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Radiological risk assessment in some imported tiles from China and its possible health implication for Nigerian users

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Abstract. This investigation was carried out to ascertain the natural radionuclides contents in the tiles imported from China for Nigerian use and its radiological risk using gamma-ray spectroscopy. Hyper-Germanium detector (HPGe) was applied to 7 brand samples of tiles exported from China to Nigeria for building-related issue in order to determine the concentrations of radioactive materials such as ^{226}Ra, ^{232}Th, and ^{40}K present in the imported tiles sample. The concentrations for the ceramics tiles in terms of average were observed to be 58.20 ± 0.52, 161.50 ± 9.45 and 455.70 ± 15.12 Bq/kg respectively for ^{226}Ra, ^{232}Th, and ^{40}K. Radiological risk indices which include radium equivalent activity, dose rate, both hazards that occur externally and internally, Annual Effective Dose (mSv/y), Gamma (Iγ) and Alpha Radiological Index (Iα) were evaluated for the imported tiles. The mean values estimated were: 317.16 Bq/kg; 153.92 nGyh^{-1}; 0.87; 1.08; 1.52 mSv/y; 1.15 and 0.29 respectively. The mean value of radium equivalent estimated in this study is within the range of international reference acceptance value while the absorbed dose rate is higher.

1. Introduction

The two main sources of natural radiation exposures include cosmic ray of high-energy particles that is incident on the surface of the earth and radionuclides that has its origin in the earth's crust and are available everywhere in the environment and this includes human body [1], and human beings are continuously exposing to ionizing radiation from natural sources which is an inescapable feature of life on earth. For most individuals, this exposure exceeds that from all man-made sources combined. The radionuclides that are of interest in the research about environmental radioactivity are ^{226}Ra, ^{232}Th and ^{40}K [2,3,4]. The exposition of humans to this natural radioactivity [5] mostly depends on the impact of the anthropogenic activities on types of the soil and geological formation of an area. In the world, building materials which contain radio-nuclides have been in use for many decades. 80% of populace spends most of their time indoors and exposition of the individual to

radiation emanating from these building materials whether internally and externally is a concern [6]. The radiation that occurs externally is as a result of gamma-ray emission from radioactive nuclides while the internal radiation occurrence results from ^{222}Rn [7]. The risk associated with these building materials such as tiles may be high as a result of high levels of radioactivity concentration and consequently posed danger to the users. This can lead to disease such as respiratory-organs disease (which includes cancer of the lung) due to an increase in radon gas present in the materials [8, 9, 10]. Individuals, group of people and various international bodies are engaged in research efforts regarding natural environmental radioactivity due to its hazardous effect [11,12,13,14,15,16,17,18,19,20,21,22,23,24,25,26,27]. In the present investigation, characterization of radiation was carried out for various brands of ceramics tiles imported from China for Nigerian use especially for building purposes. This is needed to ascertain the possible radiological risks to human societal-health. The result of this investigation serves as additional information to the pool of existing databases. This investigation, therefore, can be used in promoting standards for the kinds of ceramics tiles that would be imported for Nigerian use in the light of international reference recommendations.

2. Methods

Seven (7) samples of various types of tiles such as Virony tile, Virony Rustic, Virony unglazed, Virony Rustic Glass, Idris Tiles, Vironi Ceramic and Virony Glazed which were imported from China were employed in this study. The used tiles were bought from Lagos commercial business market in Nigeria. Table 1 gives the identity (ID) and the names of the samples. Cataloguing and labelling were done at the early stage for easy recognition. The samples were broken into smaller pieces for further action. All the sample materials used were shivered using milling machine in the laboratory of Pascall Engineering. Thereafter the milling machine used in the laboratory was cleaned adequately using a blower that uses pressure that is very high. The whole processes were repeated for each sample until all the materials were thoroughly crushed into powdered form. The crushed samples underwent sieve mesh of 250 μm size; consequently, one kilogramme of the sample was sieved, weighed, kept in a bottle made of high-density polyethene and marked with a marker for proper identification. In addition, the sieved samples were sent for gamma spectroscopy analysis in Universiti Teknologi Malaysia Nuclear Laboratory centre. Moreover, a digital balance in the range of $\pm$ 0.01 g was used to measure the samples which were later transferred into Marinelli beakers. Thereafter, 4 weeks of secular equilibrium were observed for the sieved samples in Marinelli beakers with the intention to equate nuclide parent with the daughter (progeny). Interestingly, the geometry sizes of the detector used for Marinelli beaker were the same for both the samples and International Atomic Energy Agency (IAEA) standard.

