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Comparative assessment of natural radioactivity and radiological hazards in building tiles and sharp sand sourced locally and those imported from China and India

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ABSTRACT

Background: Thirteen (13) types of building tiles and Sharp sand commonly used for building purposes were collected for their radionuclide contents analysis. Both imported and locally produced building tiles were examined. ***Materials and Methods***: The samples of tiles and sand were crushed to powder and they were prepared such that their content could be examined by the use of gamma-ray spectrometry. ***Results:*** The average activity concentration of 238U (60.61 Bq/kg), 232Th (76.55 Bq/kg) and 40K (528.40 Bq/kg) for all the samples were observed to be higher than the world recommended standard of 35, 30 and 400 Bq/kg respectively. The external and internal hazard indexes were estimated for all the building materials, the average results were found to be below the recommended limits. However, samples 3 and 6 reported internal hazard indices of 1.08 and 1.06 respectively, which are higher than the world standard. Furthermore, the estimated absorbed dose rates were observed to be within the recommended safe limits. Moreover, a comparative study of the products revealed that the results of the measured parameters from both India and China products are far higher than Nigeria products by a factor of about 1.34. ***Conclusion:*** The results obtained showed the following trend of activity concentration for the analyzed samples, India > China > Nigeria, which implies that a long exposure to both India and China products poses higher risk to the inhabitants.

Keywords: *Ionizing Building materials, tiles, sharp sand, gamma-ray spectrometry, radiological threats.*

INTRODUCTION

Tiles have become one of the widely used materials in building constructions because of the beauty and covering it provides for the cement walls and concrete floors. It is very rare in these modern days to locate a building without the beautiful touches of tiles no matter how little. Therefore, the demands for tiles have increased making the sellers to source for them from different sources both within and outside Nigeria in order to meet the recent increasing demand. Although, there is standard organization of Nigeria (SON) that is saddled with the responsibility of checking the standards of goods made both locally and those that are being imported to ascertain the quality. It has been observed that these checks are not conducted regularly and when carried out, may not test for the radioactive content of the materials being sold to consumers. This may give room for producers to compromise the standards of their products in order to make more profit. Different sectors in Nigeria have experienced compromise in the standards of their products ranging from drugs, cables,

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electronic devices and so on, therefore, standard of building materials including tiles could also be lowered by making them out of materials that can be very harmful to the users.

These harmful materials can result in different health conditions to the people that are exposed to them over a period of time. Therefore, there is need to test all types of tiles that are brought into the Nigerian market whether they are produced locally or brought into the country from the overseas. Prominent among the test recommended for building tiles is the test for their radionuclide content. This is crucial because the radiation exposures of the population can be increased appreciably by the use of building tiles that contain materials whose natural radionuclides are above the recommended standards [1-5].

Previous studies have shown that it is needful to check the radioactive content of materials that are used in buildings [6] confirmed the necessity of testing the natural radioactivity of granites used as building materials. This could be the reason to set up a database of activity concentration measurements of natural radionuclides in building material [7]. This contained about 10,000 samples of both bulk material such as bricks, concrete, cement, natural- and phosphogypsum, sedimentary and igneous bulk stones and superficial materials such as igneous and metamorphic stones used in the construction industry in most European Union Member States. It was confirmed from the assessment of commercial granites for radiological hazard that the surface exhalation rate of granites increased with the roughness of the finishes, while the thermal finish presented the highest exhalation rate according to [8].

Thus, regular studies have to be conducted to cleanse our society of building materials that could result in health hazards. Therefore, this paper is aimed at studying the level of natural radioactivity in building tiles together with the assessment of dose exposure based on the activities of the mineralogical and chemical characteristics of the constituents in order to ascertain the safety of the users and purge the market of any building tiles with the potential of exposing the users to the risk of undue radiation.

