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Comparative assessment of natural radionuclide content of cement brands used within Nigeria and some countries in the world

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ABSTRACT

The gamma spectrometric analysis of different brands of cement used as building and construction material in Nigeria has been carried out in this study. Samples of 12 brands of gray Ordinary Portland Cement (OPC) and 5 brands of white cement of six samples each were collected and analyzed for their radiological content using gamma spectrometry method. The average value of ^{226}Ra, ^{232}Th, and ^{40}K for OPC is 30.2 ± 10.4 Bq kg^{-1}, 24.6 ± 7.1 Bq kg^{-1}, and 251.3 ± 27.6 Bq kg^{-1} respectively and the average value for the white cement is 41.9 ± 16.7 Bq kg^{-1}, 30.1 ± 9.4 Bq kg^{-1} and 340.2 ± 37.7 Bq kg^{-1} respectively. The total average content of ^{226}Ra, ^{232}Th, and ^{40}K for all the cement brand samples are 36.1 ± 13.6 Bq kg^{-1}, 27.4 ± 8.3 Bq kg^{-1}, and 295.8 ± 32.7 Bq kg^{-1} respectively. These values obtained are lower when compared to the world average values (^{226}Ra-50 Bq kg^{-1}, ^{232}Th-50 Bq kg^{-1} and ^{40}K-500 Bq kg^{-1}) for building materials. The estimated radium equivalent activities (Ra_{eq}), representative index (Iγ), average absorbed γ-dose rate (D), the annual effective dose rate (AEDE), annual gonadal dose equivalent (AGDE) external and internal hazard indices and the Excess life cancer risk (ELCR) were lower than the recommended safe limit and are comparable with results from similar studies conducted in other countries. The evaluated mean gonadal dose equivalents of three cement brand samples were found to be higher than the world average for building material while others are less than the world average. A comparison of the average activity values obtained in Nigeria cement and other countries of the world show that those countries with history of high radionuclide solid minerals have activity concentration far above that of Nigerian cement, while these values agreed with those obtained in other countries.

1. Introduction

Man and the biota are continuously irradiated by this ionizing radiation from many sources broadly classified as either natural or artificial. Most of the total radiation exposure of man and his environment come from natural sources (UNSCEAR, 2000). This natural radiation comes from two main sources: cosmogenic radionuclides (^{3}H, ^{14}C, etc.) and long lived primordial radionuclides and their daughters (^{40}K, ^{238}U, ^{235}U, and ^{232}Th). The amount of the cosmogenic radionuclides is basically constant because of equilibrium between their rate of creation by cosmic radiation and their radioactive decay (Mikhail, 2008). Although the amount of primordial radionuclide keeps decreasing slowly with time due to radioactive decay, quite a significant amount still remain in the earth crust today due to their long half lives. These natural (primordial) radionuclides are known to be distributed in rocks and soil across the earth in varying concentrations depending on the geography and geological formations. Higher radiation levels are associated with igneous rocks, such as granite and phosphate, whereas lower levels with sedimentary rocks.

The inhomogeneous distribution of these radionuclides in geological formations like soils and rocks suggests that man made products derived from these substances will contain traces of these radioisotopes whose concentrations will be dictated by the origin of the soil and rocks they are derived from. One of such materials derived from rocks is Portland cement. Portland cement is a major building material used worldwide; it is derived from mixing natural clay, limestone and gypsum at high temperature (Kpeglo et al., 2011). Cement when mixed with fine and coarse aggregate in the right proportion is used for making concrete block—a basic building material worldwide.

The use of cement as a basic building and construction material cuts across all social and economic strata in Nigeria. It is used (when mixed with other materials such as fine sand) majorly in making concretes, sand blocks and for floors, walls, bridges and even roof finishing. A random survey on the application of cements in building revealed that over 90% of structures used as homes, offices, and commercial centers in Nigerian urban areas are constructed majorly using cement as binding material for concrete blocks (Sam and Abbas, 2001). In rural areas where clay/mud blocks and other locally sourced materials are used, it

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is common to see walls and floors plastered using cement paste. Consequently, Nigeria yearly consumption of cement stood at 10 million tons (Farai and Ejeh, 2006), and has been on the increase due to rapid structural and infrastructural development by government at all levels and the rapid increase in population and the consequent need for more homes and public structures. About two decades ago, a large proportion of cements used in Nigeria is imported (Esubiyi, 1995) while lesser proportion comes from the local industries. Today, the number of local cement industries has swollen such that more than 70% of locally consumed cement is manufactured locally. Generally, the local producing companies are located in areas where the raw materials are obtained while the imported product comes majorly from Asian and European countries. Although the brand names of some of the cements may suggest that they are manufactured in Nigeria, research has shown that some local companies only package imported cement dust using different brand names (Avwiri, 2005). The diverse sources of these cements imply that their natural radionuclide content will vary. Many research conducted and completed all over the world (Esubiyi, 1995; El-Bahi, 2004; Khalid and Hasan, 2001, etc.), have shown that natural radionuclides are present in Portland cement in varying concentration from country to country. Due to the diverse application of cement in building constructions, it could account for the bulk of indoor background radiation exposure to the populace. Furthermore, the grain size of cement is such that it is aerodynamic (Esubiyi, 1995), which could easily pass through respiratory track, or get blown by air into food and water. Consequently, the presence of radionuclide in cement does not only pose potential external radiation hazard but could also cause internal radiological contamination as well.

