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Assessment of radioactivity and radiological hazards in commercial ceramic tiles used in Ife-Central, local government area of Osun State, Nigeria

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ABSTRACT

The activity concentrations of the natural radionuclides (^{40}K , ^{232}Th and ^{238}U) in ceramic tiles use in homes and offices in Ile-Ife were investigated using a NaI (Tl) detector as the detecting device for gamma spectrometry. The gamma absorbed dose rate (D_R), radium equivalent activity (Ra_{eq}), annual effective dose (A_d), activity concentration index (I), external radiation index (H_{ex}), internal radiation hazard index (H_{in}), alpha index (I_α), activity concentration index (I_β), excess lifetime cancer risk (E_c), and annual gonadal dose equivalent (AG_d) associated with the radionuclides were evaluated in order to assess the radiation hazard of ceramic tiles used in Ile-Ife. The mean activity concentrations of 850, 24, and 128 Bq kg^{-1} were obtained for ^{40}K , ^{232}Th , and ^{238}U , respectively. These resulted in an annual effective dose that ranged from 0.07 to 0.2 mSv y^{-1} with a mean value of 0.14 mSv y^{-1} . The results showed that all the calculated radiological parameters are within the recommended safety limit, hence, do not pose significant radiological hazard when used as building materials.

In order to determine the similarities and correlations among various samples, all the radiological variables above were subjected to correlation analysis.

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1. Introduction

Ceramic tiles are derived from a mixture of clay, sand, and other natural materials that are shaped into slabs and fired

at high temperatures, up to 1250 °C. A glaze surface layer is commonly applied. The glaze is essentially a glass covering, usually a mixture of raw materials such as zinc oxide, feldspar, kaolin, and zircon an opacifier dispersed in water. Their composition is the same as that of all ceramic

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materials, from tableware and sanitary ware to roofing tiles [1,2].

Ceramic tiles are generally preferred for indoor finishing due to their ornamental properties along with their relatively cheap cost. Ceramic tiles may be glazed or unglazed. Zircon is a common opacifying constituent of glazes applied to ceramic tiles and is also used as an opacifier in porcelain tiles by incorporating it directly into the mixture used for forming the body of the tile. All zircon sand contains radionuclides of natural origin, primarily those in the uranium and thorium decay series. Although the concentrations of these radionuclides are low, they are significantly higher than those in normal rocks and soil. Due to the presence of zircon in the glaze, ceramic tiles can show elevated concentration of natural radioactivity [1,3,4]. The fact that ceramic tiles are fundamentally building materials, an assessment of their radiological impact on members of the public is necessary with emphasis on gamma radiation and inhalation of radon released from the product as the main exposure pathways [1].

Ceramic tiles have been extensively used in Nigeria as building materials. The knowledge of the level of natural radioactivity in ceramic tiles as building material is of great importance to determine the associated radiological hazards to human health and to develop a reference data of radiological parameters in ceramic tiles used in Ile-Ife.

Several studies have been done on building materials in Nigeria [5,6]. However, there are few works on ceramic tiles in Nigeria. In Ekiti State, Nigeria, the mean activity concentrations measured in ceramic tiles for ^{226}Ra , ^{232}Th , and ^{40}K were 74 ± 31 , 82 ± 24 , and $618 \pm 231 \text{ Bq kg}^{-1}$, respectively [7]. Jwanbot et al. [8] assessed the indoor and outdoor gamma dose rate associated with commercial building materials in Jos, Plateau Nigeria. For floor tiles, they reported an effective dose range of 0.258–0.264 mSv/yr and 0.218–0.226 mSv/yr for indoor and outdoor, respectively. These values were far below the recommended safe limit set by the International Commission on Radiological Protection (ICRP).

