Comparative Analysis of Natural Radionuclide Contents and Hazards associated with selected locally made and Imported Tiles

Akinlade Bidemi I., Pemi Stephen G.

Abstract— **Introduction: Ceramics tiles have been used for building decoration in Nigeria. However, the glazing materials and other chemical additives in their production could increase their radionuclides. Analysis of radionuclides: Uranium-238 (^{238}U), Thorium-232 (^{232}Th) and Potassium-40 (^{40}K) in commonly used tiles is hereby presented.**

Materials and Methods: Radionuclides in 16 different samples of tiles, 10 made in Nigeria (LMT) and 6 imported (IT), were analysed using Canberra NaI(Tl) Gamma ray detector at the Secondary Standard Dosimetry Laboratory, National Institute of Radiation Protection and Research, Ibadan, Nigeria. Radiological hazard indices namely, Radium Equivalent Activity (Ra_{eq}), External Hazard (H_{ex}), Internal Hazard H_{in}), Gamma Index(I_γ), Alpha Index(I_α), Absorbed Dose Rate (D_R), Annual Effective Dose (AED), and Excess Lifetime Cancer Risk (ELCR), associated with each sample were thereafter calculated. Results were compared with the international recommended limit (IRL).

Results: The mean concentration (Bq/kg) of ^{238}U, ^{232}Th and ^{40}K obtained from LMT were 18.46±10.46, 138.55±31.27 and 478.95±39.09 while that from IT were 26.87±16.34, 108.35±37.20 and 796.23±227.52 respectively. The mean values of Ra_{eq}, H_{ex}, H_{in}, I_γ, I_α, D_R, AED and ELCR obtained from LMT and IT were 253.46 and 243.12(Bq/kg); 0.68 and 0.66; 0.73 and 0.73; 0.91 and 0.90; 0.09 and 0.13; 115.50 and 115.90(nGy/h); 0.57 and 0.57(mSv/year); and 1.98 and 1.99(x 10^{-3}) respectively.

Conclusion: It was observed that Ra_{eq} and AED obtained from both LMT and IT selected for this study were less than IRL of 370 Bq/kg and 1 mSv/year respectively. Tiles of these samples are considered safe for building decoration.

Index Terms— **Building and decoration materials, imported tiles, locally made tiles, Radionuclide concentration, Radiological hazard parameters.**

I. INTRODUCTION

The use of ceramic tiles for flooring, decoration (interior and exterior) and finishing in homes, offices and public buildings has increased rapidly in the last two decades due to their attractive nature and beauty. It is fast replacing the use of concrete, marble, terrazzo and wood for flooring among builders [1].

The raw materials used for the production of tiles originated from rocks and soil, which are rich in primordial radionuclides such as radioactive Uranium-238 (^{238}U), Thorium-232 (^{232}Th), and Potassium-40 (^{40}K). The primordial radionuclides refer to the radionuclides formed during the formation of the universe and their half-lives are so long that they are still present today [2]. These radionuclides find their way into building materials, air, water, food and human body through various activities like mining, construction, manufacturing, oil and gas exploration.

Building materials such as ceramic tiles are the major sources of radiation exposures at home and offices [3]. As individuals spend more than 80% of their time indoors (at home or in the office), the internal and external radiation exposure from building materials could results in prolonged radiation exposure [4]

Ceramic tiles are derived from a mixture of plasticity clay, sand, and other natural materials that are shaped into slabs and fired at high temperature of about 1250 °C. [5]. A glaze surface layer, commonly applied on tiles, is essentially a mixture of zinc oxide, feldspar, quartz, kaolin, and zircon. The use of glazing materials and other chemical additives in the production of ceramic tiles has the potential to increase its radiological content significantly [6].

The radioactivity concentration in ceramics tiles depend on the geographical location and geochemical conditions under which the raw materials are obtained. The manufacturing process may sometimes enhance the natural radionuclide concentration in tiles by turning naturally occurring radioactive materials (NORM) in the raw materials to technologically enhanced naturally occurring radioactive materials (TENORM) so that the end products give rise to elevated radiation exposure above the background radiation.

In the past, Nigerians depend solely on the use of imported tiles for home, offices and public buildings. Now, the emergence of indigenous ceramic tiles manufacturing companies in Nigeria has given Nigerians the opportunity to choose among varieties of both locally made and imported tiles. The raw materials used for the production of locally made tiles and imported tiles were obtained from different geological location with varying radionuclides concentration and this has made the assessment of radiological risk associated with ceramic tiles to become a global concern.