Some works have been done in Nigeria on building materials including cements without indication of brand name and their acceptability and covering the entire country (Avwiri, 2005; Esubiyi, 1995; Farai and Ejeh, 2006; Ibrahim, 1999). Thus, one is not sure about the nature and the distribution of the cement referred to in those works. In this study, the natural distribution of the cement brands considered is emphasized using the Nigeria map for the first time. Furthermore, this work considered brands that make up of more than 96% of those presently used in Nigeria including those used as tile adhesive. This work would serve as indicator of the indoor and outdoor radiation exposure of Nigerian people due to cement.

In the light of the diverse application of cement in Nigeria building construction industries and the fact that an average Nigerian spends about 80% of their time indoors, the knowledge of radiological content and associated hazard from cement is thus a necessity. The aim of this study is therefore to quantify the natural radionuclide content of cements available in local market in Nigeria and estimate the potential radiological hazard to the dwellers of buildings constructed from such cements and compare them with values obtained in other parts and countries of the world. The data from this study may be used by the Nigerian authority for the development and implementation of radiation protection guidelines for the use and management of cements in the country. The data in this work could also assist builders in Nigeria in considering radiological factor when making choices for cements rather than the traditional factors of cost and availability. Moreso, the data obtained in this study will add to the world data base of radioactivity content in cement as a building/construction material.

2. Materials and method

2.1. Site description

In order to collect cement brand samples that represent a fair proportion of major cement brands used in Nigeria, a survey was carried out. The survey includes visiting major cement factory, re-bagging site, suppliers' stores and sites where buildings were under construction throughout the six geopolitical zones in Nigeria. The survey revealed that the acceptability of a particular brand of cement is dictated by its availability and cost. The availability of a particular brand in a region is closely related to the proximity to the manufacturer or distributor and the cost of all local packaged cement brands is almost the same.

2.2. Sample collection

The survey revealed that 12 gray cements and 5 white cements used as tile adhesive are generally used in different parts of Nigeria. The color of each of the brands is given in Table 1. The cement brands and the places in Nigeria where they are mostly used are shown in Fig. 1. Six samples of each brand of the 17 brands totaling 102 samples were collected from major suppliers, factories, re-bagging site and building/road construction sites. The samples were collected into clean plastic containers, sealed, marked and transported to the laboratory.

2.3. Sample preparation and analysis

The samples of cements collected from construction site and major distributors and factories in locations across the country where they are mostly found were collected for radioactivity measurement. Each sample was air dried and pulverized into powdered form. 200 g of each powdered sample was put in a cylindrical polystyrene container and sealed with tapes to prevent radon permeability and left for more than four weeks in order to allow for radon and its short lived progeny to reach radioactive equilibrium. After this period, the radioactivity measurement was carried out for 7 h using a 7.6 cm × 7.6 cm NaI(Tl) detector with a resolution of 8% at 662 keV and housed in 10 cm thick lead shield to reduce background gamma radiation. The power supply and the data acquisition of the energy spectra were achieved by using an integrated spectroscopy system from Bicron. The system utilized SAMPO S100 software package from Canberra. The energy calibration of the detector was performed using IAEA standard point sources (^{109}Cd, ^{57}Co, ^{137}Cs, ^{54}Mn, and ^{22}Na) of gamma energy range between 83 keV and 1275 keV being the energy range of the radionuclide to be identified. To simulate the cement samples, 200 g of IAEA-375 reference soil was used. The radioactivity concentrations of ^{226}Ra were determined from the photopeaks of 609.32 keV (^{214}Bi), 1120.20 keV (^{214}Bi) and 352.6 keV (^{214}Pb) and that of ^{232}Th from 969.3 keV (^{228}Ac) and 583.78 keV (^{208}Tl) while the radioactivity of ^{40}K was determined from 1460.3 keV photopeak following the decay of ^{40}K as shown in Fig. 2. The background spectrum measured under the same conditions for both the standard and sample measurements, was used to correct the calculated sample activities concentration in accordance with Arogunjo et al. (2005), Kabir et al. (2009) and Zarie and Al Mugren (2010). The activity concentrations (C) in Bq kg^{-1} Bql^{-1} of the radionuclides in the samples were calculated after decay correction using the expression:

$$C_s = \frac{N_{Ey}}{\varepsilon_{Ey} \times M_s \times t_c \times P_\gamma} \left(\mathrm{Bqkg}^{-1}/\mathrm{Bql}^{-1}\right) \quad (1)$$

where C_s = Sample concentration, N_{Ey} = net peak area of a peak at energy, ε_{Ey} = Efficiency of the detector for a γ-energy of interest, M_s = Sample mass, t_c = total counting time, P_γ = Emission probability of radionuclide of interest.

2.4. Quality control and assurance

Energy calibration and efficiency calibration of the spectrometer system were performed every two months using the standard reference radionuclide sources and the reference soil. The samples of 10% were selected at random for replicate analysis (n = 3) and the relative standard deviations of results were found as <3.4%. It is therefore indicated that the detector system is relatively stable, and the results are reliable (Gang et al., 2012).

Table 1
Specific activities concentration of various Nigerian Portland and white cement brands.

