



Radionuclides Activity and Associated Radiological Indices of Two Commonly Used Brands of Nigerian Cements

Lasun Tunde Ogundele* and Joyline Onaopemipo Adejayan
Department of Physics, University of Medical Sciences, Ondo, Nigeria
Correspondence: logundele@unimed.edu.ng

Abstract

Cement is a composite natural product that is widely used in building construction and it is reported to contain varying levels of naturally occurring radioactive materials (NORMs) that might be a source of gamma radiation exposure to human. The radioactivity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in two commonly used brands of cement (CA and CB) for building purposes were determined using gamma-ray spectrometry and the radiological indices [Radium Equivalent activity (R_{eq}), Absorbed Dose rate (DR), Annual Effective Dose rate (AED), Criteria formula (CF), Representative level index (RLI), Activity utilization index (AUI), Gamma index (I_γ), Alpha index (I_α), External hazard (H_{ex}), and Internal hazard (H_{in})] were estimated. The average values of activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K are 82.72 ± 5.73 , 3.71 ± 0.18 , $189.83 \pm 9.75 \text{ Bqkg}^{-1}$ and 84.04 ± 7.30 , 3.82 ± 0.35 , $183.50 \pm 8.86 \text{ Bqkg}^{-1}$ for CA and CB brand, respectively. The ranges of estimated values of R_{eq} were $93.72 - 113.00 \text{ Bqkg}^{-1}$, and $87 - 114.99 \text{ Bqkg}^{-1}$ for CA and CB brand. The mean values of H_{in} were 0.50 and 0.51 for CA and CB brand. The H_{in} , H_{ex} and RLI values were less than unity. None of the average values of the I_γ nor the values obtained for each samples exceeded the exemptions limit I_γ . The average absorbed dose rate in CA and CB brands were 48.44 and 48.85 nGyh^{-1} , respectively. The excess life cancer risk (ELCR) obtained for CA and CB cement brands ranged from $7.80 \times 10^{-6} - 9.55 \times 10^{-6}$ and $7.24 \times 10^{-5} - 9.79 \times 10^{-6}$. The values of radiological indices were lower than the permissible limits stipulated by International Commission on Radiological Protection (ICRP) and United National Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) indicating the safe use of such cement brands as materials for building constructions. Therefore, it can be concluded that no significant radiological health risk is envisaged by using the brands of cements under consideration for the construction building for human dwelling.

Keywords: NORMs, Cement, Gamma Spectrometry, Exposure, Radiological risks.

INTRODUCTION

Cement is a composite product that occurs as a combination of limestone, clay and shale in varying proportion (Al-Dadi *et al.*, 2014). Cements are used principally for making bricks, blocks and concrete during construction activities. They are also used for plastering and smoothing of building surfaces, and several other aggregates for physical infrastructure and building construction for human dwelling (Ozdis *et al.*, 2017). Globally, over 90 % of buildings are constructed using blocks made from cement and sand (Sam and Abbas, 2001). Cement could also be mixed with fine aggregate to produce mortar for masonry, or with sand and gravel in concrete making. Chemically, the compositions of cement materials are complex with varying levels of radionuclides and trace metals (Alkhomashi *et al.*, 2017) but the basic compounds in cement are Calcium oxide (CaO), Magnesium oxide (MgO), Silica (SiO_2), Aluminum oxide (Al_2O_3), Iron oxide (Fe_2O_3), gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), and so on. Portland cement is the most well-known brand and it is produced by heating limestone, clay and gypsum, silica, alumina at high temperature (1400°C) in order to control chemical proportions in the final products (Mujahid *et al.*, 2008; Kosmatka *et al.*, 2002). The cement production processes had been reported as a major source that

