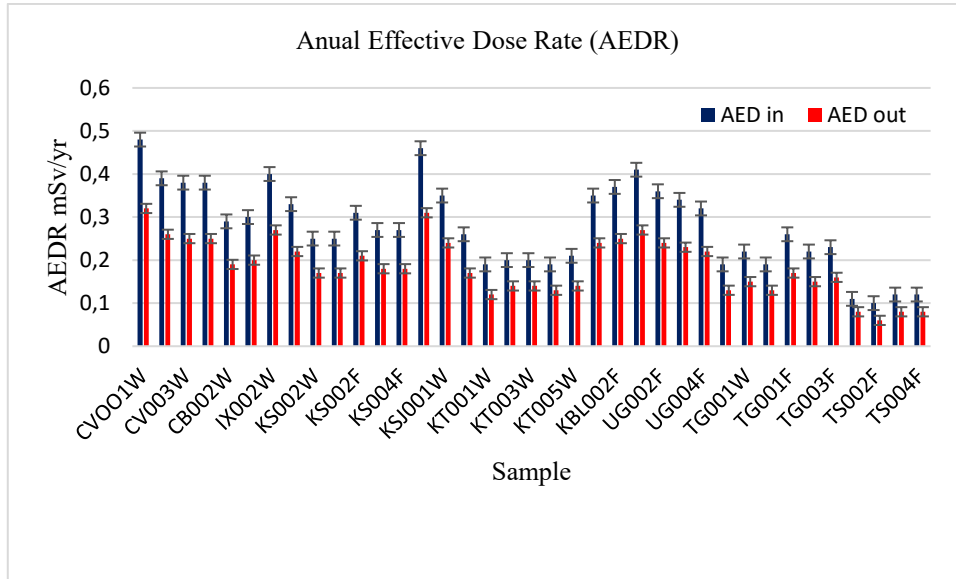


Figure 4: Annual effective dose indoor (red) and outdoor (blue).

The mean values from this study were 0.28 and 0.19 mSv⁻¹ for indoor and outdoor annual effective doses respectively which are consistent with many similar studies and bellow the recommended safety limits [28].

3.5 Hazard indices.

The internal hazard index is the measure of radiation exposure to an individual from radioactive substances within the body while the external hazard index is the measure of the radiation exposure to an individual from radioactive substances outside the body. Equation 8 and 9 have been used to estimate the internal hazard index from the sampled tiles and the results are presented in figure 5.

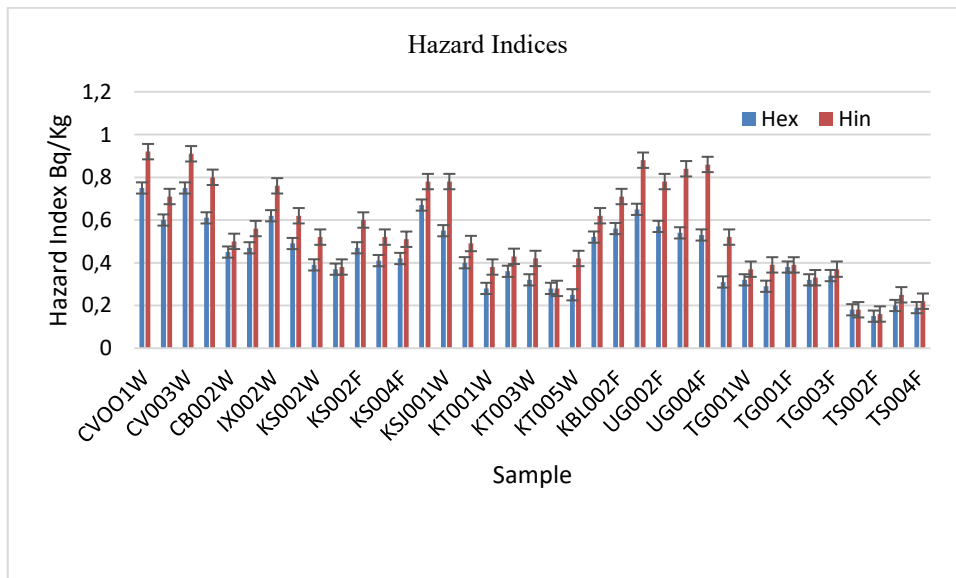


Figure 5: Internal (red) and external (blue) hazard indices.

The highest value of the internal hazard index was 0.92 while the lowest was 0.16. The largest value of the external hazard index was 0.75 and the lowest was 0.15. All the samples used in this study gave indices lower than a unit and therefore pose no health risk to human beings.

4. CONCLUSION

Measurement of the natural radioactivity in ceramic tiles used in Kenya was done for 37 tile samples using gamma-ray spectrometry. The mean activity concentration obtained was 53.73 ± 2.34 Bqkg⁻¹ for ²³²Th, 43.17 ± 3.40 Bqkg⁻¹ for ²³⁸U and 525.99 ± 36.10 Bqkg⁻¹ for ⁴⁰K. The findings on radioactivity agree well with spatial natural abundances of the radionuclides ²³⁸U > ²³²Th > ⁴⁰K. The dosimetric parametric values from activity concentrations including radium equivalent, absorbed dose, indoor annual effective dose, and internal hazard indices (internal and external) were 159.59 BqKg⁻¹, 75.55 nGyh⁻¹, 0.28, 0.54 and 0.43 respectively. The range of values for various dosimetric quantities compares well with other similar research and is lower than the levels recommended by ICRP and UNSCEAR reports. From the radiological viewpoint, the ceramic building tiles used in Kenya have no potential harm to the users.

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