In this work, gamma-ray spectrometry was used to measure the activity concentrations of natural radionuclides in seven varieties of ceramic tiles that are used as flooring materials in Ile-Ife, Osun State Nigeria. Their associated radiological hazards were assessed by calculating the absorbed dose rate, radium equivalent activity, and the external and internal hazard indices. The possibility of the activity concentration associated with ceramic tiles to elicit anomalous response from body tissues was assessed by determining the corresponding Gonadal dose equivalent as well as the excess lifetime cancer risk.

2. Materials and methods

2.1. Sample collection and preparation

A total of 7 different varieties of tiles were collected from the dealers in Ile-Ife based on their type and place of origin. The sample preparation was carried out in Centre for Energy Research and Development (CERD). Each of the samples with a mass of 200 g was weighed into cylindrical polyvinylchloride (PVC) containers, hermetically sealed and kept for 28 days in order to attain secular equilibrium between the parent and the daughter nuclides present.

2.2. Gamma ray spectroscopic technique

The activity concentration of natural radioactivity in the selected samples were determined using a $7.62 \text{ cm} \times 7.62 \text{ cm}$ NaI (Tl) detector surrounded with adequate lead shielding that reduces the background by a factor of approximately 95%. A counting time of 25,200 s was used. The activities of various radionuclides were determined in Bq kg^{-1} from the count spectra obtained from each of the samples using the gamma ray photo peaks corresponding to energy of 1120.3 keV (^{214}Bi), 911.21 keV (^{228}Ac) and 1460.82 keV (^{40}K) for ^{238}U , ^{232}Th and ^{40}K , respectively. For a counting time of 25,200 s, the detection limits of the NaI (Tl) detector system were calculated as 6.77, 11.40, and 12.85 Bq kg^{-1} for ^{40}K , ^{232}Th , and ^{238}U , respectively.

3. Results and discussion

3.1. Activity concentrations of ^{238}U , ^{232}Th , and ^{40}K

The specific activity concentration of ^{232}Th , ^{238}U , and ^{40}K in the samples is presented in Table 1. As can be seen in Table 1, the activity concentration of ^{40}K ranged from 58 to 1768 Bq kg^{-1} while a range of 16 to 43 Bq kg^{-1} and 84 to 168 Bq kg^{-1} were observed for ^{238}U and ^{232}Th , respectively.

Besides the mean values of ^{238}U , the mean values of ^{232}Th and ^{40}K are higher than the worldwide average values of 35, 30, and 400 Bq kg^{-1} for ^{232}Th , ^{238}U , and ^{40}K , respectively [9]. As shown in Table 1, the distribution of the activity concentration in the samples is not uniform; this is likely due to their region of origin and processing composition. The higher activity concentrations found in the studied ceramic tiles may be due to the presence of zircon in the glaze [1,10].

Table 1 – Sample code and the specific activity concentration of the different variety of tiles used in Ile-Ife.

Sample code	Tile Variety	K-40 Bq kg^{-1}	U-238 Bq kg^{-1}	Th-232 Bq kg^{-1}
Ida1	Vitrified	758 ± 5	26 ± 1	168 ± 29
Ida2	Semi vitrified	1768 ± 8	43 ± 1	135 ± 23
Ida3	Colored local	1120 ± 6	22 ± 1	111 ± 21
Ida4	Plain imported	271 ± 3	16 ± 1	84 ± 17
Ida5	Terracotta	1124 ± 6	23 ± 1	150 ± 26
Ida6	Plain local	58 ± 3	16 ± 1	85 ± 19
Ida7	Colored imported	852 ± 5	22 ± 1	163 ± 28
Arithmetic mean		850	24	128

Table 2 – Dose rate, annual effective dose, radium equivalent dose, internal and external radiation hazards, activity concentration index, alpha index, excess lifetime cancer risk and gonadal dose equivalent in different ceramic tiles used in Ile-Ife.