introduce large quantity of dust into the environment, causing pollution and degradation of water, air and soil around the factory (El-Taher *et al.*, 2010; Agbazue, 2013). Apart from environmental pollution from dust generated from production activities, studies had indicated that cement materials contain naturally occurring radioactive materials (NORMs), such as Uranium (^{238}U), Thorium (^{232}Th) and their decay products and the radioactive isotopes of Potassium (^{40}K) (Kpeglo *et al.*, 2011; Senthilkumar *et al.*, 2013; Al-Dadi *et al.*, 2014; Alkhomashi *et al.*, 2017). The NORMs in cement are originated from the Earth's crust from which the cement materials are extracted. Most radionuclides disintegrate with the emission of energy in form of gamma ray even at low level. The concentrations of ^{238}U , ^{232}Th and ^{40}K in cement materials could vary, depending on the geological origin of the rocks and soil of the location of formation (Alkhomash *et al.*, 2017). In the ^{238}U decay series, radium (^{226}Ra) is the most important radiological member of the series and, therefore, it is commonly considered as replacement for ^{222}Ra (Turhan, 2008; Altum *et al.*, 2017). The two principal routes of exposure of natural radionuclides in cements are external and internal exposure routes (Stoulos *et al.*, 2003; Kpeglo *et al.*, 2011; Ozdis *et al.*, 2016). Gamma radiation that originates from ^{226}Ra , ^{232}Th and their daughter radionuclides and ^{40}K are responsible for external radiation exposure while the short-lived progenies of Radon (^{222}Rn) after the decay of ^{226}Ra are the main cause of internal radiation exposure. Both external and internal radiation exposure routes have detrimental implications on human health. The inhalation of radon gas and its short-lived daughters from cement material in the indoor environment presents radiation risks for the population. Also, once radon and its progenies enter the human body, they are deposited in the respiratory tract. They can cause damage in the sensitive tissues of the lungs. Radon gas could also penetrate into the blood stream from the lungs and irradiate the whole human body through internal gaseous exchange, leading to a permanent source of internal radiation exposure (Kakati *et al.*, 2013; Ozdis *et al.*, 2017). On the average over a lifetime, people spend about 80 % in the indoor micro-environment such as houses, offices and workplaces. The total gamma radiation burden might be increased, most especially, when cement material that contains significant amounts of NORMs are used for building. Some of the radiological implications due to long term exposure and inhalation of radionuclide are acute leucopenia, chronic lung disease, anemia, necrosis of the mouth, cataract, leukemia, pancreas, hepatic, bone and kidney cancers (Ononugbo *et al.*, 2016). Therefore, determination of the activity concentrations of the radionuclides and possible radiological risks as a results of radiation hazards is fundamentally relevant from human health perspective since cements are used in a large scale for the construction of human dwellings. The studies on the measurement of the concentration of NORMs in cement materials had been carried out by many researchers in various parts of the world (Mujahid *et al.*, 2008; Kpeglo *et al.*, 2011; Trevisi, *et al.*, 2012; Senthilkumar *et al.*, 2013; Ndontchueng *et al.*, 2013; Kakati *et al.*, 2013; Majid *et al.*, 2013; Baz, 2015; Fathivand and Amidi, 2017). In most of these studies, the NORMs were measured and the associated radiological risks were assessed. In Nigeria, few studies on the determination of radionuclides concentrations and the assessment of radiological hazards from cement samples had been conducted (Ademola, 2008; Agbalagba *et al.*, 2014). The urban development, characterized by construction of buildings for human dwelling, had been increasing rapidly in the past few years and there is need to assess the radiological content of the cement materials used for such buildings. The present study attempted to contribute to the existing studies on measurement of ^{226}Ra , ^{232}Th and ^{40}K in cement products and estimate their radiological hazards. This study is of great relevance because it assessed the suitability of cement materials that are used on a large scale for the construction of human dwelling places in Nigeria. The radiological indices such as representative index level, absorbed dose rate, criterion index, annual effective dose rate, external radiation index, internal radiation index, criterion factor, gamma index, alpha index and excess life cancer risk were estimated to assess the radiation risk associated with the cements used for construction of human dwellings in Ile-Ife, southwestern, Nigeria. The results were also compared with the limits stipulated by the International Commission on Radiological

Protection (ICRP) and United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR).

MATERIALS AND METHODS

Sampling

Two different brands of cement that are commonly used for construction purposes were used in this study. The choice of the two brands were based on the availability and they are the leading brands of cements used largely for construction of building for human dwelling. For each brand, four subsamples (about 250 g) were bought at different retail shops and at different periods. The samples from each brand were coded (CA 01 - CA 12 and CB 01 - CB 13) to avoid cross contamination. The names of the brands were not stated to protect the patent right of the manufacturers. Since the cement samples were in the powdered form, sample preparation was limited to weighing without any processing such as crushing, sieving and homogenization (Ndontchueng *et al.*, 2013). Two hundred grams (200 g) of each sample was accurately weighed using electronic weighing balance and later packed into cylindrical polyvinylchloride containers (radon impermeable plastic of 28 mm height and 70 mm diameter). The samples were hermetically sealed with adhesive masking tape around its screw neck to ensure that the radon gas was confined within the sample. Prior to gamma spectroscopic measurements, the sealed samples were kept for 30 days to enable the parent radionuclides attain a state of secular equilibrium, where the rate of decay of the daughter nuclides is equal to the rate of decay of the parent.