This study was carried out to do a comparative analysis of the radionuclides concentration and hazards associated with locally made and imported tiles; compared the results with the international recommended levels and assess their radiological impact on the health of the general public.

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II. MATERIALS AND METHODS

A. Samples Collection and Preparation

A total of 16 tiles comprise of 10 locally made and 6 imported from either China, Spain, Italy and United Arab Emirates were purchased from different commercial tiles' dealers at the building materials market, Iwo road Ibadan, Oyo state Nigeria and used for this study. All samples of tiles were taken to the laboratory of the Geo Solutions Nigeria Limited, Ibadan where each tile was crushed, pulverized, dried and pass through 250µ sieve mesh size in order to achieve homogeneity in the powder sample. Each powder sample was sealed in new polythene nylons and labeled accordingly. Tile samples before preparation are shown in figure 1.

The samples were thereafter taken to the Environmental Monitoring Laboratory (EML) of the National Institute of Radiation Protection and Research (NIRPR), University of Ibadan, Ibadan Nigeria for analysis of their radionuclides' concentration. Before the analysis, each sample was thoroughly mixed, weighed with digital chemical balance and recorded. 300g of each sample was put into a non-radioactive plastic cylindrical container of dimension 70 mm diameter and 90 mm height. The containers were hermetically sealed and left for more than 4 weeks in order to allow Radium and its short-lived progeny to attain secular equilibrium, prior to gamma counting.

B. The Counting Assembly (Gamma Ray Spectrometer)

The Coaxial NaI (Tl) scintillation gamma spectrometer at the NIRPR was used as counting unit for all samples. It has Canberra vertical coaxial NaI (Tl) detector, crystal size 76 mm x 76 mm, Model 802 and serial number 13000850. The crystal detector is encapsulated with an Aluminium casing and enclosed in a 60 mm thick lead shield. The unit is coupled to a Canberra Multichannel Analyser (MCA) through a pre-amplifier and then connected to a computer system. To minimize the effect of scattered radiation from the shield, the detector was placed at the centre of the chamber. The computer system uses Genie 2000 spectroscopy software for processing the detector responses. In addition, the counting assembly is connected to a solar inverter system (alternative power supply) which automatically switches on whenever there is power failure to prevent interruption in the counting. The Counting assembly is as shown in figure 2.

C. Energy Calibration of the Detector

The energy calibration of the coaxial NaI (Tl) detector was done and relative efficiency obtained was used for the calculation of the radioactivity concentration. The Genie 2000 spectroscopy software has counts versus channels scale. The energy calibration was carried out to match these channels with the energy of gamma ray produced. This involves the use of three points sources of known energy: Co-60 (1173.22 keV and 1332.49 keV), Cs-137 (661.65 keV) and Am-241 (59.54 keV). These sources were placed in the detector and counted for 600 seconds using the energy calibration method and the counts in the region of interest (ROI) were plotted against the corresponding channels. The software uses the following formula to calibrate energy between two points (ROI):

$$E = a(Ch) + b \qquad (1)$$

Where *E* is the energy at a particular channel, *a* is the slope of the plot and *b* is the intercept.

The NaI detector used for this study has a resolution of 7.5% specified at 661.65 keV peak of ^{137}Cs. This is capable of distinguishing the gamma ray energies considered during this measurement.

D. Measurements (counting)

Three type of measurements were carried out to estimate the activity concentrations of U-238, Th-232 and K-40 from the tile samples. These measurements are:

(i) **Background Measurement:** This was carried out by placing an already washed empty plastic cylindrical container in the detector and counted for 10800 seconds and recorded.

(ii)Standard Measurement: This involved placement of 100g IAEA 315 standard source consisting of radionuclides with known activity in the container and counted for 10800 seconds.

(iii) **Sample Measurement:** This was carried out by placing each tile sample of known mass in the container under similar condition and counted for 10800 seconds.

The background measurement count was thereafter subtracted from each sample measurement count in order to get the actual measurement count due to the activity of the radionuclides present in the tile samples.