Table 1. Tiles specimen imported from China

Specimen trademark	Co	Size (mm)
	C	40×40
VT		
	C	40×40
VR		
	C	30×30
VU		
	C	40×40
VRG		
IRT	C	60×60
VC	C	40×40
VG	C	60×60

VT-Virony tile; VR- Virony rustic; VU- Virony unglazed; VRG- Virony rustic glazed; IRT- Idris tile; VC- Virony ceramic; VG- Virony glazed; C- China; Co- Country

2.1 Gamma Spectroscopy Analysis

In measuring the gamma energy spectrum that ranges from 50 keV to 2000 keV a spectrometer of the high resolution was used. This comprises of an HPGe of the relative efficiency of about 20 % and 1.8 keV of resolution. Genie-2000 was the software used in measuring gamma ray. The liquid nitrogen at 77 K was used to cool the detector hence, causing the reduction of current leakage and heat noise, and its heat-up sensor is connected to the high voltage detector [17]. The background radiation was minimised via pre-amplifier put inside a lead shield [17]. The investigation of [27] revealed that Minimum Detectable Activity (MDA) for Uranium, Thorium and Potassium as 1, 2 and 13Bq/kg accordingly. Table 2 shows the gamma energy, isotopes and disintegration of gamma rays.

Table 2. Gamma energy, isotopes and disintegration of gamma rays [28,29]

Radioactive Series	Decay Isotopes	Energy (keV)	Gamma Disintegration (%)
Uranium	214-Pb	352.0	35
	214-Bi	609.4	43
Thorium	208-Tl	583.1	30
	228-Ac	911.1	29
Potassium	^{40}K	1460.8	10.68

2.1.1 Standard Sample Preparation for Gamma Spectrometry

The standard sample (S-14) and Lake-Sediment (SL-2) which are IAEA established samples were employed as source materials and blended with Silicon (IV) Oxide in Marinelli beakers. The content of U-238 and Th-232 in the standard sample is 29 and 610 ppm. 20.00 g of weight measured from the standard sample was rigorously blended with 100.00 g of Silicon (IV) Oxide in a Marinelli beaker (marked as S-14). After mixture, the concentrations of U-238 and Th-232 were 4.63 and 97.3 ppm accordingly. The IAEA (SL-2) which is a standard sample was used in calculating the activity concentration of potassium (K) (which is 240 Bq kg^{-1} in value). Another Marinelli beaker of 74.18 g of IAEA standard sample was also mixed with SiO2 at 100.00 g and marked as SL-2. And this serves as a background value for the standard samples (Table 3).

Table 3. Standard samples of IAEA used in the investigative study [28, 29]. Where C typifies concentrations.

Standard Sample Code	C		
	Uranium (ppm)	Thorium (ppm)	Potassium (%)
S-14 (Thorium ore)	29	610	-
SL-2 (Late sediment)	-	-	240

2.1.2 Calculation of ^{238}U *and* ^{232}Th *(for IAEA standard sample S-14) and* ^{40}K *in SL-2)*

^{238}U:

The concentration of Uranium-238 in the standard sample (S-14) is equivalent to 29 ppm

The measured weight of the sample used is equivalent to 20.00 g

The concentration of Uranium-238 in the standard sample (S-14) used = 29 x 20.01μg g^{-1}

$$= 580.29 \text{ ppm} \tag{1}$$

^{232}Th:

For ^{232}Th in standard sample (S-14) = 0.061 (wt. %) $= \frac{0.061 \times 100}{1000000}$

$$= 0.061 \times 10^{-4}$$

The weight of the measured standard sample (S-14) used is equivalent to 20.00 g

The concentration of ^{232}Th in the standard sample (S-14) used = 610 x 20.00

$$= 12206 \text{ ppm.} \tag{2}$$

^{40}K:

Concentration of activity of ^{40}K = 240 Bq kg^{-1} (IAEA SL-2)

The weight of lake sediment used = 74.18 g

Concentration of activity of ^{40}K in SL-2 used $= \frac{240 Bq}{1000} \times 74.18g = 0.24 \times 74.18 = 17.8 \text{ Bq}$ (3)

The concentration of the Uranium-238 and Thorium-232 was determined using equation (4) and equation (5). Equations (6) and (7) was used for ^{40}K

$$C_{samp} = \frac{W_{std} \times N_{samp}}{W_{samp} \times N_{std}} C_{std} \tag{4}$$

where C_{samp} and C_{std} are a concentration in ppm for both samples collected and standard sample; W_{std} and W_{samp} are standard and collected sample weight measured in gram; N_{samp} and N_{std} are net counts of the photopeak area for standard and collected sample.

The uncertainty in the concentration measured was determined using the approach of [30,31].

$$C_{samp}(ppm) = (\frac{\Delta W_{std}}{W_{std}})^2 + (\frac{\Delta W_{samp}}{W_{samp}})^2 + \left(\frac{\Delta N_{samp}}{W_{samp}}\right)^2 (\frac{\Delta N_{std}}{N_{std}})^{\frac{1}{2}} \times C_{std} \tag{5}$$

2.1.3 The concentration of activity present in potassium was determined using equation (6):

$$A_{samp} = \frac{W_{std} \times N_{samp}}{W_{samp} \times N_{std}} A_{std} \tag{6}$$

where A_{samp} and A_{std} are activity concentration in ppm for both sample collected and standard sample; W_{std} and W_{samp} are standard and collected sample weight; N_{samp} and N_{std} are net counts of the photopeak area for standard and collected sample.

The uncertainty values for the concentration of potassium were determined using [7] [30, 31]:

$$\Delta A_{samp}(ppm) = \left(\frac{\Delta W_{std}}{W_{std}}\right)^2 + \left(\frac{\Delta W_{samp}}{W_{samp}}\right)^2 + \left(\frac{\Delta N_{samp}}{W_{samp}}\right)^2 \left(\frac{\Delta N_{std}}{N_{std}}\right)^{\frac{1}{2}} \times A_{std}$$

(7)

3. Results and Discussion

3.1 Determination of Concentration of radionuclides

The evaluated activity concentration in the sample tiles is presented in Table 4. The observed concentration of radioactivity content in the samples used ranged between 42.50 ± 0.53 and 75.0 ± 0.52 Bq/kg for ^{226}Ra, 41.50 ± 9.41 and 405.5 ± 9.41 Bqkg for ^{232}Th, and 290 ± 15.21 and 740 ± 15.10 Bqkg^{-1} for ^{40}K accordingly. The results show that VC (40 x 40 mm) had the highest ^{226}Ra concentration of 75 ± 0.52 Bq/kg and 405 ± 9.40 Bq/kg for ^{232}Th respectively while Idris (60X60 mm) tile has a value of 740 ± 15 Bq/kg for ^{40}K. The lowest values of 42.5 ± 0.53, 41.50 ± 9.41, and 290 ± 15.10 Bq/kg are found for the tiles samples VRG (40 x 40 mm), Virony Rustic (40 x 40 mm) and Vironi Ceramic (40 x 40 mm). The mean value concentration of the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K for the tiles are 58.20 ± 0.52, 161.50 ± 9.45, and 455.70 ± 15.12 Bq/kg respectively. The result shows that the mean values were below the standard value when compared with [18, 19, 20, 21] UNSCEAR report.