Gamma spectroscopic measurement

The activity concentrations of the natural radionuclides in the cement samples were measured using gamma-ray spectrometry equipped with Cs(Tl) detector connected to a power supply and amplifier. The multi-channel analyzer (model URSA II) was used to record the gamma spectra. The background radiation was reduced to about 95% with a shield made of a custom Pb having a moveable cover. The analytical quality control and energy calibration of gamma spectrometry system were carried out using standard sources of known gamma ray energies and activities. The activity concentrations of radionuclides were determined in Bqkg⁻¹ using the count spectra obtained from each of the samples. The gamma ray photo peaks corresponding to energy of 1460 keV for ⁴⁰K, 352 keV (²¹⁴Pb) for ²²⁶Ra and 583 keV (²⁰⁸Tl) for ²³²Th were considered to determine the activity of ⁴⁰K, ²³²Th and ²³⁸U in each sample. All the samples were subjected to gamma spectral analysis with a counting time of 36,000 sec (10 hours) so as to achieve minimum counting error and to obtain gamma spectrum with reliable statistics (Ogundele *et al.*, 2020). All measurements were conducted in duplicate and means (\pm standard deviation) of the data sets were reported.

Estimation of radiological hazard indices

Radium equivalent

The Radium Equivalent (Ra_{eq}) was calculated on the assumption that the distribution of activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in the cement samples under consideration were not uniform. Therefore, the homogeneity with respect to the exposure to radiation due to various radionuclides in the cement samples could be estimated and it is defined by Radium equivalent activity (Ra_{eq}) in Bqkg⁻¹. The Ra_{eq} values reveal a weighted sum of the mixture of the activities of the measured natural radionuclide in the samples and it is estimated using the equation proposed by Bereka and Mathew, (1985):

$$Ra_{eq} = A_{Ra} + 43.1 A_{Th} + 0.077 A_K \quad (1)$$

where A_{Ra} , A_{Th} and A_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively. The estimation of Ra_{eq} is based on the fact that 370 Bqkg⁻¹ of ²²⁶Ra, 259 Bqkg⁻¹ of ²³²Th and 4810

Bqkg⁻¹ of ⁴⁰K produce equal amount of gamma ray dose rate. Also, it relates both the external dose and the internal dose from radon and its daughter radionuclides. The maximum permissible value of Ra_{eq} is 370 Bqkg⁻¹ which is equivalent to the effective dose of 1.5 mSvyr⁻¹ (Bereka and Mathew, 1985).

Internal and external hazard indices

The suitability of any materials to be used for construction of building for human dwelling is determined by internal hazard (H_{in}) and external hazard (H_{ex}) indices. The H_{in} quantifies the internal exposure to carcinogenic radon and its short-lived daughters (Ravisankar *et al.*, 2014; Raghu *et al.*, 2017). In this study, H_{in} and H_{ex} indices due to ²²⁶Ra, ²³²Th and ⁴⁰K in the cement samples from the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in Bqkg⁻¹ were evaluated using Equations 2 and 3 (Raghu *et al.*, 2017; Alkhomashi *et al.*, 2017; Altum, *et al.*, 2017):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (2)$$

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively. For radiation hazard to be insignificant and for a safe use of any material in building construction, the estimated values of H_{ex} and H_{in} must be less than unity.

Representative Level Index

The Representative Level Index (RLI) is index used to correlate the annual dose rate due to the external gamma radiation caused by superficial materials and acts as a screening tool for identifying materials that might pose health concerns when used in the construction building for human dwelling. The gamma radioactivity level associated with the ²²⁶Ra, ²³²Th and ⁴⁰K was calculated as a representative level index (RLI) using Equation 4 (Altum, *et al.*, 2017; Raghu *et al.*, 2017):

$$RLI = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{150} \quad (4)$$

The maximum permissible limit for RLI is unity (Raghu *et al.*, 2017):

Criterion Formula

The criterion formula (CF) is a useful comparison tool for limiting the annual radiation dose from building material. In this study, CF was determined as follows (Altum, *et al.*, 2017; Raghu *et al.*, 2017):

$$CF = \frac{A_{Ra}}{740} + \frac{A_{Th}}{520} + \frac{A_K}{9620} \quad (5)$$

Gamma Index

The gamma activity concentration (I_γ) has been defined as an index that can be used to estimate the level of gamma-ray hazard related to natural radioactivity (Bruzzi, 2006; Altum *et al.*, 2017; Al-Hamarneh, 2017). For all sorts of building materials, it is determined as follows:

$$I_{\gamma} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \quad (6)$$

For superficial building materials and natural stones, of which cement material is an example, the recommended limit for the exemption annual effective dose criterion of 0.3 mSvyr⁻¹ equivalent is I_γ ≤ 2 and the dose criterion of 1 mSv⁻¹ is met for I_γ ≤ 6 (EC, 1999; Ravisankar *et al.*, 2014).