Cement brand	Sample size	Cement color	^{226}Ra (Bq kg^{-1})		^{232}Th (Bq kg^{-1})		^{40}K (Bq kg^{-1})		Ra_{eq} (Bq kg^{-1})
			Range	Mean	Range	Mean	Range	Mean	
Ashaka	6	Gray	19.1–26.7	23.3 ± 9.6	17.3–23.0	20.7 ± 6.9	216.0–233.4	227.1 ± 30.6	70.4 ± 21.8
Atlas	6	Gray	28.4–44.2	39.5 ± 14.6	15.6–21.1	19.4 ± 8.3	250.2–261.3	254.8 ± 32.3	86.9 ± 28.9
Bua	6	Gray	42.0–50.2	44.7 ± 12.5	29.0–35.6	32.5 ± 11.0	266.1–282.2	275.3 ± 27.7	112.4 ± 30.4
Burham	6	Gray	33.3–42.5	38.5 ± 11.7	26.2–34.1	30.5 ± 10.6	389.0–421.9	401.9 ± 38.1	113.1 ± 29.8
Dangote (Obajana)	6	Gray	18.6–24.2	22.6 ± 6.7	13.5–18.6	19.3 ± 7.1	208.1–215.5	211.2 ± 28.3	66.6 ± 19.3
Dangote (Ibese)	6	Gray	20.8–29.2	25.6 ± 6.3	16.4–20.6	18.1 ± 5.6	206.5–210.0	208.3 ± 25.2	67.5 ± 16.2
Eagle	6	Gray	29.8–35.4	31.1 ± 8.4	22.6–29.4	26.0 ± 9.0	209.5–217.3	213.4 ± 27.9	84.7 ± 23.4
Elephant	6	Gray	21.1–34.1	28.8 ± 7.2	18.6–24.2	20.1 ± 8.6	212.6–220.1	217.1 ± 27.6	74.3 ± 21.6
Ibeto	6	Gray	32.9–40.7	36.4 ± 11.6	23.9–30.4	27.4 ± 9.0	281.1–298.8	289.4 ± 34.4	97.9 ± 27.1
Madewell	6	Gray	36.4–43.3	38.9 ± 12.3	21.3–34.6	29.3 ± 9.8	211.8–222.1	217.6 ± 29.1	97.6 ± 28.6
Sokoto	6	Gray	25.5–30.3	26.6 ± 7.9	17.0–24.2	21.3 ± 7.8	198.2–209.8	205.9 ± 24.3	72.9 ± 20.9
c	6	Gray	39.4–46.0	43.3 ± 13.4	26.2–33.7	30.1 ± 9.4	288.4–297.5	293.7 ± 34.0	109.0 ± 29.5
Sub-average of gray cement			18.6–50.2	30.2 ± 10.4	13.5–35.6	24.6 ± 7.1	198.2–421.9	251.3 ± 27.6	84.7 ± 22.7
ABS	6	White	37.1–42.8	40.2 ± 12.9	22.2–30.4	27.4 ± 9.5	301.2–324.9	318.6 ± 36.9	103.9 ± 29.3
JK	6	White	46.3–51.8	49.5 ± 14.7	24.9–34.1	29.6 ± 8.2	339.5–349.0	342.4 ± 37.8	118.2 ± 29.3
Maggen roi	6	White	45.6–47.2	46.7 ± 14.6	26.0–34.3	30.7 ± 8.8	440.4–463.9	452.9 ± 43.5	125.5 ± 32.8
Moulders	6	White	34.0–40.0	37.5 ± 10.9	28.2–35.1	32.8 ± 10.4	277.1–285.5	283.8 ± 35.6	106.3 ± 28.5
Rak white	6	White	31.5–38.4	35.5 ± 11.3	24.6–32.1	29.8 ± 9.7	294.2–308.0	303.5 ± 34.8	101.5 ± 27.9
Sub-average of white cement			31.5–51.8	41.9 ± 16.7	22.2–35.1	30.1 ± 9.4	301.2–463.9	340.2 ± 37.7	111.1 ± 33.0
Aver Age	102		18.6–51.8	36.1 ± 13.6	13.5–35.6	27.4 ± 8.3	198.2–463.9	295.8 ± 32.7	98.1 ± 28.0

3. Results and discussion

3.1. Radionuclide concentrations

The measured activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the various brands of cement considered in this work is given in Table 1. From the table the mean activity concentrations varied generally from 23.3 ± 9.6 Bq kg^{-1} to 49.5 ± 14.7 Bq kg^{-1}, from 18.1 ± 5.6 Bq kg^{-1} to 32.8 ± 10.4 Bq kg^{-1}, and from 205.9 ± 24.3 Bq kg^{-1} to 452.9 ± 43.5 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively. The mean concentrations of the gray ordinary Portland cement (OPC) brands varied from 23.3 ± 9.6 Bq kg^{-1} to 44.7 ± 12.5 Bq kg^{-1}, from 18.1 ± 5.6 Bq kg^{-1} to 32.5 ± 11.0 Bq kg^{-1} and from 205.9 ± 24.3 Bq·kg^{-1} to 401.9 ± 38.1 Bq·kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively. While the mean concentrations of the white cement used as tile adhesive varies from 35.5 ± 11.3 Bq kg^{-1} to 49.5 ± 14.7 Bq kg^{-1}, from 27.4 ± 9.5 Bq kg^{-1} to 32.8 ± 10.4 Bq kg^{-1}, and from 283.8 ± 35.6 Bq kg^{-1} to 452.9 ± 43.5 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively. The mean concentrations of the radionuclides in OPC brands were found to be low when compared with those of white cements. The variation of the mean activity concentrations of the cement brands could be attributed to the variations in the geological origin of