E. Spectra Evaluation

The spectra were analyzed using Genie 2000 spectra analysis software which matched various gamma energy peaks to a library of possible radionuclides. The activity concentration of U-238 was estimated using Bi-214 with energy 1764.18 keV at emission percentage of 17%, activity of Th-232 was estimated using Tl-208 with energy 2614.5 keV at 100% emission percentage and K-40 was estimated using its own gamma ray energy of 1460.5 keV. The area of the spectrum (counts) and its associated uncertainty were estimated by placing markers (cursor) at the two tail of the spectrum.

F. The Radioactivity Concentration

The radioactivity concentration of ^{238}U, ^{232}Th, and ^{40}K present in each of the tile sample was calculated using equation below:

$$C_n = \frac{N}{\epsilon t Y m} \qquad (2)$$

Where C_n is the radioactivity concentration (Bq/Kg) of the radionuclides in each sample, m is the mass (kg) of the sample, t is the counting time (second), Y is the gamma yield, ϵ is the efficiency of the detector, N is the Net counts (sample counts – background counts) (counts/sec).

G. The Radiological Hazard Indices Associated with Ceramic Tiles.

The radioactivity concentration of ^{238}U, ^{232}Th, and ^{40}K obtained from the analysis of each of the tile sample was thereafter used to calculate the radiological hazard indices associated with the tile using the following respective equation:

(i) **Radium Equivalent (Ra_{eq}):**

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

where C_U, C_{Th} and C_K are the average activity concentration of ^{238}U, ^{232}Th and ^{40}K measured in Bq/Kg respectively. The international acceptable level for Radium Equivalent is 370 Bq/kg. [7]–[8].

(ii) **External Hazard Index (H_{ex}).**

$$H_{ex} = \frac{C_U}{370\ (Bq/Kg)} + \frac{C_{Th}}{259\ (Bq/Kg)} + \frac{C_K}{4810\ (Bq/Kg)} \quad (4)$$

where, C_U, C_{Th} and C_K are the average activity concentrations of ^{238}U, ^{232}Th and ^{40}K, in Bq/kg respectively. For safe use of a material in the construction of dwellings, the recommended reference level for (H_{ex}) is 1. [8]

(iii) **Internal Hazard Index (H_{in}).**

$$H_{in} = \frac{C_U}{185\ (Bq/Kg)} + \frac{C_{Th}}{259\ (Bq/Kg)} + \frac{C_K}{4810\ (Bq/Kg)} \quad (5)$$

where C_U,C_{Th} and C_K are the average activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively in Bq/kg.For the safe use of tiles for decorative purposes in the construction of buildings, recommended level for H_{in} is 1 [8]–[9].

(iv) **Gamma Index (Iγ)**

$$I_\gamma = \frac{C_U}{300\ (Bq/Kg)} + \frac{C_{Th}}{200\ (Bq/Kg)} + \frac{C_K}{3000\ (Bq/Kg)} \quad (6)$$

where C_U, C_{Th} and C_K are the average activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively in Bq/kg.

The recommended value for Iγis 1. [7], [8], [10]

(v) **Alpha Index (Iα)**

$$I_\alpha = \frac{C_U}{200\ (Bq/Kg)} \quad (7)$$

where C_U is the average activity concentration of ^{238}U in Bq/kg. The recommended level for Iα in building materials is 1. [1], [8].

(vi) **The Absorbed Dose Rate (D_R).**

$$D_R(\text{nGy/h}) = 0.642C_U + 0.642C_{Th} + 0.0417C_K \quad (8)$$

where C_U, C_{Th} and C_K are the average activity concentrations of ^{238}U, ^{232}Th and ^{40}K respectively in Bq/kg. The recommended level for D_R is 84 nGy/h. [8], [11].

(vii) **Annual Effective Dose (AED)**

The annual effective dose was estimated using the dose conversion factor (DCF) of 0.7 Sv/Gy and occupancy factor (OF) of 0.8 for indoor exposure. Time (T) = 8760 hours per year. The occupancy factor is used to convert the absorbed dose in air to effective dose. The indoor annual effective dose (AED) from tiles was calculated using equation below:

$$AED\left(\frac{mSv}{y}\right) = D_R\left(\frac{nGy}{h}\right) \times T \times OF \times DCF\ X\ 10^{-6} \quad (9)$$

The recommended level for AED from building materials, such as tiles, is 1 mSv/year [1], [8].