Alpha Index

The alpha index (I_α) is another radiological index introduced to explore the exposure level due to inhalation of radon originating from disintegration of radioactive nuclei in the building materials (Al-Hamarneh *et al.*, 2017). It was determined as:

$$I_{\alpha} = \frac{A_{Ra}}{200} \quad (7)$$

In the estimation of I_α, only the activity concentration of ²²⁶Ra was used and it was based on the assumption that ²²⁶Ra nuclei in the cement samples were in secular equilibrium with ²³⁸U. The

recommended lower and upper limit of ^{226}Ra concentrations are 100 and 200 Bqkg^{-1} for which the value of $I\alpha$ must be unity (Flores *et al.*, 2008; Pantelic *et al.*, 2015)

Absorbed dose and annual effective dose rate

The absorbed dose rate (DR) in the air and the annual effective dose rate (AED) with respect to gamma-ray emission from the ^{226}Ra , ^{232}Th and ^{40}K in the cement samples were evaluated using equations developed by the United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR, 2000). In assessing the DR, the absorbed dose conversion coefficients (0.7 SvGy^{-1}) of a standard room stipulated by UNSCEAR, (2000) were considered. The volume of a standard room was taken as 56 m^3 (4 m by 5 m by 2.8 m). The thickness of the concrete walls and the density of the building were 20 cm and 2350 kgm^{-3} , respectively. Also, the DR was calculated taking into account the internal exposure rates from terrestrial gamma radiation, indoor occupancy factor of 0.8, considering the fact that the population spent, on the average, 80 % of their time indoors (Shala *et al.*, 2017). The DR and AED values were calculated by the Equations 8 and 9 (Taskin, *et al.*, 2009):

$$D(\text{Gyh}^{-1}) = 0.92A_{\text{Ra}} + 1.11A_{\text{Th}} + 0.08A_{\text{K}} \quad (8)$$

$$\text{AED} = (\mu\text{Svy}) * D(\text{Gyh}) * 8760 * 8.0 * 0.7 * \text{SvGy} \times 10^6 \quad (9)$$

where A_{Ra} , A_{Th} and A_{K} are the activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively. The worldwide average value of the absorbed dose rate in the air and annual effective dose from the natural sources of radiation were 1 mSvy^{-1} and 84.00 mGyh^{-1} . These values were adopted as reference for comparison of DR and AED values in this study.

Excess Life Cancer Risk

The excess lifetime cancer risk (ELCR) is the probability of development of cancer over a lifetime by exposure to ionizing radiation. Apart from living in a house built with cement as major building material, people working in cement factory and staying in cement distribution (retail) store may also be exposed to some level of radiation leading to increasing chance of cancer risk. For external gamma dose exposure, ELCR were determined from annual effective dose using equation (Taskin *et al.*, 2009):

$$\text{ELCR} = \text{AED} * \text{LE} * \text{RF} \quad (10)$$

where LE is the average life expectancy (70 years) and RF is the fatal cancer risk per Sievert (Sv^{-1}) which is 0.05 for public as reported in ICRP-60 for stochastic effects.

Results and Discussion

Activity concentration of ^{226}Ra , ^{232}Th and ^{40}K results.

Table 1 shows the distribution, average, standard deviation and range of activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the two brands of cements under consideration. The measured activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K ranged from 74.39 – 92.88 Bqkg^{-1} with an average value of $82.72 \pm 5.73 \text{ Bqkg}^{-1}$, from 3.35 – 4.02 Bqkg^{-1} with an average value of $3.71 \pm 0.18 \text{ Bqkg}^{-1}$, and from 174.69 – 203.63 Bqkg^{-1} with an average value $189.83 \pm 9.75 \text{ Bqkg}^{-1}$, respectively for CA brand. The measured activity concentrations of ^{226}Ra , ^{232}Th , ^{40}K ranged from 63.39 – 96.06 Bqkg^{-1} with an average value of $84.04 \pm 7.30 \text{ Bqkg}^{-1}$, from 3.35 – 4.67 Bqkg^{-1} with an average value of $3.82 \pm 0.35 \text{ Bqkg}^{-1}$, and from 166.70 – 197.64 Bqkg^{-1} with an average value of $183.50 \pm 8.86 \text{ Bqkg}^{-1}$, respectively for CB brand. The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the cement samples under investigation showed a small variation as could be observed in the values of ranges for each brand and the radionuclide. This might be as a result of the difference in composition of the crustal materials, formation processes of the cement materials and availability of different types of raw materials. The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in cements depend on the raw materials and probably the radionuclide contents in various geological

conditions, geochemical characteristics and geographical features of the areas where the raw cement materials were obtained (Hassan *et al.*, 2003). The estimated averages of $^{226}\text{Ra}/^{232}\text{Th}$, $^{226}\text{Ra}/^{40}\text{K}$, and $^{232}\text{Th}/^{40}\text{K}$ activity concentrations ratios were 9.9, 0.8 and 0.2. The $^{226}\text{Ra}/^{232}\text{Th}$ ratios are higher than unity while $^{226}\text{Ra}/^{40}\text{K}$, and $^{232}\text{Th}/^{40}\text{K}$ activity ratios were less than unity, implying that the activity concentrations of ^{40}K were greater than ^{232}Th and ^{226}Ra . It also means that cement materials were enriched with ^{40}K more than ^{232}Th and ^{226}Ra . The ^{40}K had been reported as one of the most abundant terrestrial radionuclides (Altum *et al.*, 2017) and this might be the reason why it was more than the other radionuclides in the investigated cement samples. The high concentration of ^{40}K could also be associated with its high occurrence in most geological materials. In the earth crust, radioisotope ^{40}K with high relative abundance in the enriched potash feldspar minerals (plagioclase feldspar, 39% and alkali feldspar, 12%) are geochemical sources of ^{40}K in the cement materials (Hassan *et al.*, 2013; Raghu *et al.*, 2017).