MATERIALS AND METHODS

Sample collection and preparation

Twelve (12) common building tiles in Nigeria markets were purchased from their major distributors in Orile-Iganmu international market in Lagos state, Nigeria and Sharp sand commonly used for building purposes was collected making thirteen (13) building materials in all (table 1). Initial labeling of the samples was done to ease identification. The samples were transported to the Obafemi Awolowo University material science laboratory for pulverization. The samples were first broken into pulverizable pieces using the Pascall Engineering Laboratory milling machine. After crushing each sample of tiles, the milling machine was thoroughly cleaned with the aid of blower before using it to crush another sample in order to avoid cross contamination of materials. After crushing each sample, it was immediately transferred to the "Christy and Norris" pulverizer to further grind the crushed samples into fine powder. The pulverizer was also blown with high pressure blower after each sample was grinded to prevent cross contamination of materials. The whole process was repeated until all the samples were grinded into fine powder. Furthermore, the samples in their fine powdered form were sieved with 250 μm mesh size and 1 kg of each sample was measured and separated to be packaged in thoroughly washed and very dry high density polyethylene bottles, that were duly labeled according to the brand of packaged tiles. The sieved samples were transferred to Canada in order to analyze them for radioactive contents using the High-Resolution Germanium detector. Prior to sample analysis, the samples were stored in air-tight cylindrical polythene containers of 70 mm × 75 mm dimension and kept for a minimum period of 4 weeks to allow ^{226}Ra to come into equilibrium with its short-lived progenies. Each container was completely filled to allow uniform distribution of ^{220}Rn and ^{222}Rn progenies in the sample and to avoid any accumulation in a residual surface air layer [9].

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Gamma spectrometric analysis of the selected samples

All available brands of imported and locally produced ceramic tiles and Sharp sand commonly sold in most Nigerian markets and used as building materials were purchased from different suppliers and were prepared according to International Atomic Energy Agency Technical Report Series-295 (IAEA TRS-295). Analysis of the samples was conducted at the Universiti Teknologi Malaysia Nuclear Laboratory, Department of Physics, Faculty of Science using High-Resolution Germanium detector, Canberra Lynx™ Digital Signal Analyzer (DSA), a 32 K channel integrated signal analyzer and a top-opening lead shield (4" lead, copper/tin liner) to prevent high background counts with 50 % relative efficiency and resolution of 2.1 keV at 1.33 MeV gamma energy of ^{60}Co. The Genie-2K V3.2 software locates and analyzes the peaks, subtracts background, identifies the nuclides. The efficiency curves for this analysis were corrected for the attenuation and self-absorption effects of the emitted gamma photons. CAMET and IAEA standards (DL-1a, UTS-2, UTS-4, IAEA-372 and IAEA-447) were used for checking the efficiency calibration of the system [10-14]. For the activity measurements, the samples were counted for 86,400 seconds with the background counts subtracted from the net count. The minimum detectable activity of the detector was determined with a confidence level of 95 %. The uncertainty errors were estimated keeping into account the associated errors from gamma courting emission probability and efficiency calibration standard of the system [15]. The progeny of radium, ^{214}Bi and ^{214}Pb emits gamma line 609 keV, 934 keV, 2204 keV, 1764 keV and 351 keV, 295 keV were used but the resolution of radium was from the emission of 1764 keV since it has low self-attenuation effect at high energy [16]. Since ^{232}Th cannot be directly detected, the estimated activity via its progeny ^{208}Tl and ^{228}Act using 2614.53 keV, (35.63%) 583 keV (30.3%) and 911 keV, 338 keV, 463 keV. The gamma line of 1461 keV (10.7%) was used to resolve ^{40}K. The activity concentrations were calculated according to the methods of [8].

Radium equivalent (Raeq) activity

It is important to estimate the radium equivalent activity in the analyzed samples. The knowledge of the radium equivalent concentration is used as the common index to ascertain the sum of the activities. Raeq activities are determined based on the estimation of 370 Bq/kg (10pCi^{-1}) of ^{238}U, 259 Bq/kg (7 pCi^{-1}) of ^{232}Th and 4810 Bq/kg (130pCi^{-1}) of ^{40}K each to produce the same gamma ray dose rate [27, 19]. In this study, Raeq was calculated using equation (1).

$$Raeq = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (1)$$

Where, C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K measured in Bq/kg respectively. The same external and internal gamma dose rate was produced from the radium equivalent activity. In accordance to [17] regulations, the maximum value of Ra_{eq} in building materials must be less than 370 Bq/kg, which is equivalent to 1.5 nGryy^{-1} [17, 18].

External hazard index

This index is used to characterize materials that are used for building construction purposes, so as to define an acceptable limiting value for the recommended equivalent dose according to [19]. The recommended value of the radiation dose from a construction material is 1.5 mSvy^{-1}, therefore, the value of Hex must be less than unity [20-22]. The external hazard index is also an additional criterion required for assessing the radiological suitability of building materials [17]. The Hex was calculated using equation (2).