(viii) **Excess Life Cancer Risk (ELCR)**

$$ELDR = AED(mSv/y) \times LT \times RF \quad (10)$$

where AED is the Annual effective dose, LT is the lifetime (average 70 years), and RF is the fatal cancer risk factor

(0.05/Sv) for the general public. [12]

The recommended average value of ELCR is 0.29 x10^{-3} [8]

III. RESULTS

The mean activity concentration of ^{238}U, ^{232}Th, ^{40}K found in locally made tiles are 18.46 ± 10.46, 138.55 ± 31.27, 478.95 ± 239.09 Bq/kg respectively while that from imported tiles are 26.87 ± 16.34, 108.35 ± 37.20, 796.23 ± 227.56 Bq/kg respectively and are presented in Table 1 and Table 2 respectively. The Radiological hazards indices namely, The Radium equivalent, External hazard index, Internal hazard index, Gamma Index, Alpha index, Absorbed dose rate, Annual effective dose, and Excess lifetime cancer risk associated with the activity concentration of ^{238}U, ^{232}Th, ^{40}K in both the locally made and imported tiles are presented in Table 3(a and b) and Table 4 (a and b) respectively.

Figure 1: Different Tile samples considered for the study

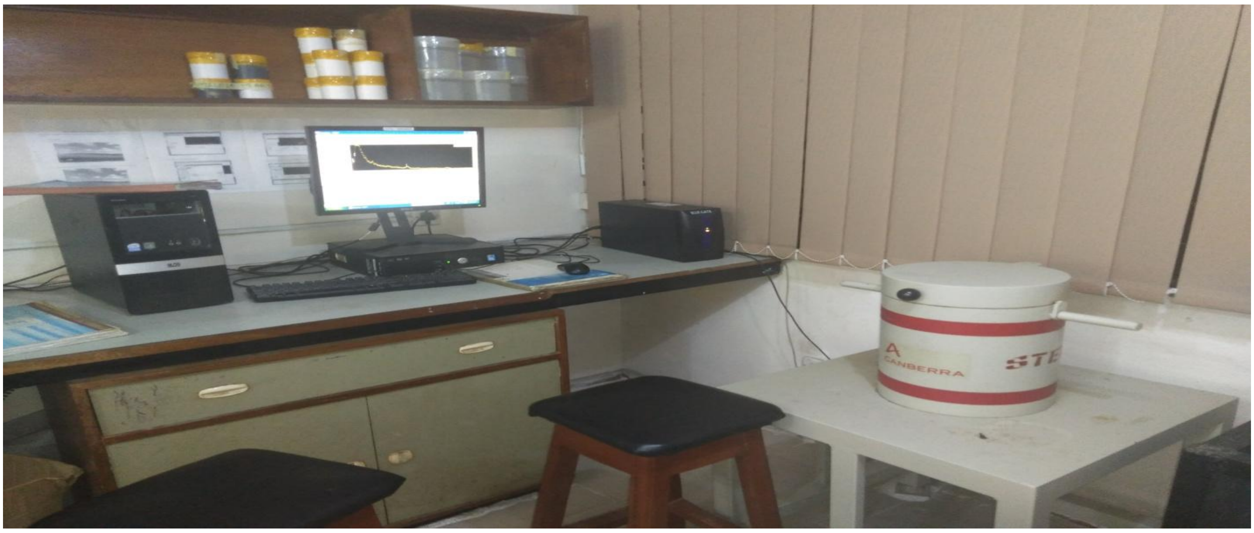

Figure 2: The Gamma Counting Assembly

Table 1: **Radioactivity Concentration in Locally Made**

Sample Name	Size cm^2	^{238}U Bq/kg	^{232}Th Bq/kg	^{40}K Bq/kg
Royal floor	40x40	12.45±1.91	202.28±14.50	676.8±42.13
Royal wall	25x40	26.74±3.93	100.51±7.96	789.32±46.78
NISPO floor	30x30	36.25±3.49	140.16±8.54	550.94± 29.40
Goodwill floor	40x40	33.12±4.46	173.47±13.00	735.77±45.93
Golden floor	40x40	3.73±0.79	120.34±8.76	214.92±15.80
PNT floor	30x30	10.80±1.81	119.50±8.70	382.17±25.44
PNT wall	25x40	20.21±2.08	129.27±7.89	717.61±37.90
Golden wall	25x40	14.86±2.19	135.13±10.28	209.40±15.46
Goodwill wall	25x50	12.41±1.95	107.91±8.09	259.16±1.93
Goodwill glaz wall	25x40	14.00±2.30	156.91±10.74	253.44±18.53
Mean ± SD		18.46 ± 10.46	138.55 ± 31.27	478.95±239.09