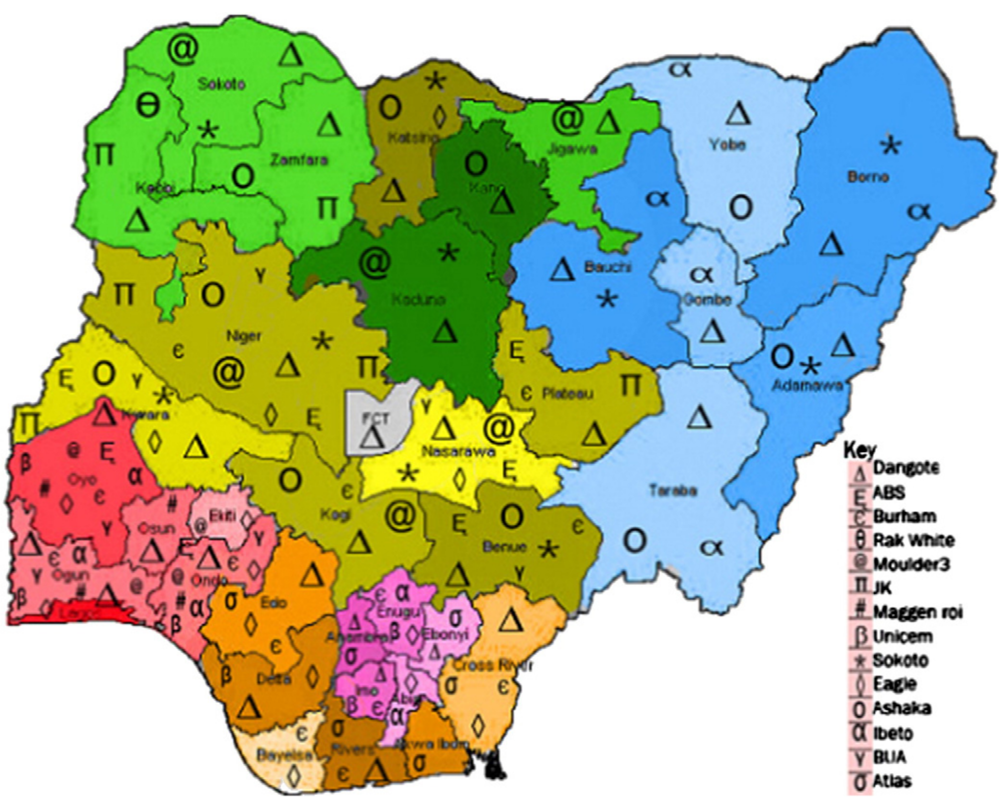

Fig. 1. Distribution of major cement brands in Nigeria.

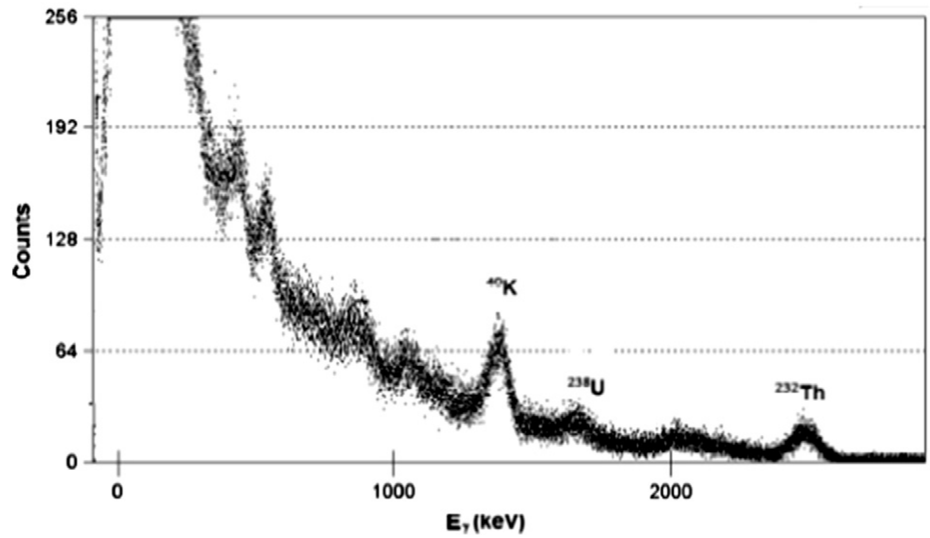

Fig. 2. A typical γ-ray spectrum obtained for one of the samples.

the raw material used in their production. All the white cement brands considered in this work were imported while only Burham and Bua are imported OPC. From the result it can be concluded that imported white cements in Nigeria tend to have higher activity concentrations when compared with the local OPC brands. Generally, the mean activity concentration of ^{40}K was the highest in all the samples when compared with the other two radionuclides. This is typical and expected from any geologically derived material due to the relative abundance of ^{40}K in the natural environment (IAEA, 2003). The range of radionuclide concentrations in the cement brands were found to be below world average of 50 Bq kg^{-1}, 50 Bq kg^{-1}, and 500 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively in building materials (UNSCEAR, 1993), while the upper limits of the range was found to be higher than the world average of 35 Bq kg^{-1}, 30 Bq kg^{-1} and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th, and ^{40}K respectively in soil (UNSCEAR, 2000).