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (2)$$

Where, $A_{Ra} \sim A_U$, A_{Th} and A_K are the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg respectively. For the radiation hazard to be acceptable, it is recommended that the Hex must be less than unity i.e. 1.

Internal hazard index

The internal hazard index is used to quantify the internal exposure of the inhabitants to radon

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and its progeny because the inhaled radon as well as the short-lived progeny presents a radiological risk to the respiratory organs [23]. This index can be estimated using equation (3).

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \quad (3)$$

According to [24-26] and [19]. For a building material to be considered safe for use, the internal hazard index must be less than 1 [21, 25].

Absorbed gamma dose rate (DR)

The contribution of the absorbed dose rate to indoor air (D_R) and the corresponding annual effective doses (AEDR) to gamma ray emission from the natural radionuclide (^{226}Ra, ^{232}Th and ^{40}K) in building materials were estimated according to equation (4) initiated by [17] and [27]. In the UNSCEAR and European Commission reports, the dose conversion coefficients were calculated for the center of a standard room. The dimensions of this room were 4 m x 5 m x 2.8 m. The thickness of the walls, floors, ceiling and the density of the structure were 20 cm and 2350 kg/m^3 (concrete), respectively. For the conversion of γ-radiation emanating from ^{226}Ra, ^{232}Th and ^{40}K, the facts of 0.436 nGyh^{-1} Bq^{-1} kg^{-1} for ^{226}Ra, 0.599 nGy/h Bq^{-1} kg^{-1} for ^{232}Th, and 0.0417 nGy h^{-1} Bq^{-1} kg^{-1} for ^{40}K were used for the estimation of the Dout. The conversion factors have been considered from literature of [28-30]. It has been reported by [31] that, "^{137}Cs, ^{90}Sr, ^{87}Rb, ^{138}La, ^{176}Lu, and ^{235}U decay series have negligible contributions to the total dose emanating from the environment. The DR was estimated using equation (4) below as given by [32, 17].

$$DR = 0.436A_{Ra} + 0.599A_{Th} + 0.0417A_K \text{ (nGy h}^{-1}\text{)} \quad (4)$$

The external absorbed dose rate (Dout)

The external absorbed dose rate (D_{out}) in nGyh^{-1} delivered by the radionuclides under investigation to the general public in the outdoor air was calculated using the equation (5) as given by [32].

$$D_{out} = 0.427C_{Ra} + 0.662C_{Th} + 0.0432C_K \text{ (nGy h}^{-1}\text{)} \quad (5)$$

Where, C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K measured in Bq/kg respectively.

The internal absorbed dose rate (Din)

The indoor exposure to gamma rays is naturally higher than the outdoor because of the predominantly earth originated materials used in building construction. When the duration of occupancy is taken into account, the indoor exposure becomes more significant. Since the investigated materials such as tiles and Sharp sand are extensively used as building materials in homes, it is important to evaluate their effects on indoor risk exposure. Considering the fact that the indoor dose contribution is 1.4 times higher than the outdoor dose contribution, the gamma dose indoor (D_{in}) in the indoor environment that is delivered by radionuclides (gamma emission from ^{226}Ra, ^{232}Th and ^{40}K) in the assessed construction materials using equation (6) by [17] and [33].

$$D_{in} = 1.4\ D_{out} \quad (6)$$

Table 1. Name of samples (Tiles and sharp sand), sizes and their country of production.

s/n	Name of Samples	Sample Description and Size	Source/Country
1	Sample 1	PNT Ceramic Tile (250x400)mm	Nigeria
2	Sample 2	PNT Vitrified Tile (400x400)mm	Nigeria
3	Sample 3	Rose Bite Tiles (600x300)mm	India
4	Sample 4	Royal Classic Ceramics (400x400)mm	Nigeria
5	Sample 5	Royal Classic Tiles (600x300)mm	Nigeria
6	Sample 6	Royal Porcelain (600x600)mm	Nigeria
7	Sample 7	Sharp sand	Nigeria
8	Sample 8	Tam Brown India (600x300)mm	India
9	Sample 9	Time Ceramics Tiles (400x400)mm	India
10	Sample 10	Virony Tiles (400x400)mm	China
11	Sample 11	Virony Tiles (300x300)mm	China
12	Sample 12	Virony Tiles (600x600)mm	China
13	Sample 13	Virony Rustic Tiles (400x400)mm	China