Sample ID	D_R (nGy hr ⁻¹)	A_d (mSv y ⁻¹)	Ra_{eq} (Bq kg ⁻¹)	H_{in}	H_{ex}	I_γ	I_α	$E_C \times 10^{-3}$ (mSv y ⁻¹)	AG_d (μSv y ⁻¹)
Ida1	34 ± 4	0.17 ± 0.02	324 ± 42	0.94 ± 0.11	0.88 ± 0.11	0.17	0.13 ± 0.02	0.58	1.02
Ida2	41 ± 3	0.20 ± 0.02	372 ± 33	1.12 ± 0.09	1.01 ± 0.09	0.20	0.21 ± 0.02	0.70	1.25
Ida3	29 ± 3	0.14 ± 0.01	267 ± 30	0.78 ± 0.08	0.72 ± 0.08	0.14	0.11 ± 0.01	0.50	0.88
Ida4	16 ± 2	0.08 ± 0.01	157 ± 24	0.47 ± 0.07	0.42 ± 0.07	0.08	0.08 ± 0.01	0.28	0.48
Ida5	35 ± 4	0.17 ± 0.02	324 ± 37	0.94 ± 0.10	0.88 ± 0.10	0.17	0.12 ± 0.02	0.59	1.05
Ida6	14 ± 3	0.07 ± 0.01	141 ± 27	0.42 ± 0.07	0.38 ± 0.07	0.07	0.08 ± 0.01	0.25	0.42
Ida7	34 ± 4	0.17 ± 0.02	321 ± 40	0.93 ± 0.11	0.87 ± 0.11	0.17	0.11 ± 0.01	0.58	1.01
Average	29 ± 1	0.14 ± 0.01	272 ± 13	0.80 ± 0.03	0.74 ± 0.03	0.14	0.12 ± 0.02	0.50	0.87 ± 0.04

3.2. Absorbed gamma dose rate (D_R) and the associated annual effective dose (A_d)

The data and formulas provided by the United Nations Scientific Committee on Effect of Atomic Radiation [9] and the European Commission [11] were used to determine the absorbed dose rate (D_R) and the associated annual effective dose (A_d) due to gamma ray emission from the studied ceramic tiles.

In order to convert the activity concentration to absorbed dose rate in air at 1 m above the ground surface for uniform distribution of naturally occurring radionuclides, dose coefficients of 0.12 nGy h⁻¹ per Bq kg⁻¹ for ²³⁸U, 0.14 nGy h⁻¹ per Bq kg⁻¹ for ²³²Th, and 0.0096 nGy h⁻¹ per Bq kg⁻¹ for ⁴⁰K. These coefficients were used to calculate the dose rates of superficial material with a thickness of about 3 cm and a density of about 2600 kg m⁻³ on all walls [11].

$$D_R \text{ (nGy hr}^{-1}\text{)} = 0.0096C_K + 0.12C_U + 0.14C_{Th} \quad (1)$$

Where C_U is the specific activity concentration of ²³⁸U, C_{Th} is the specific activity concentration of ²³²Th, C_K specific activity concentration of ⁴⁰K in Bq kg⁻¹, and D_R is the dose rate in nGy h⁻¹.

In order to determine the annual effective dose rate to whole body indoor, an indoor occupancy factor of 0.8, a conversion factor of 0.7 Sv Gy⁻¹ from absorbed dose in air to effective dose as proposed by Ref. [9] was employed.

$$A_d \text{ (mSv y}^{-1}\text{)} = D_R \text{ (nGy hr}^{-1}\text{)} \times 8760 \text{ (h)} \times 0.7 \times 0.8 \times 10^{-6} \quad (2)$$

where D_R (nGy hr⁻¹) is defined in Eq. (1).

The estimated D_R values are presented in column 2 of Table 2. As shown in the table, the dose rate varies from 14 (Ida6) to 41 (Ida2) nGy h⁻¹ with a mean value of 29 nGy h⁻¹. This mean

value is lower than the world average indoor absorbed gamma dose rate of 84 nGy h⁻¹. In the same vein, the estimated mean value of the indoor annual effective dose rate of 0.14 mSv y⁻¹ is less than the world indoor average value of 0.49 mSv y⁻¹ [9] and 1.0 mSv y⁻¹ limit set by the International Committee on Radiation Protection (ICRP).