Table 4. Measured Radioelement concentration in China tiles used for this study

Specimen trademark	Size	AC($Bqkg^{-1}$)		
		^{226}Ra	^{232}Th	^{40}K
Virony	40X40	55.5 ± 0.51	126.5 ± 9.51	530.0 ± 15.11
Virony Rustic	40X40	59.5 ± 0.52	41.5 ± 9.41	480.0 ± 15.10
Virony unglazed	30X30	55.0 ± 0.52	52.0 ± 9.41	440.0 ± 15.10
Virony Rustic Glass	40X40	42.5 ± 0.53	63.0 ± 9.32	390.0 ± 15.12

IDRIS Tiles	60X60	65.0 ± 0.51	337.0 ± 9.43	740.0 ± 15.10
Vironi Ceramics	40X40	75.0 ± 0.52	405.5 ± 9.41	290.0 ± 15.21
Virony Glazed	60X60	55.0 ± 0.53	105.0 ± 9.70	320.0 ± 15.13
Mean values		58.2 ± 0.52	161.5 ± 9.45	455.7 ± 15.12

AC – Activity concentration

3.2 Radiological Hazard Indices Assessment

3.2.1 Radium equivalent (Raeq) Estimation

The amount of radioactive concentrations present in ^{226}Ra, ^{232}Th and ^{40}K in the analysed building materials is not spread uniformly. The radium equivalent activity of the measured radio-isotopes is usually based on comparison factor with the activity of each of ^{226}Ra, ^{232}Th and ^{40}K contents in the materials (for instance tiles) used for building purposes. Radium equivalent activity with the unit as $Bqkg^{-1}$ was calculated using equation (8).

$$Raeq = AC_{RA} + 1.43AC_{Th} + 0.077AC_{K} \qquad (8)$$

where the activities concentration of ^{226}Ra, ^{232}Th and ^{40}K are represented by AC_{RA}, AC_{Th} and AC_{K} and are measured in $BqKg^{-1}$ respectively. According to [1]) the maximum value of Radium equivalent in tiles used materials must be less than $370\,Bqkg^{-1}$. The estimated value for radium equivalent activity in this present investigation ranged between 155.81 and 677.9 $Bqkg^{-1}$. The mean result of the radium-equivalent is 317.16 $317.16Bqkg^{-1}$ and is lower than the reference value as reported by [18, 19, 20, 21]. This is presented in Table 5.

3.2.2 The Dose Rate

Furthermore, the dose rates (DR) were obtained from the estimated activity concentrations as shown in Table 5. The overall air absorbed dose rate in the open air is 1 m above the ground level due to the ejection of gamma radiation from the radioactive nuclides of ^{226}Ra, ^{232}Th and ^{40}K in $Bqkg^{-1}$ obtainable in a habitat is determined using equation (9) [1,20]

$$D(nGyh^{-1}) = 0.642C_{Ra} + 0.604C_{Th} + 0.0417C_{k} < 80nGyh^{-1} \qquad (9)$$

As displayed in Table 5, the peak value of 305.17 nGyh^{-1} was noted in VC tiles while the least value of 81.60 nGyh^{-1} was observed in VRG. In comparing the average value of dose rate in this present investigation with the reference value of 80 nGyh^{-1} as suggested by [20-27], it was observed that the value of absorbed dose rate evaluated is higher than the recommended value of 80 nGyh^{-1}.

3.2.3. Determination of H_{ex}

The emission of gamma radiation hazard index due to the prescribed natural radioactivity was appraised by external hazard radiation and evaluated by applying equation (10) [1,20].

$$H_{ex} = (C_{Ra}/370) + (C_{Th}/259) + (C_K/4810) \qquad (10)$$

where,

C_{Ra}, C_{Th} and C_K are the concentrations of activities of ^{226}Ra, ^{232}Th and ^{40}K in $Bqkg^{-1}$ respectively. For the radiation hazard to be accepted, it has been established that H_{ex} is smaller than unity. In this study, the estimated external hazard index for the tile samples used ranges between 0.42 and 1.82 with the elevated value noted in Vironi ceramic tile of size 40X40 mm while the lowest value was also noted to be Virony Rustic tile. The estimated highest value for the tiles is 1.82. This is presented in Table 5.