Table 1: Activity concentration of ^{226}Ra , ^{232}Th , and ^{40}K (Bqkg^{-1})

Sample	^{226}Ra	^{232}Th	^{238}U
CA 01	81.58 ± 1.05	3.67 ± 0.26	179.86 ± 2.15
CA 02	76.45 ± 2.55	3.73 ± 0.31	193.65 ± 2.15
CA 03	74.39 ± 2.21	3.57 ± 0.41	184.70 ± 2.11
CA 04	87.16 ± 2.04	3.60 ± 0.33	199.64 ± 3.26
CA 05	78.06 ± 2.22	3.57 ± 0.40	174.69 ± 3.78
CA 06	92.88 ± 3.14	4.02 ± 0.53	186.67 ± 2.08
CA 07	87.16 ± 2.44	3.95 ± 0.46	200.64 ± 3.41
CA 08	84.81 ± 1.34	3.75 ± 0.11	1.95 ± 2.37
CA 09	88.14 ± 2.24	3.85 ± 0.34	206.63 ± 3.82
CA 10	83.05 ± 1.23	3.68 ± 0.23	194.68 ± 2.37
CA 11	83.45 ± 1.33	4.00 ± 0.54	166.70 ± 2.91
CA 12	84.38 ± 2.25	4.67 ± 0.51	178.46 ± 3.60
Range	74.39 – 92.88	3.35 – 4.02	174.69 – 203.63
Average	82.72 ± 5.73	3.71 ± 0.18	189.83 ± 9.73
CB 01	83.58 ± 1.18	3.76 ± 0.76	175±0.68 ± 3.35
CB 02	65.45 ± 2.05	3.35 ± 0.13	188.65 ± 2.57
CB 03	68.39 ± 2.31	3.37 ± 0.18	186.70 ± 2.10
CB 04	77.60 ± 2.03	3.92 ± 0.30	189.4 ± 3.62
CB 05	96.06 ± 2.20	3.72 ± 0.70	174.69 ± 3.02
CB 06	94.89 ± 3.64	4.12 ± 0.53	184.67 ± 2.08
CB 07	90.16 ± 2.14	3.45 ± 0.46	187.64 ± 3.41
CB 08	84.81 ± 1.43	3.95 ± 0.21	187.65 ± 2.52
CB 09	88.04 ± 2.43	3.35 ± 0.34	193.63 ± 3.82
CB 10	82.05 ± 1,32	3.86 ± 0.43	189.65 ± 2.73
CB 11	82.16± 1.33	3.66 ± 0.23	176.67 ±2.74
CB 12	79.40 ± 1.45	3.67 ± 0.45	185.87 ± 2.05
CB 13	81.20 ± 1.24	3.90 ± 0.55	173.56 ± 2.55
Range	68.39 – 96.06	3.35 – 4.67	166.70 - 189.64
Average	84.04 ± 7.50	3.82 ± 0.35	183.50 ± 8.86

Comparison of activity levels of cement samples from different countries.

Table 2 shows the comparison of the activities of ^{226}Ra , ^{232}Th and ^{40}K determined in this study with reported values in other studies in various part of the world. Generally, the activities of ^{40}K were higher in all the studies compared to ^{226}Ra and ^{232}Th . The high concentration of ^{40}K followed the similar trend with other studies around the world. The activity concentrations recorded can be attributed to the difference in the geology and geochemical constituent of the rock from which the cements were derived.

Table 2: Comparison of activity concentrations of cement samples from different countries.

Country	^{226}Ra	^{232}Th	^{40}K	References
Turkey	34.0	15.0	220.0	Ozdis <i>et al.</i> , (2017)
Saudi Arabia	18.9	15.7	114.8	Alkhomashi <i>et al.</i> , (2017)
Egypt	35.6	43.2	82.1	El-Taher <i>et al.</i> , (2010)
Ghana	35.9	25.4	233.0	Kpeglo <i>et al.</i> , (2011)
Saudi Arabia	32.2	23.3	177.3	Baz, (2015)
Malaysia	34.7	32.9	190.6	Majid <i>et al.</i> , (2013)
Nigeria	30.2	24.6	251.3	Agbalagba, <i>et al.</i> , (2014)
Algeria	41.0	27.0	422.0	Al-Hamarneh, (2017)
Cameroon	27.0	15.0	277.0	Ngachin <i>et al.</i> , (2007)
Brazil	61.7	58.5	564.0	Malanca <i>et al.</i> , (1993)
Sicily	38.0	22.0	218.0	Rizzo <i>et al.</i> , (2001)
Nigeria	82.7	3.7	189.8	This study
Nigeria	84.0	3.8	183.3	This Study