3.2. Radium equivalent activity (Ra_{eq})

Radium equivalent activity is a single quantity that combines the radiological effects of ^{226}Ra, ^{232}Th, and ^{40}K in material used for buildings (Beretka and Mathew, 1985; Roy et al., 2000; Sam and Abbas, 2001). It is a weighted sum of activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K based on the assumption that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th, and 4810 Bq kg^{-1} of ^{40}K produce the same gamma radiation dose rates (Diab et al., 2008). The radium equivalent is calculated using equation (Zarie and Al Mugren, 2010); The percentage contribution of the three natural radionuclides is estimated using the assumption of the radium equivalent (Gang et al., 2012).

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (2)$$

where C_{Ra}, C_{Th}, and C_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K respectively. Any material whose radium equivalent activity concentration exceeds 370 Bq·kg^{-1} is not recommended for safe use as building material (Sam and Abbas, 2001). The evaluated Ra_{eq} for the cement is given in Table 2. For the gray cement, the mean Ra_{eq} varies from 66.6 ± 19.3 Bq·kg^{-1} to 113.1 ± 29.8 Bq·kg^{-1}, while for the white cement it varies from 101.5 ± 27.9 Bq·kg^{-1} to 125.5 ± 32.8 Bq·kg^{-1}. Generally, the Ra_{eq} for all the cement brands considered are less than the limit, 370 Bq·kg^{-1}. A comparison of the mean radium equivalent

Table 2
Estimated radiation hazard indices of Nigerian cement.

S/N	Cement brand	Cement origin	Iγ (Bq kg^{-1})	D (ηGy·h^{-1})	AEDE (mSv y^{-1})	AGDE (mSv y^{-1})	Hazard index		$ELCR \times 10^{-3}$	Ra_{eq} (Bq kg^{-1})
							H_{ex}	H_{in}		
1	Ashaka	Nigerian	0.5	33.1	0.04	0.17	0.19	0.25	0.14	70.4 ± 21.8
2	Atlas	Nigerian	0.6	40.9	0.05	0.28	0.23	0.34	0.18	86.9 ± 28.9
3	Bua	Imported	0.8	52.3	0.06	0.36	0.30	0.42	0.21	112.4 ± 30.4
4	Burham	Imported	0.8	53.5	0.07	0.37	0.28	0.41	0.25	113.1 ± 29.8
5	Dangote (Obajana)	Nigerian	0.5	31.2	0.04	0.22	0.18	0.27	0.14	66.6 ± 19.3
6	Dangote (Ibese)	Nigerian	0.5	31.7	0.04	0.22	0.18	0.25	0.14	67.5 ± 16.2
7	Eagle	Nigerian	0.6	39.4	0.05	0.27	0.23	0.31	0.18	84.7 ± 23.4
8	Elephant	Nigerian	0.5	34.8	0.04	0.24	0.20	0.28	0.14	74.3 ± 21.6
9	Ibeto	Nigerian	0.7	45.9	0.06	0.32	0.26	0.36	0.21	97.9 ± 27.1
10	Madewell	Nigerian	0.6	45.2	0.06	0.24	0.26	0.36	0.21	97.6 ± 28.6
11	Sokoto	Nigerian	0.5	34.1	0.04	0.24	0.20	0.27	0.14	72.9 ± 20.9
12	Unicem	Nigerian	0.8	50.9	0.06	0.35	0.29	0.41	0.21	109.0 ± 29.5
13	ABS	Imported	0.8	48.9	0.06	0.34	0.28	0.39	0.21	103.9 ± 29.3
14	JK	Imported	0.9	55.5	0.07	0.38	0.32	0.45	0.25	118.2 ± 29.3
15	Maggen roi	Imported	0.9	59.5	0.07	0.42	0.34	0.47	0.25	125.5 ± 32.8
16	Moulders	Imported	0.8	49.5	0.06	0.31	0.29	0.39	0.21	106.3 ± 28.5
17	Rak white	Imported	0.7	47.5	0.06	0.33	0.27	0.37	0.21	101.5 ± 27.9
Average			0.7	45.1	0.06	0.31	0.26	0.36	0.21	98.1 ± 24.6
World standard			≤1.0	{60 (18–93)}	1.0	0.36	≤1.0	≤1.0	0.29	370.0

Table 3
Comparison of mean radium equivalent Ra_{eq} (Bq kg^{-1}) in Nigeria brands of cements with reported values in others countries of the world.

Country	Ra_{eq} (Bq kg^{-1})	References
Ghana	90.1	Kpeglo et al. (2011)
Malaysia	188	Ibrahim (1999)
Zambia	79	Hayambu et al. (1995)
South Korea	80.8	Lee et al. (2001)
China	127.7	Xinwei (2004)
Greece	221.6	Papaefthymiou and Gouseti (2008)
India	580.1	Sonkawade et al. (2008)
Egypt	291.9	Ahmed (2005)
Lebanon	93.8	Kobeissi et al. (2008)
Turkey	246.1	Baykara et al. (2011)
Nigeria	96.4	Present work

activities in this work and those obtained from recent published work from other countries is given in Table 3. The variation in the Ra_{eq} from other countries can be attributed to the difference in the Geology and consequent geochemical constituent of the rock from which the cements were derived.