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RESULTS

Activity Concentrations in the Measured Building Material Samples

The specific activities of ^{238}U, ^{232}Th and ^{40}K measured in the samples are presented in table 3. The activity concentration of ^{238}U, measured in the samples ranged between 112.35 ± 0.52 and 12.03 ± 0.62 Bq/kg as detected in samples 6 and 7 respectively. For ^{232}Th, the activity concentration varied between 104.28 ± 8.12 and 33.20 ± 9.81 Bq/kg as observed in samples 3 and 7 while the highest and lowest activity concentrations of 715.64 ± 15.04 and 134.10 ± 15.26 Bq/kg were noticed in samples 12 and 1 for ^{40}K. The mean activity concentrations were estimated for the three radionuclides and they were found to be 60.61 ± 0.55, 76.55 ± 8.53 and 528.40 ± 15.14 Bq/kg for Uranium, Thorium and Potassium respectively. The estimated mean values obtained from this present study are far higher than the corresponding worldwide average values of 35, 30 and 400 Bq/kg by factors of 1.73, 2.55 and 1.32 respectively [17, 25].

The values of Raeq obtained from this present study ranged between 79.14 and 304.09 Bq/kg as presented in figure 1. The highest value was observed in sample 3 while the lowest value was noted in sample 7. The mean value for the analyzed samples was found to be 208.81 Bq/kg.

The results of the absorbed dose rates (figure 2), were observed to vary between 35.76 and 125.48 $nGyh^{-1}$ with the lowest value noticed in sample 7 while the highest value of 125.48 $nGyh^{-1}$ was observed in sample 6. This study went further to compare the highest value of absorbed dose rate with the standard safe limit of 84 $nGyh^{-1}$ recommended by [34-36], it was found to be higher by a factor of 1.49. The average value of the analyzed samples was found to be 108.5 $nGyh^{-1}$ which is still far higher than the world average of 84 $nGyh^{-1}$ by a factor of 1.29.

The results of the D_{out} presented in figure 2 varied between 38.13 and 143.32 nGy h^{-1} with an average value of 99.39 nGy h^{-1}. The lowest value was observed in sample 7 while the highest value was reported in sample 3. The mean value of 99.39 nGy h^{-1} may not be sufficient enough to influence the result of gamma activity up to 370 Bq/kg, which could increase the value of the external dose rate to 1.5 $mSvy^{-1}$ in line with the report of [17]. In a similar trend, the result of the internal absorbed dose presented in figure 2 for this present study varied between 53.38 and 200.65 nGy h^{-1} with a mean value of 139.14 nGy h^{-1}.The lowest value was noted in sample 7 while the highest value was reported in sample 3.

The estimated external hazard index obtained from this study is presented in figure 3. The estimated Hex for all the samples varied between 0.21 and 0.82 with a mean value of 0.57. The highest external hazard index was noticed in sample 3 while the lowest value was observed in sample 7. Similarly, the internal hazard index (H_{in}) was calculated and the results obtained are presented in figure 3. In the present study, the Hin was observed to vary between 0.25 and 1.08. The highest value was observed in sample 3 while the lowest value was noted in sample 7. The calculated mean value is 0.73, which is less than the recommended standard of less than or equal to 1 considered as the limit for safe building materials in the world.

Table 3. Activity concentration of ^{228}U, 232Th and ^{40}K in the Building materials

Samples	^{238}U (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)
1	36.96±0.55	62.47±8.68	134.10±15.26
2	35.64±0.56	59.83±9.27	220.54±15.21
3	94.47±0.53	104.28±8.12	785.77±15.03
4	89.82±0.52	70.79±8.34	612.23±15.10
5	81.21±0.53	67.89±8.74	624.10±15.10
6	112.35±0.52	81.54±8.28	663.07±15.08
7	12.03±0.62	33.20±9.81	254.93±15.24
8	18.68±0.60	84.79±8.34	670.45±15.32
9	79.30±0.53	80.84±8.15	351.90±15.16
10	18.68±0.53	84.79±8.68	670.45±15.07
11	68.52±0.53	68.60±8.52	634.84±15.05
12	71.32±0.53	101.99±7.92	715.64±15.04
13	68.93±0.53	94.20±8.10	531.18±15.12
mean	60.61 ± 0.55	76.55 ± 8.53	528.40 ± 15.14