3.3. Radium equivalent activity concentration (Ra_{eq})

In order to compare the specific activity of materials containing different amount of ²³⁸U, ²³²Th and ⁴⁰K, the radium equivalent activity Ra_{eq} is used as defined by the following expression [12,13]:

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_K \quad (3)$$

Where C_U , C_{Th} , and C_K are the specific activity concentrations of ²³⁸U, ²³²Th, and ⁴⁰K in Bq kg⁻¹, respectively. All values of Ra_{eq} in the studied samples are lower than the recommended limit (370 Bq kg⁻¹). As shown in column 4 of Table 2, the mean value of Ra_{eq} (272 Bq kg⁻¹) obtained for the ceramic tiles of Ile-Ife is below the recommended safe limit and thus does not pose any radiological hazard when used in a building. The comparison of the specific activities and Ra_{eq} with literature is presented in Table 3.

3.3.1. Internal hazard index (H_{in})

Radon, a gaseous radionuclide and its shortlived daughters are hazardous to the respiratory organs. The internal hazard index (H_{in}) gives the internal exposure to carcinogenic radon and its shortlived progeny. H_{in} is given by the following equation [17].

$$H_{in} = \frac{C_U}{185 \text{ Bq/kg}} + \frac{C_{Th}}{259 \text{ Bq/kg}} + \frac{C_K}{4810 \text{ Bq/kg}} \leq 1 \quad (4)$$

Table 3 – Activity concentrations and radium equivalent activities compared.

Country	Sample	²³⁸ U	²³² Th	⁴⁰ K	Ra_{eq}	References
India	Tile	41.88	57.38	527.53	164.56	[13]
Italy	Ceramic tile	150	70	930	322	[10]
Israel	Ceramic tile	46	48	776	-	[14]
Syria	Ceramic tile	55	54	654	-	[15]
Egypt	Ceramic tile	61–118	55–98	730–1050	267	[16]
Nigeria	Ceramic tile	24	128	850	272	Present study

where C_U , C_{Th} , and C_K are the specific activity concentrations of ^{238}U , ^{232}Th , and ^{40}K in $Bq\ kg^{-1}$, respectively. The values of H_{in} as shown in column 5 of Table 2 ranged from 0.42 to 1.12 with a mean value of 0.8.

3.3.2. External hazard index (H_{ex})

The external hazard index is an additional measure used in order to evaluate the external gamma radiation emanating from building materials. It is used to assess the suitability of a building material. It was calculated as follows [13]:

$$H_{ex} = \frac{C_U}{370\ Bq/kg} + \frac{C_{Th}}{258\ Bq/kg} + \frac{C_K}{4810\ Bq/kg} \leq 1 \quad (5)$$

where C_U , C_{Th} , and C_K are the specific activity concentrations of ^{238}U , ^{232}Th , and ^{40}K in $Bq\ kg^{-1}$, respectively. To ensure the safe use of these samples as a building material and to keep the radiation hazard insignificant, the value of H_{ex} should be less than unity [18]. As shown in Table 2, it can be seen that all but one (Ida2) value are below the standard limit of 1.

3.4. Alpha index (I_α)

It has been shown that there is a linear relationship between radon exhalation rate and sample thickness [19,20] particularly for samples with a thickness that is less than the diffusion length of radon. The diffusion length of radon has been estimated to be $16.0 \pm 0.7\ cm$ [21]. The average thickness of ceramic tiles is less than 5 cm. Hence, alpha index was used to assess the excess alpha radiation exposure caused by radon inhalation from building materials. The Alpha index (I_α) was determined as follows [4]:

$$I_\alpha = \frac{C_U}{200\ Bq/kg} \quad (6)$$

where C_U is the specific activity concentration of ^{238}U in $Bq\ kg^{-1}$. The recommended limit for I_α in building materials is 1.0 as suggested by the Radiation Protection Authorities in Denmark, Finland, Iceland, Norway, and Sweden [22]. As can be observed in Table 2 column 8, all the values of I_α obtained are below the recommended limit. This shows that radon inhalation from the ceramic tiles studied would not cause any excess radiation exposure when used as a building material.