3.3. Representative level index (Iγ)

Another radiation hazard index used for the estimation of gamma radiation associated with the natural radionuclides in soil is called representative level index Iγ, (Alam et al., 1999; Ashraf et al., 2010)

$$I\gamma = C_{Ra}/150 + C_{Th}/100 + C_k/1500 \leq 1. \tag{3}$$

The representative level index (Iγ) value obtained in all the brand of cements samples ranged from 0.5 Bq kg^{-1} OPC cement to 0.9 Bq kg^{-1} in imported white cement. This shows that the (Iγ) values of the cements are within the world standard tolerable range.

3.4. Absorbed dose rate (D)

The absorbed dose rates outdoor (D) due to gamma radiations in air at 1 m above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{226}Ra, ^{232}Th and ^{40}K) were calculated based on guidelines provided by UNSCEAR (2000). The conversion factors used to compute absorbed γ-dose rate (D) in air per unit activity concentration in Bq kg^{-1} (dry weight) corresponds to 0.462 nGyh^{-1} for ^{226}Ra (of U-series), 0.621 nGyh^{-1} for ^{232}Th and 0.0417 nGy h^{-1} for ^{40}K (Ashraf et al., 2010; UNSCEAR, 2000).

$$D\left(\text{nGyh}^{-1}\right) = 0.462C_{Ra} + 0.621C_{Th} + 0.0417C_K. \tag{4}$$

The absorbed dose rate to man, outdoor varies from 31.2 nGy h^{-1} to 59.5 nGy h^{-1} with an average value of 45.1 nGy h^{-1}. This shows that the obtained data for absorbed dose rate is still within the standard limit of 60 nGy h^{-1}. This variation in absorbed dose is also an indication of the different geological sites where these cement raw materials are gotten from, which is a factor of the geological formation of the source rock.

3.5. Annual effective dose rate (AEDE)

To estimate the annual effective dose rates outdoor, one has to take into account the conversion coefficient from absorbed dose in air to effective dose (0.7 SvG y^{-1}) and outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) are used. Therefore, the annual effective dose rate (mSv y^{-1}) was calculated by the formula (UNSCEAR, 2000):

$$\begin{aligned} \text{AEDE}\left(\text{mSvy}^{-1}\right) &= \text{D}\left(\text{nGyh}^{-1}\right) \times 8760\ \text{h.y}^{-1\times}\ 0.7 \\ &\times \left(10^3\text{mSv}/10^9\right)\text{nGy} \times 0.2 \\ \text{AEDE}\left(\text{mSvy}^{-1}\right) &= \text{D} \times 1.2264 \times 10^{-3}. \end{aligned} \tag{5}$$

The worldwide annual effective dose from the natural sources of radiation in areas of normal background is estimated to be 1 mSv y^{-1} by UNSCEAR (1993). Table 2 shows the estimated annual effective dose rate obtained in the general brands of cements analyzed, the values obtained ranged from 0.04 mSv y^{-1} to 0.07 mSv y^{-1} with an average value of 0.06 mSv y^{-1}. The result shows that they are well below the world standard limit. This indicates that the cements used in Nigeria and by Nigerian are radiologically safe going by the annual effective dose rate World standard.

3.6. Annual gonadal dose equivalent (AGDE)

Since the gonads are considered as the organs of interest, together with the active bone marrow and bone surface cell, the annual gonadal

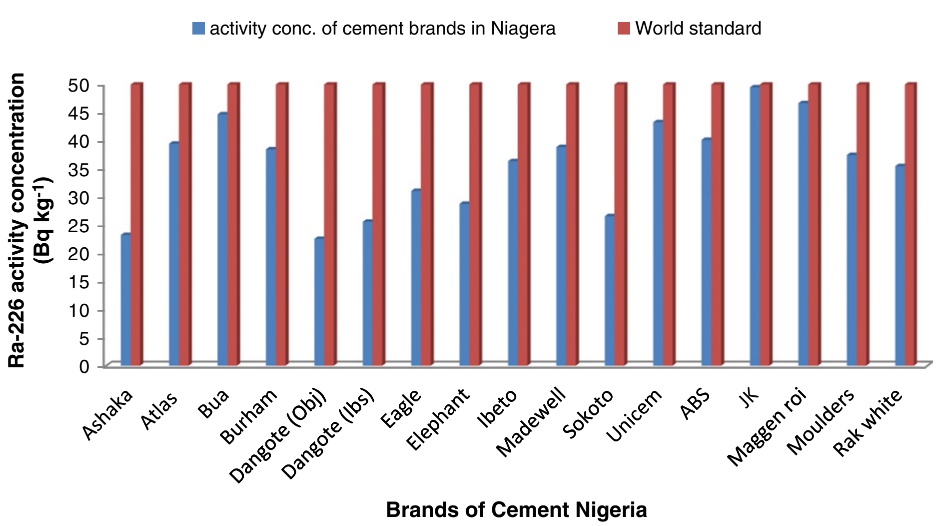

Fig. 3. Comparison of ^{226}Ra concentration in the some brands of Nigerian cement with World standard.