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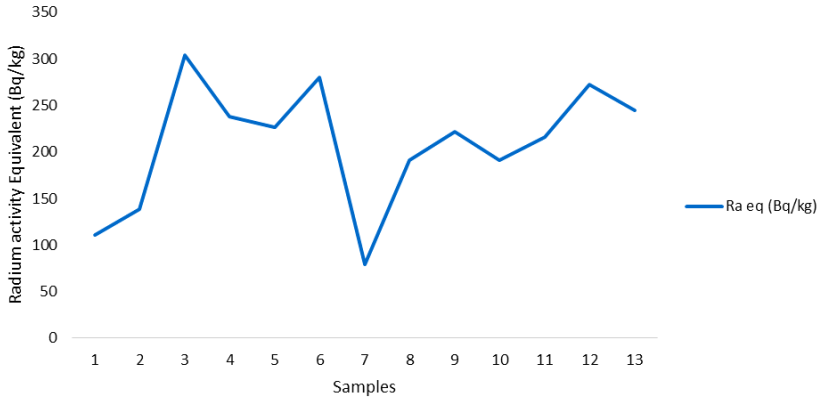


Figure 1. Radium equivalent activity.

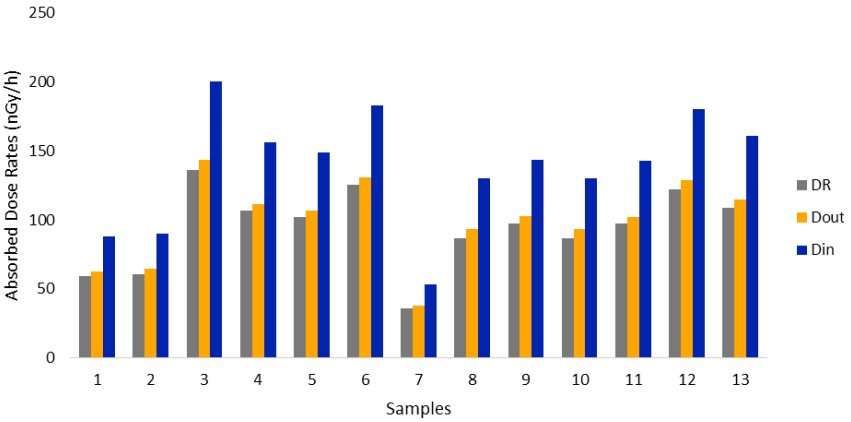


Figure 2. Graph of radium equivalent activity, absorbed dose rates.

Table 4. Comparing the Activity Concentration of Sand in the present study with some other countries of the world

Sand					
Country	^{238}U	^{232}Th	^{40}K	Raeq	Reference
India	90.27	101.67	280.71	-	[20]
Australia	3.7	40	44.4	64.32	[19]
China	39.4	47.2	573	151.02	[21]
Egypt	9.2	3.3	47.3	17.56	[22]
Pakistan	21.50	31.90	520	107.16	[23]
Nigeria	12.03	33.20	254.93	79.14	Present study
World Std*	35	30	400	-	[17]

Std* means Standard

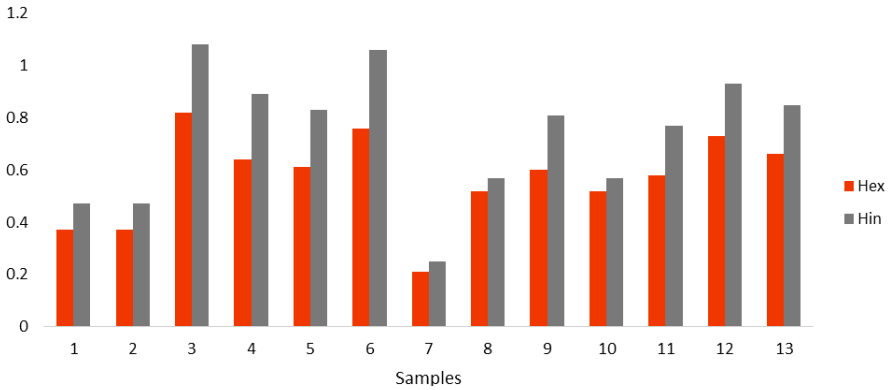


Figure 3. External and internal hazard indices.