Radiological hazard indices results

The results of the estimated radiological hazard indices consisting of Radium equivalent (Ra_{eq}), internal hazard (H_{in}), external hazard (H_{ex}), representative level index (RLI), criteria formula (CF), gamma index (I_γ), alpha index (I_α), absorbed dose rate (DR), annual effective dose rate (AED), and excess life cancer risk are presented in Table 3. The average values and ranges of estimated Ra_{eq} were 101.64 and 93.72 - 113.00 Bqkg^{-1} , and 104.63 and 87 - 114.99 Bqkg^{-1} for CA and CB brand respectively. The estimated Ra_{eq} values were lower than recommended limit value of 370.00 Bqkg^{-1} (ICRP, 1990) for any materials to be used for the construction of building for human dwelling. Thus, the investigated cement brands had their Ra_{eq} values within the recommended safety limit. The computed values of H_{in} for CA and CB brand ranged from 0.45 – 0.56 with the average value of 0.50, and 0.42 – 0.57 with an average value of 0.51, respectively. The H_{in} values were less than unity, which is the permissible dose equivalent limit set by International Commission on Radiological Protection (ICRP, 1990). This implies that the hazardous effects of radon and its short-lived progenies to the respiratory organs are negligible (UNSCEAR, 2000). The H_{in} values being less than unity also implied that the maximum concentration of ^{226}Ra is half that of the normal acceptable limit of 370 Bqkg^{-1} .

The H_{ex} values for the studied samples were less than unity, hence, the cement samples were safe to be used for construction purposes. None of the measured samples in the present work had RLI value exceeding the recommended limit value of 1.0. The calculated CF values from the sum of the three quotients for the annual radiation dose associated with the cements samples under investigation were close to the acceptable value of unity in all the samples. This indicates that the gamma radiation in the studied cement samples did not exceed the criterion limit and they could be used safely in the construction of human habitation structures. None of the average values of

the I_γ nor the values obtained for each samples exceeded the exemptions limit I_γ ($I_\gamma \leq 2$) for annual effective dose criterion of 0.3 mSvy^{-1} (EC, 1999). The results of the I_γ indicated that the exposure level due to radon inhalation originating from the radionuclide in cement samples are insignificant. From the results of the I_α indices, the I_α values and the computed averages are well below the recommended maximum value of unity. Similarly, the calculated values of I_γ and I_α further implied that the hazardous radiation effects of the radionuclides in the cement brands under consideration were insignificant and confirmed the safe use in building construction for human dwelling. The calculated absorbed gamma dose rate due to ^{226}Ra , ^{232}Th and ^{40}K ranged from $44.29 - 53.19 \text{ nGyh}^{-1}$ and $41.70 - 4.09 \text{ nGyh}^{-1}$ in CA and CB brands, respectively. The average absorbed dose rate in CA and CB brands were 48.44 and 48.85 nGyh^{-1} , respectively. The average absorbed doses from both samples of the brands of cement were lower than the worldwide average value of 84 nGy h^{-1} stipulated for building materials (UNSCEAR, 1993; UNSCEAR, 2008). The average values and range of annual effective dose rate of CA and CB brand of cement samples under consideration were $0.25, 0.21 - 0.27 \text{ }\mu\text{Svy}^{-1}$ and $0.25, 0.21 - 0.28 \text{ }\mu\text{Svy}^{-1}$. The annual effective dose rate values were lower than the world average annual effective dose received from terrestrial gamma radiation of $70 \text{ }\mu\text{Svy}^{-1}$ (Amrani, 2001). The results of the I_α and I_γ values were also less than unity. The computed values of excess life cancer risk (ELCR) obtained for CA and CB cement brands ranged from $7.80 \times 10^{-6} - 9.55 \times 10^{-6}$ with the average values of 8.60×10^{-6} , and $7.24 \times 10^{-5} - 9.79 \times 10^{-6}$ with the average values of 8.71×10^{-6} , respectively. The world average limit values of ELCR for outdoor terrestrial gamma radiation is 2.4×10^{-5} (ICRP, 1990). Therefore, the calculated values of ELCR were lower than the world average values in the samples for both brands of cement. The relevance of the calculated ELCR being lower than the world average is that the risk of dying because of cancer due to the gamma radiation from ^{226}Ra , ^{232}Th and ^{40}K in the brand of cement is very low and there might not be any significant radiological health risks among the people. Therefore, the cement products under investigation did not pose a significant radiological hazard when used for building constructions.