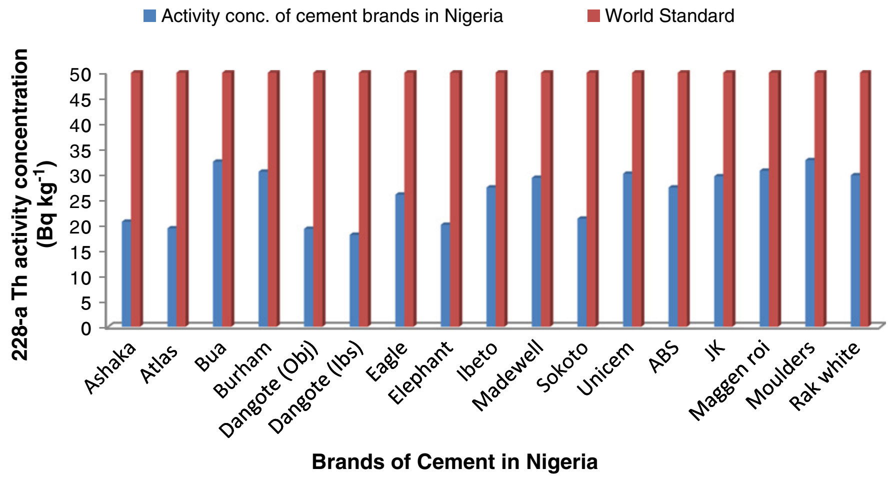

Fig. 4. Comparison of ^{228}Th concentration in the some brands of Nigerian cement with World standard.

dose equivalent (AGDE, mSv y^{-1}) due to the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K was calculated using (Al-Jundi et al., 2006):

$$AGDE = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K. \quad (6)$$

According to this model, a house is considered as a cavity with infinitely thick walls which makes it possible to make comparison of AGDEs of a house whose materials contain concentrations of ^{226}Ra, ^{232}Th, and ^{40}K with that of the world average of 35, 30, and 400 Bq kg^{-1} respectively in soil (UNSCEAR, 2000). The implication of this is that, if a building has materials whose radioactivity of the three primordial radionuclides is lower than that of the world average, such building could act as a radiation shield for the inhabitant. On the other hand, if the radioactivity is higher than the world average, the building itself could be a source of radiation to the inhabitant whose AGDE would then be greater than the world average of 0.36 mSv y^{-1} in soil. The mean AGDE of the different cement brands varies from 0.17 mSv y^{-1} to 0.42 mSv y^{-1} with a mean of 0.31 mSv y^{-1}. In all the 12 OPC brand examined, only Burham and Bua have AGDE greater than the world average value, while JK and Magen roi have AGDE values greater than the world average. These values obtained in the four cement brands show that their rock sources may be of high radioactive rock most probably from igneous rock. A further study is needed to ascertain the origin of the rock geological formations.

3.7. Internal and external gamma indices

Other indices used for testing the suitability of any material for safe use as building material are the external hazard (H_{ex}) and internal hazard (H_{in}) indices which are defined according to (Zarie and Al Mugren, 2010) as:

$$H_{ex} = {}^{C_{Ra}}\!/_{370} + {}^{C_{Th}}\!/_{259} + {}^{C_K}\!/_{4810} \quad (7)$$

$$H_{in} = {}^{C_{Ra}}\!/_{185} + {}^{C_{Th}}\!/_{259} + {}^{C_K}\!/_{4810} \quad (8)$$

where C_{Ra}, C_{Th}, and C_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K respectively.

The activity limits in terms of these limits are 1 for safe use of the material in building construction.

The external gamma indices for all the cements considered in this work are less than unity. The obtained values of the external hazard index for all the samples are given in Table 2. It varies from 0.18 for Dangote (Obajana and Ibese) cement brands, locally produced, to 0.34 for magen roi, imported white cement.

The internal hazard index (H_{in}) quantifies the internal exposure to carcinogenic radon and its short lived progeny. The values of the calculated H_{in} for the various brand of cement are also given in Table 2, and

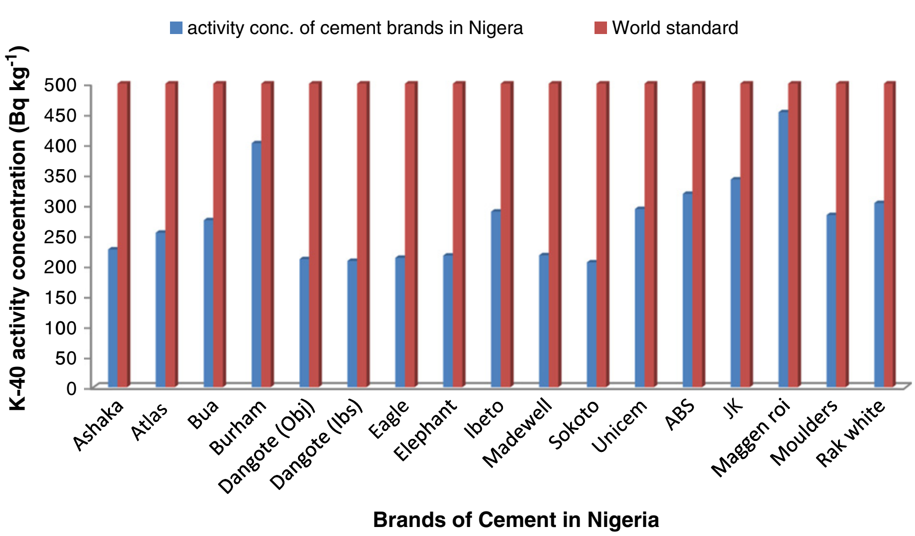

Fig. 5. Comparison of ^{40}K concentration in the some brands of Nigerian cement with World standard.