3.2.4 Estimation of H_{in}

The internal hazard (H_{in}) which is defined in relation to hazard indices can be estimated by the use of equation (3) [11]:

$$H_{in} = (C_{Ra}/185) + (C_{Th}/259) + (C_K/4810) \qquad (11)$$

where C_{Ra}, C_{Th} and C_K are concentrations of natural radioactivity of ^{226}Ra, ^{232}Th and ^{40}K in Bqkg^{-1}. In order to ascertain the safety of using building materials such as tiles for decorative purposes, the internal hazard index value should be less than 1(Table 5). The estimated values for the sample in terms of internal hazard index ranges from 0.55 to 2.03 with an average value of 1.08 with the IDRIS Tile value above than the reference limit value of 1.

Table 5. Evaluation of Raeq, D, H_{ex} and H_{in} radiological parameters from China tiles

Specimen trademark	Sample size	Ra$_{eq}$(Bq/kg)	D(nGyh^{-1})	H_{ex}	H_{in}
Virony	40X40	227. 21	134.14	0.74	0.89
Virony Rustic	40X40	155.81	83.28	0.42	0.92
Virony unglazed	30X30	163.24	85.07	0.44	0.59
Virony Rustic Glass	40X40	162.62	81.60	0.44	0.55
IDRIS Tiles		603.89	276.14	1.63	1.81

	60X60				
Vironi Ceramic	40X40	677. 9	305.17	1.82	2.03
Virony Glazed	60X60	229.79	112.07	0.62	0.77
Mean values		317. 16	153.92	0.87	1.08

3.2.5 Application of Annual Effective Dose Rate

The indoors effective dose rate which is usually experienced by human beings from the internal dose rate in terms of occupancy factor is elucidated as the human occupancy level in an area closest to source of radiation is established as 80 per cent of 8760 hours per year, and the transformation factor of 0.7 Sv Gy^{-1} is usually used in the conversion of the absorbed dose rate in the air to effective dose [1, 20-27]. The yearly effective dose is evaluated using the application of equation (12).

$$AED = (0.49 \times C_{Ra}) + (0.76 \times C_{Th}) + (0.048 \times C_{K}) \times (8.76 \times 10^{-3}) \qquad (12)$$

Table 6 shows the values of the annual effective dose rate of the tiles. The value of the AEDE ranged between 0.73 and 3.14 m.Sv/y with a mean value of 1.52. It was observed that the mean value estimated in the samples is above the recommended limit value of 0.07 mSv y^{-1}.

3.2.6 Gamma Index Determination (Iγ)

Gamma-ray index is another important radiological hazard that can be used to estimate the **γ**-emission risk relating to the natural radioactive materials in the specific varieties under study. The gamma ray index representative (Iγ) is calculated using equation (13) as reported by [28].

$$I_{\gamma} = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_{K}}{3000} \qquad (13)$$

The check on the radionuclides of building decorating materials as reported by [32] is dependent on the standard level of the dose used for exemption and control. The effective doses that are greater than the standard level of 1 mSvy^{-1} should be considered in terms of protection of emission. Recommendation of dose that ranged between 0.3 and 1 mSvy^{-1} was considered as a control, which serves as the excessive gamma dose to that absorbed outdoors. The gamma emission activities index is useful in the identification of whether a criterion dose is met [32]. This gamma emission activities index describe the processes and aggregate in which the building materials

are used, with the limitation of their value indices not greater than the reference value and are dependent on the criterion dose level as displayed in Table 6. In this present study, the dose evaluated has excluded the doses at the background level that has been protected by the building materials that was used in bulk. The gamma activity index values for the samples used ranged between 0.57 (VR) and 2.37 (VC). In considering the international recommendation value of criterion of unity (1) in m.Sv, all the evaluated values are lower than the criterion value of 1. This conforms to the protection level except for IDRIS tile and Vironi Ceramic of size 40X40 mm.