Table 3: Results of radiological hazard indices

Codes	R_{eq}	H_{in}	H_{ex}	RLI	CF	I_γ	I_α	AD	AED	ELCR x (10^6)
CA 01	100.66	0.49	0.27	0.70	1.22	0.89	0.41	47.46	0.24	8.46
CA 02	96.69	0.47	0.26	0.68	1.09	0.92	0.38	45.71	0.23	8.03
CA03	93.72	0.45	0.25	0.65	1.27	0.88	0.37	44.29	0.22	7.80
CA 04	107.71	0.53	0.29	0.75	1.14	0.97	0.44	50.84	0.26	9.04
CA 05	96.62	0.47	0.26	0.67	1.35	0.86	0.39	45.57	0.23	8.10
CA 06	113.00	0.56	0.31	0.70	1.27	0.95	0.46	48.27	0.27	9.55
CA 07	108.26	0.53	0.29	0.75	1.24	0.98	0.44	51.89	0.26	9.07
CA 08	105.24	0.51	0.28	0.73	1.29	0.95	0.42	49.67	0.25	8.82
CA 09	109.23	0.53	0.30	0.76	1.21	0.99	0.44	51.56	0.26	9.15
CA 10	103.30	0.50	0.23	0.72	1.22	0.94	0.42	48.77	0.25	8.65
CA 11	102.48	0.50	0.28	0.71	1.10	0.88	0.42	51.19	0.25	8.63
CA 12	94.77	0.46	0.26	0.66	1.09	0.90	0.38	44.80	0.23	7.90
Min	93.72	0.45	0.25	0.65	1.09	0.86	0.37	44.29	0.22	7.80
Max	113.00	0.56	0.31	0.76	1.35	0.99	0.46	53.19	0.27	9.55
Mean	102.64 ± 7.43	0.50	0.28	0.72	1.20	0.93	0.41	48.44	0.25	8.60
CB 01	87.59	0.42	0.24	0.61	1.14	0.87	0.34	41.47	0.21	7.24
CB 02	97.81	0.47	0.26	0.68	1.40	0.91	0.39	46.19	0.23	8.14
CB03	114.83	0.57	0.31	0.79	1.38	0.92	0.48	53.97	0.28	9.79
CB 04	114.99	0.57	0.31	0.80	1.32	0.95	0.47	54.09	0.28	9.74

CB 05	110.31	0.54	0.30	0.77	1.24	0.98	0.45	52.04	0.27	9.30
CB 06	104.91	0.51	0.28	0.73	1.28	0.93	0.42	49.46	0.25	8.80
CB 07	107.74	0.53	0.29	0.75	1.20	0.96	0.44	50.83	0.26	9.08
CB 08	102.17	0.50	0.28	0.71	1.20	0.92	0.41	48.21	0.24	8.55
CB 09	101.00	0.49	0.27	0.70	1.16	0.88	0.41	47.60	0.24	8.50
CB 10	98.96	0.48	0.27	0.69	1.19	0.90	0.40	46.71	0.24	8.28
CB 11	100.14	0.49	0.27	0.70	1.22	0.87	0.41	47.17	0.24	8.41
CB 12	102.01	0.50	0.28	0.71	1.23	0.85	0.42	47.99	0.25	8.61
CB 13	104.80	0.51	0.28	0.73	1.00	0.90	0.42	49.33	0.25	8.78
Min	87.60	0.42	0.24	0.61	1.00	0.85	0.34	41.47	0.21	7.24
Max	114.99	0.57	0.31	0.80	1.40	0.98	0.48	54.09	0.28	9.79
Mean	103.63 ±5.64	0.51	0.28	0.72	1.23	0.91	0.42	48.85	0.25	8.71

CONCLUSION

The activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in samples of two major brand of cements had been determined by employing gamma-ray spectroscopy. Among the measured radionuclides, the concentrations of ^{40}K were higher than ^{226}Ra and ^{232}Th . The level of natural radioactivity of ^{226}Ra , ^{232}Th and ^{40}K were lower than the international recommended limit. The estimated radiological hazard indices were also lower than the international recommended limit set by ICRP and UNSCEAR. It can, therefore, be concluded that the cements products investigated were radiologically safe, and hence could be used for construction of building for human dwellings. It is important to point out that these values are not the representative values for all the regions in the country but only for the part of the country where the samples were obtained.

ACKNOWLEDGEMENT

The authors appreciate the efforts of Mr C. Iyanuoluwa for his assistance during the sampling activities.

Funding: This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sector.