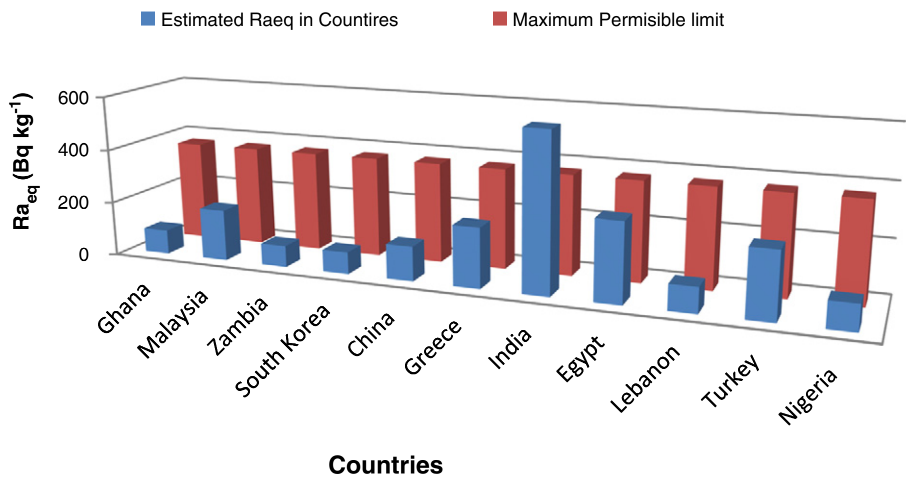

Fig. 6. A comparison of various countries cement estimated radium equivalent with Nigeria (present study).

are all less than 1. The evaluation of the internal and the external hazard indices reveal that the two brands of Dangote cement are most safe cement for building construction.

3.8. Excess lifetime cancer risk (ELCR)

Excess lifetime cancer risk (ELCR) can be defined as the excess probability of developing cancer at a lifetime due to exposure level of human to radiation. Excess lifetime cancer risk is calculated using the equation

$$ELCR = AEDE \times DL \times RF \quad (9)$$

where AEDE, DL and RF is the annual effective dose equivalent, duration of life (70 years) and risk factor (Sv^{-1}), fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public (Kaleel and Mohammad, 2012; Ramasamy et al., 2009; Taskin et al., 2009). Working in cement factory, staying in cement distribution (retail) store or living in a house build with cement as major building material may increase once chance of cancer risk. If the radioactivities in these environments are higher than the world average, it could be a source of radiation to the inhabitant whose ELCR would be greater than the world average of $0.29 mSvy^{-1}$ in such environment. From Table 2, obtained ELCR levels in all the samples analyzed are well below the world standard limit. Figs. 3–5 show the comparison of the three naturally occurring radionuclides examined in the various cement brands in Nigeria with World standard. It was observed that in all the cement sample brands analyzed, none exceeded the world permissible limit and the imported brand of cement was observed to have higher activity concentration compared to locally manufactured cements.

Fig. 6 shows the comparison of the average radium equivalent activities obtained in different countries of the world with the international standard limit. It revealed that all other countries except Indian radium equivalent were below the permissible level, which shows that cement

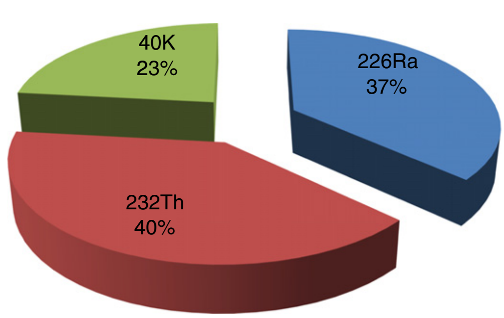

Fig. 7. Percentage contribution of the three naturally occurring radionuclides in the cement samples.

from India may contain high level of radioactivity. The reviewed work in ten different countries of the world shows that activity concentration (radium equivalent) in Malaysia, China, Greece, India, Egypt and Turkey is well above the average value obtained in Nigeria. This indicates that the areas with high background ionizing radiation due to the present of some radioactive minerals like monazite and limonite have their radioactive concentration relatively high compared to others. This is particularly observed in the result from Asian countries like China, Turkey and India where we have trace of these radioactive minerals and their background ionizing radiation above world average, thus the geological formation, i.e. the rock and geography of an area are highly responsible for the radionuclide content in raw materials used cement manufactured in the area. Fig. 7 shows the average percentage contribution of the three naturally occurring radionuclides in the cement samples. The percentage contribution of the three naturally occurring radionuclides in soil and sediment was calculated from the radium equivalent (Gang et al., 2012), the average percentage contribution of ^{226}Ra, ^{232}Th and ^{40}K in all the cement samples is 37%, 40% and 23% respectively, which shows that ^{232}Th contributes the highest radioactivity to the cement activity concentration.

4. Conclusion

The natural radionuclide content and their consequent radiation hazard indices were evaluated in gray and white cement used in Nigeria. The radionuclide concentration of cement was found to be dictated by the origin of the soil and rocks they are derived from. Although the mean specific activities of imported cement brands are higher than the locally produced ones, their total mean activities were less than the world average in building materials. The radium equivalent activities obtained for all the cement brands considered in this work were all below the criterion limit of radiation dose (1.5 mSv y^{-1}). Calculations of hazard indices show that none of the samples exceeded their recommended permissible level except the AGDE in two cement samples. The results of this study have clearly shown that cement used in Nigeria for construction and building of houses is radiologically safe and may not cause any significant health hazard to dwellers with Dangote two brands of cement most safe for use. The Nigerian cement activity concentrations are well within values obtained in other countries of the world, but a geological correlation of data obtained with cement sources (mother rocks) is recommended for further study.

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