Table 2: Radioactivity Concentration in Imported

Sample Name	**Size cm^2**	**Origin**	**^{238}U (Bq/kg)**	**^{232}Th Bq/kg**	**^{40}K (Bq/kg)**
Virony glaze floor	40x40	China	45.17±5.45	179.94±13.43	587.36±38.78
RAK Ceramic floor	40x40	UAE	44.54±5.15	93.95±8.00	570.96±35.82
NATO floor	30x60	Italy	29.35±3.68	82.64±6.47	839.47±49.26
Cantabria Wall	25x50	Spain	6.08±1.44	79.01±6.18	1016.01±56.30
Virony Wall	20x30	China	11.20±1.74	101.63±8.05	658.50±39.31
AB Ceramic floor	60x60	China	24.89±3.45	112.94±8.68	1105.1±62.13
Mean ± SD			26.87±16.34	108.35±37.20	796.23±227.56

Table 3a: Radiological Hazard Indices in Locally made Tiles

Sample Name	**Raeq Bq/kg**	**Hex**	**Hin**	**Iγ**
Royal floor	353.82	0.95	0.99	1.28
Royal wall	231.25	0.62	0.70	0.85
NISPO floor	279.10	0.75	0.85	1.01
Goodwill vitrified floor	337.84	0.91	1.00	1.22
Golden Crown floor	192.37	0.52	0.53	0.69
PNT ceramic floor	211.11	0.57	0.60	0.76
PNT ceramic wall	260.32	0.70	0.76	0.95
Golden crown wall	224.22	0.61	0.65	0.79
Goodwill crack wall	186.68	0.50	0.54	0.67
Goodwill glaze wall	257.90	0.70	0.73	0.92
Mean ± SD	253.46±57.01	0.68±0.2	0.73±0.2	0.91 ± 0.2

Table 3b: Radiological Hazard Indices in Locally made Tiles

Sample Name	Iα	DR nGy/h	AED mSv/y	ELCR x 10 -3
Royal floor	0.06	158.39	0.78	2.72
Royal wall	0.13	110.79	0.54	1.90
NISPO floor	0.18	130.90	0.64	2.25
Goodwill vitrified floor	0.17	156.72	0.77	2.69
Golden Crown floor	0.02	84.04	0.41	1.44
PNT ceramic floor	0.05	95.05	0.47	1.63
PNT ceramic wall	0.10	120.98	0.59	2.08
Golden crown wall	0.07	99.89	0.49	1.72
Goodwill crack wall	0.06	83.95	0.41	1.44
Goodwill glaze wall	0.07	114.33	0.56	1.96
Mean ± SD	0.09 ± 0.05	115.50±26.83	0.57± 0.1	1.98±0.46

Table 4a: Radiological Hazard Indices in Imported Tiles

Sample name	Origin	Ra_{eq} Bq/kg	H_{ex}	H_{in}
Virony glaze floor	China	347.71	1.06	0.94
RAK Ceramic floor	UAE	222.85	0.72	0.60
NATO floor	Italy	212.16	0.65	0.57
Cantabria Wall	Spain	197.30	0.55	0.53
Virony Wall	China	207.24	0.59	0.56
AB Ceramic floor	China	271.49	0.80	0.73
Mean ± SD		243.12±57.45	0.66±0.10.2	0.73±0.19

Table 4b: Radiological Hazard Indices in Imported Tiles

Sample name	Origin	I_γ	I_α	D_R nGy/h	AED mSv/y	ELCR x 10^{-3}
Virony glaze floor	China	1.25	0.23	162.18	0.80	2.78
RAK Ceramic floor	UAE	0.81	0.22	109.15	0.54	1.87
NATO floor	Italy	0.79	0.15	103.76	0.51	1.78
Cantabria Wall	Spain	0.75	0.03	93.99	0.46	1.61
Virony Wall	China	0.76	0.06	96.03	0.47	1.65
AB Ceramic floor	China	1.02	0.12	130.28	0.64	2.24
Mean ± SD		0.90±0.20	0.13±0.08	115.90±26.14	0.57±0.13	1.99± 0.45