3.5. Activity concentration index (I_r)

The activity concentration index (I_r) is derived to indicate whether the annual dose due to the excess external gamma radiation in a building may exceed 1 mSv. The activity index is calculated for superficial material in the following way [11]:

$$I_r = \frac{C_U}{1701\ Bq/kg} + \frac{C_{Th}}{1458\ Bq/kg} + \frac{C_K}{21259\ Bq/kg} < 1 \quad (7)$$

where C_U is the specific activity concentration of ^{238}U , C_{Th} is the activity concentration of ^{232}Th and C_K is the activity concentration of ^{40}K in $Bq\ kg^{-1}$. The activity concentration index is a screening tool for determining materials that might be of concern when used as a building material. The values of I_r for

the ceramic tiles studied in this work are presented in Table 2. The calculated values ranged from 0.07 to 0.17 with a mean value of 0.14. This shows that these materials can be safely used in building construction.

3.6. Excess lifetime cancer risk (E_c)

The excess lifetime cancer risk was calculated using the following equation [13,23]:

$$E_c = A_d (mSv\ y^{-1}) \times LT \times RF \quad (8)$$

where A_d , LT , and RF are the annual effective dose equivalent, lifetime (duration of life 70 years) and the risk factor ($0.005Sv^{-1}$), respectively. For stochastic effects, a value of 0.05 was used for the public. The world average value of excess lifetime cancer risk is 0.29×10^{-3} [9]. The result obtained are presented in Table 2. From the table, the average value of excess lifetime cancer risk is slightly higher than the world average value.

3.7. Annual Gonadal dose equivalent AG_d ($\mu Sv\ y^{-1}$)

The annual genetically significant dose equivalent (AG_d) to the population is a measure of the genetic significance of the yearly dose equivalent received by the population's reproductive organs (gonads) [24]. The gonads are particularly radiosensitive. A single dose of only 0.3Gy (30 rads) to the testes may result in temporary sterility among men; for women, a 3-Gy (300-rad) dose to the ovaries may lead to temporary sterility. Higher doses increase the period of temporary sterility [25]. The Annual Gonadal dose equivalent was calculated using the equation below [26].

$$AG_d = 3.09C_U + 4.18C_{Th} + 0.314C_K \quad (9)$$

The calculated AG_d according to Eq. (9) ranged from 0.42 to $1.25\ \mu Sv\ y^{-1}$ with a mean value of $0.87\ \mu Sv\ y^{-1}$. Therefore, all the ceramic tiles sampled have values lower than $1\ mSv\ y^{-1}$ as recommended by the International Commission on Radiological Protection (ICRP) for members of the public.

4. Statistical evaluation

4.1. Descriptive statistics

Descriptive statistics include basic statistics (mean, median, variance, etc.), frequency counts, and correlation coefficients of a set of data. It provides an overview of the entire data. The list of basic statistical data, (mean, median, standard deviation, sum, median, range) of the ceramic samples are presented in Table 4. The table helps to simplify the data sets in a way that gives meaningful insight and indicate natural correlations between the variables.

The spread of the data is evaluated by the standard deviation. From Table 4, the range of each variable can easily be deduced from the minimum and maximum values. For ^{40}K , the range is 57.67–1768.48, for ^{238}U the range is 15.89–42.78. Out of the seven types of ceramic tile studied, ^{40}K has a mean value

Table 4 – Descriptive statistics.