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DISCUSSION

The distinct variation of ^{238}U, ^{232}Th and ^{40}K noticed in the different samples considered for this study (table 3), could be as a result of the differences in the geochemical and mineralogical composition of the soil from which the tiles were produced because of the varying regional geology of the different countries [25, 29]. It was also noted that ^{232}Th contributed highest to the environmental radioactivity in all the samples considered. This could be that the geological formations are composed of geological materials that are more dominant in Thorium. The results of the activity concentration of ^{238}U in sand presented in Table 4 compared the result of the present study with others. It was discovered that while the result of this study was higher than that of Egypt and Australia, it was quite lower than that of India [20], China [21] and Pakistan [23]. A trend similar to Uranium was observed for ^{232}Th, where, the activity concentrations for Thorium in India [20], Australia [19], China and Pakistan were noticed to be far higher than in Nigeria while Egypt was lower. The activity concentration of ^{40}K shown in Table 4 revealed that Nigeria is less than India, China and Pakistan but far less than Australia and Egypt [22]. The activity concentration was noted to vary from one country to another, which could be as a result of the varying mineralogical contents of the site where the raw materials were sourced. The different dominant radionuclides noticed in various regions could be as a result of the concentration of the dominant radionuclides in the geochemical compositions of the source country [37-38].

Moreover, the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in tiles produced in other parts of the world were compared with the results obtained in the present study (table 5). The following countries were randomly selected for this comparison, India, Israel, Algeria, Syria and Finland.

The results of the activity concentration obtained in this study compared with Algeria according to [25] and lower than Finland as presented in [18] and reported in table 5.

Further comparison of the results obtained from the estimation of radium activity equivalent, absorbed dose rate, external hazard index and internal hazard index were found for both sand and tiles as presented in Table 6. The radium equivalent (Raeq) of Sand in the present study is observed to be lower than the result of [20] by a factor of 3.25, as noted in Table 6. Similarly, the values of the absorbed dose rate in this investigation is far lower than the result obtained in [20] by a factor of 1.73. Likewise, both external and internal indices for Sand in [20] are far higher than the result obtained for the same material in the present study by factors of 3.31 and 3.76 respectively. In contrast to what was observed in the results of the estimated parameters for sand, building Tiles showed an opposite trend as most of the parameters considered in the study were far higher than the reference study of [20] as seen in table 6. Radium equivalent for Tiles in the present study was higher than [20] by a factor of 1.21. Likewise, both external and internal indices in this study were much higher than the compared study by [20]. However, the result of the absorbed dose rate in [20] was higher than the present study.

Table 5. The comparison of the activity concentrations in Tiles from different countries of the world.

Materials	Raeq	D_R	Hex	Hin	Reference
Sand	257.27	217.34	0.6948	0.9388	[20]
	79.14	125.48	0.21	0.25	Present study
Tiles	164.56	143.86	0.4444	0.5576	[20]
	198.79	91.03	0.55	0.744	Present study

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Table 6. Comparison of various estimated parameters with a previous study

Country	^{238}U	^{232}Th	^{40}K	Reference
India	41.88	57.39	527.53	(20)
Israel	46	48	776	(24)
Algeria	65	41	410	(25)
Syria	55	54	654	(26)
Finland	78	62	962	(18)
Nigeria	71.20	68.50	450.81	Present study

Raeq-Radium activity equivalent
DR-Absorbed Gamma Dose rate
Hex-external hazard index
Hin-internal hazard index

CONCLUSION

The natural radionuclide content together with other radiological parameters such as the internal, external hazard indices, the absorbed dose rate, the internal absorbed dose rate and the external absorbed dose rate for both building tiles and sharp sand commonly used for construction purposes in Nigeria were determined. The average activity concentration of ^{226} Ra (60.61 Bq/kg), ^{232}Th (76.55 Bq/kg) and ^{40}K (528.40 Bq/kg) in these samples of building materials are higher than the recommended safe limits of 35 Bq/kg, 30 Bq/kg and 400 Bq/kg respectively. This result showed that the raw materials used for producing tiles that are used in buildings contain very high naturally occurring radionuclides. Furthermore, the hazard indices and the absorbed dose rates were estimated, all the building materials gave results that were below the world recommended safe limit except in samples 3 and 6. The results of this study revealed that radiation exposure and its associated risk can be minimized by the choice of materials used for buildings [39-41].

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Conflicts of interest*: *Declared none.

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