IV. Discussion

The radioactivity concentration and health hazards associated with the use of ceramic tiles made locally and of foreign origin for building decorations in Nigeria have been analysed. It can be seen from Tables 1 and 2 that the mean activity concentration of ^{238}U, ^{232}Th, ^{40}K radionuclides in tiles made in Nigeria were found to be slightly varied from each other even though they are within the range of recommended values [13]. It was also observed that the activity concentration of ^{40}K was higher than that of ^{238}U and ^{232}Th for all the tile samples under investigation. This could be due to the abundant presence of ^{40}K in the earth crust, where most of the raw materials used for the production of tiles were extracted. This same trend was observed in a similar study carried out by Ademola et al, [14] where the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K reported (72 ± 14, 84 ± 18 and 629 ± 198 Bq/kg) for imported tiles showed higher value for ^{40}K. In another report by Gbenu et al. [5], the mean activity concentration of ^{40}K, ^{232}Th, and ^{238}U in ceramic tiles used in homes and offices were found to be 850, 24, and 128 Bq/kg respectively. Similarly, Joel et al. [15] found the mean activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in tiles made in Nigeria to be 61.1 ±5.5 Bq/kg, 70.2 ± 6.08 Bq/kg and 514.7 ±59.8 Bq/kg respectively and those made in China to be 58.2 ± 0.5, 161.5 ± 9.4 and 455.7 ± 15.1 Bq/kg respectively. All these authors reported higher activity concentration values for K-40.

In Tables 3 and 4, it can be seen that the mean Radium Equivalent obtained for locally made tiles (253.46 Bq/kg) and imported tiles (243.1 Bq/kg) are within the international recommended value of 370 Bq/kg. These values are similar to the values reported by Ademola et al, for wall tiles (259 Bq/kg) and floor tiles (241 Bq/kg) and by Joel et al (204.42 Bq/kg). The external hazard index obtained from both locally made tile and imported tiles are within the international recommended level of 1 except for an imported tile from China (Virony Glaze floor) in Table 4 that has a value that is slightly higher (1.06) than 1. On the other hand, the mean internal hazard of all samples of tiles from Nigeria origin and imported tiles are within the recommended level of one. On the average, the gamma index of all the samples investigated are within the recommended value of one except the following locally made tiles: NISPO floor tile (1.01), Goodwill vitrified floor tile (1.22), Royal floor tile (1.28) and the following tiles imported from China: AB ceramic floor tile (1.02) and Virony glaze floor tile (1.25). The mean Alpha index of all the samples of Tiles considered in this study are within the recommended value of one. As seen in Tables 4 and 5, the mean absorbed dose rate for Tiles made in Nigeria (115.50 nGy/h) and imported tiles (115.90 nGy/) obtained in this study are higher than the reference level of 84 nGy/h (8) by 38%. The only exception is the Goodwill crack wall tile, made in Nigeria, which has a value of 83.95 nGy/h.

The mean annual effective dose doses (0.57 mS/year) derivable from all samples are within the recommended level of 1mSv/year (1, 8). The mean excess lifetime cancer risk obtained in this study as presented in Table 4 and 5 for tiles made in Nigeria (1.98 x 10^{-3}) and imported tiles (1.99 x 10^{-3}) is higher than the recommended value of 0.29 x10^{-3} [8, 12]. In all the tiles samples investigated, the Nigeria made tiles (Golden Crown floor tiles and Goodwill Crack wall tile) and Imported tiles (Cantabria wall tile from Spain) have the lowest value for all radiological hazard indices estimated. Hence, they are among the safest tiles to use. Whereas, the Nigeria made tiles (Royal floor tile) and imported tiles (Virony glaze floor from China) has the highest value for all radiological hazard indices estimated.

V. Conclusion

The measurement of radioactivity concentration of 16 tile samples comprises of 10 Nigeria made and 6 Imported tiles used for building purposes in Nigeria has been analysed using Canberra vertical coaxial NaI (Tl) detector Model 802 at the

Environmental Monitoring Laboratory of the National Institute of Radiation Protection and Research, Ibadan, Nigeria. While the Annual Effective Dose for imported tiles ranged from 0.46 to 0.80 mSv/year and the mean Annual Effective Dose was 0.57mSv/year. The Radium Equivalent activity and Annual Effective Dose for all the tiles samples investigated is less than the recommended value of 370 Bq/Kg and 1 mSv/year respectively. Hence, they are safe for building and decoration purposes.

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