Variables	^{40}K	^{238}U	^{232}Th
Mean	850.14	24.01	127.92
Standard deviation	572.06	9.02	35.40
Sum	5950.98	168.10	895.42
Minimum	57.67	15.89	83.54
Median	852.44	22.24	135.08
Maximum	1768.48	42.78	167.73
N analysis	7	7	7

Table 5 – Spearman correlation coefficient.

	^{40}K	^{238}U	^{232}Th	D_R	A_d indoor	R_{eq}	H_{in}	H_{ex}	I_γ	Alpha index	ELTCR	AGDE
" ^{40}K "	1											
" ^{238}U "	0.78571	1										
" ^{232}Th "	0.35714	0.60714	1									
" D_R "	0.85714	0.92857	0.64286	1								
" A_d indoor"	0.85714	0.92857	0.64286	1	1							
" R_{eq} "	0.85714	0.92857	0.64286	1	1	1						
" H_{in} "	0.75	0.96429	0.71429	0.96429	0.96429	0.96429	1					
" H_{ex} "	0.85714	0.92857	0.64286	1	1	1	0.96429	1				
" I_γ "	0.85714	0.92857	0.64286	1	1	1	0.96429	1	1			
"Alpha index"	0.78571	1	0.60714	0.92857	0.92857	0.92857	0.96429	0.92857	0.92857	1		
"ELTCR"	0.85714	0.92857	0.64286	1	1	1	0.96429	1	1	0.92857	1	
"AGDE"	0.85714	0.92857	0.64286	1	1	1	0.96429	1	1	0.92857	1	1

of 850.14 Bq kg⁻¹, while ^{238}U has 24.01 Bq kg⁻¹ and ^{232}Th has 127.92 as mean. This indicates that gamma radiation activities from the ceramic tiles studied are due more to ^{40}K than to the other two primordial radionuclides that is ^{238}U and ^{232}Th .

4.2. Correlation analysis

Correlation is a bivariate analysis that measures the strengths of association between two variables. In statistics, the value of the correlation coefficient varies between +1 and -1. When the value of the correlation coefficient lies around ± 1 , then it is said to be a perfect degree of association between the two variables. As the correlation coefficient value goes toward 0, the relationship between the two variables will be weaker.

Correlation coefficient is used as a measure of relationship between two variables. Spearman's correlation coefficient is a statistical measure of the strength of a monotonic relationship between paired data. Spearman correlation is a common substitute for Pearson correlation coefficient when non-parametric statistics is involved.

All the evaluated variables were subjected to Spearman's correlation analysis to determine the mutual relations and the degree of association between pairs of variables. Table 5 shows the correlation coefficient matrix of all the radiological variables considered for the ceramic tile types in Ile-Ife. The strength of the correlation is defined for the absolute value of the Spearman coefficient as 0.00–0.19 (very weak), 0.20–0.39 (weak), 0.40–0.59 (moderate), 0.60–0.79 (strong), and 0.80–1.0 (very strong). It can be seen from the table that a high positive correlation exists among all the variables. The Spearman's correlation coefficient value of 1 in the matrix is a reflection of the perfect monotonic relationship that exists between the pairs. Similarly, from the data in Table 5, all but one coefficient fall in the

strong to very strong correlation range. ^{40}K has a very high correlation with all other variables except ^{232}Th where it has a weak correlation. The high positive correlation coefficient observed between ^{232}Th and ^{238}U is due to the fact that radium and thorium decay series occur together in nature [27].

5. Conclusions

The activity concentration, gamma absorbed dose rate, radium equivalent activity, annual effective dose and various radiological hazard indices of ceramic tiles used in Ile-Ife have been analyzed using gamma spectrometry technique. The extracted values are, in general, comparable to the average worldwide ranges. The values obtained in this study are within the recommended safety limits, demonstrating that these ceramic tiles do not pose any significant radiation hazard when used as a building material. This study can be used as a baseline for future research work and also be used as a reference data for monitoring possible radioactivity pollutions in the future.

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