3.2.7 Alpha Index Estimation (Iα)

The evaluation of alpha index is one of the cogent aspects of radiological hazard risk analysis that is concerned with the estimate of the excess of alpha emission radiation as a result of radon inhalation which originates from materials used for constructing buildings such as marble. The alpha index is calculated using equation (14) [30, 31] is:

$$I_\alpha = \frac{C_{Ra}}{200} I\alpha \qquad (14)$$

where C_{Ra} is the radium concentration in Bq/kg. If the activity of radium level in building material is far beyond 200 Bq/kg there is a probability that the radon-exhalation emanating through the material may pose concentration of indoor radon. This is shown in Table 6. The ICRP suggested the level of activity of 200 Bqm^{-3} as standard value radon for dwelling house [33]. Furthermore, if the activity level of radium is less than 100 Bqkg^{-1}, it reveals that radon-exhalation emanating through the building materials may likely not cause harm since the interior concentration is not above 200 Bqm^{-3} [34]. It has been documented that the suggested excluded value and the suggested upper limit for concentrations of radon are 100 Bqkg^{-1} and 200 Bqkg^{-1} when dealing with building materials [35] and the upper limit for concentrations of radon (Iα) is equivalent to 1 [34]. The concentration of radon estimated in tiles in this present study reveals that it ranged between 0.21 and 0.33.

Table 6: Estimated AED; I_γ and I_α

Sample Name	Sample size	Annual Effective Dose (mSv/y)	Gamma activity Index(Iγ)	Alpha Index (Iα)
Virony	40X40	1.30	0.99	0.28
Virony Rustic	40X40	0.73	0.57	0.29
Virony unglazed	30X30	0.77	0.59	0.28
Virony Rustic Glass	40X40	0.77	0.59	0.21
IDDRIS Tiles	60X60	2.83	2.14	0.33

Virony Ceramic	40X40	3.14	2.37	0.38
Virony Glazed	60X60	1.07	0.82	0.28
Mean value		1.52	1.15	0.29

4. Conclusions

The radioactivity concentration in tiles imported from China for Nigerian use have been made using Hyper germanium Detector (HPGe) and its radiological risks were estimated for seven (7) different brands tiles samples imported from China for Nigerian use in connection with the buildings related issue. The following conclusions were reached: The estimated mean values for the concentration of the radionuclides are 58.2 ± 0.52, 161.5 ± 9.45 and 455.7 ± 15.12 Bq/kg respectively and these were below international standard value. The radium-equivalent estimated was found be less than 370 Bq/kg as established by UNSCEAR [1] report except for IDDRIS tile and Vironi Ceramic (40X40 mm). The dose rate evaluated was found to be between 81.60 nGyh^{-1} to 305.17 nGyh^{-1} with a mean value of 153.92 nGyh^{-1} and higher than the recommended value as established by [12] and [1]. The estimated mean for H_{ex} and H_{in} are 0.87 and 1.08 respectively except IDRIS tiles, which has value higher than the international reference value of unity. The higher value of the effective dose rate in tile samples Vironi Ceramic (40X40 mm), Virony Glazed (60X60 mm) and IDRIS tile were observed and are greater than the international reference value of 1 mSvy^{-1}, likewise on the average level. The average values of gamma ($I\gamma$) and alpha ($I\alpha$) radiological activity indices for the tiles sample used are 1.15 and 0.29 respectively. The result showed that the tiles sample such IDRIS tile (with gamma index value of 2.37); Vironi Ceramic (with gamma index value of 2.14) and Virony Glazed (with gamma index value of 0.82) should undergo monitoring before it is being used for building related issues in order to ensure the health safety of the users. The higher values of gamma hazard index activity recorded in some tiles such as IDRIS tile, Vironi Ceramic (40X40 mm) and Virony Glazed (60X60 mm) could be associated to the nature of the sources of the raw substances used in the manufacturing of these tiles.

5. Acknowledgements

6. Conflict of Interest
The authors declare that they have no conflict of interest

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