Declaration of Conflict of interest: The authors declared that no conflict of interest exists

Disclaimer

The products used for this study are predominantly for research purposes. There is absolutely no conflict of interest between the authors and producers of the products because the products were only used for advancement of knowledge

REFERENCES

- Ademola, J. A., (2008). Assessment of natural radionuclide content of cements used in Nigeria. *Journal of Radiological Protection*, 28:581–588.
- Agbalagba, E. O., Osakwe, R. O. A, and Olarinoye, I. O., (2014). Comparative assessment of natural radionuclide content of cement brands used within Nigeria and some countries in the world. *Journal of Geochemical Exploration*, 142:21–28.
- Agbazue, V. E., (2013). Analysis of source contribution to particulate matter pollution in Benue Cement Factory. *Sacha Journal of Environmental Studies*, 31:16–22.
- Al-Dadi, M. M., Hassan, H. E., Sharshar, T., Arida, H. A., and Badran, H. M., (2014). Environmental impact of some cement manufacturing plants in Saudi Arabia. *Journal of Radioanalytical and Nuclear Chemistry*, doi:10.1007/s10967-014- 3383-8.
- Al-Hamarneh, I. F., (2017). Radiological hazards for marble, granite and ceramic tiles used in buildings in Riyadh, Saudi Arabia. *Environmental Earth Science*, 76:516–526.

- Alkhomashi, N., Almasoud, F. I., Alhorayess, O., Alajayan, T. M., Alsalamah, A. S., Alssalim, Y. A., and Ababneh, Z. Q., (2017). Assessment of radioactivity and trace elements of cement produced in Saudi Arabia. *Environmental Earth Science*, 76:280–288
- Altum, M., Sezgin, N., Nemlioglu, S., Karakelle, B., Can, N., and Temelli, U. E., (2017). Natural radioactivity and hazard-level assessment of Portland cements in Turkey. *Journal of Radioanalytical and Nuclear Chemistry*, doi:10.1007/s10967-014-3383-8.
- Amrani, D., and Tahtat, M., (2001). Natural radioactivity in Algerian building materials. *Applied Radiation and Isotopes*, 54:687–689
- Baz, S. S., (2015). Evaluation of natural radioactivity and the associated radiation hazards in local cement used in Saudi Arabia. *Life Sciences Journal*, 12(2):189–195.
- Bereka, J., and Mathew, P J., (1985). Natural radioactivity of Australian building materials, wastes and by-product. *Health Physics.*, 48, 87–95.
- Bruzzi, R. S., (2006). Natural radioactivity and radon exhalation in building materials used in Italian dwellings. *Journal of Toxicology and Environmental Health, Part A: Current Issues*, 88:158–170.
- EC (1999). European Commission on Radiological Protection and Principles Concerning the Natural Radioactivity of Building Materials. Directorate-General Environment. Nuclear Safety and Civil Protection. 112.
- El-Taher, A., Makhluif, S., Nossair, A., and Abdel Halim, A., (2010). Assessment of Natural Radioactivity levels and Radiation hazards due to cement industry. *Applied Radiation and Isotopes*, doi:10.1016/j.apradiso.2009.09.001.
- Fathivand, A. A. and Amidi, J., (2017). Assessment of natural radioactivity and the associated hazards in Iranian cement. *Radiation Protection and Dosimetry*, 124(2):145 – 147.
- Flores, O. B., Estrada, A. M., Suarez, R. R., Zerquera, J. T., and Perez, A. H., (2008). Natural Radionuclide content in Building materials and gamma dose rate in dwellings in Cuba. *Journal of Environmental Radioactivity*. 99(12):1834-7.
- Hassan, N. M, Mansour N. A, and Hassan M. F., (2013). Evaluation of Radionuclides Concentration and Associated Radiological hazard indexes in building materials used in Egypt. *Radiation Protection Dosimetry*, 157:214–220.
- ICRP-60., (1990) Recommendations of the International Commission on Radiological Protection (ICRP). Pergamon Press, Oxford.
- Kakati, R. K., Kakat, L., and Ramachandran, T. V., (2013). Measurement of uranium, radium and radon exhalation rate of soil samples from Karbi Anglong district of Assam, India using EDXRF and Can technique method. *APCBEE Procedia*, 3:186–191.
- Kosmatka, S., Kerkhoff, B., and Panerese W., (2002). Design and Control of Concrete Mixtures, 14th edition. Portland Cement Association, Skokie.
- Kpeglo, D. O., Lawlubi, H., Faanu, A., Awudu, A. R., Deatanyah, P., Wotorchi, S. G., Arwui, C. C., Emi-Reynolds, G., and Darko, E. O., (2011). Natural Radioactivity and its Associated Radiological hazards in Ghanaian cement. *Journal of Environmental Earth Sciences*, 3(2):160–166.
- Majid, A. A., Ismail, A. Fand Yasir, M. S., Yahaya, R., and Bahari, I., (2013). Radiological dose assessment of naturally occurring radioactive materials in concrete building materials. *Journal Radioanalytical and Nuclear Chemistry*, 297:277–284.
- Malanca, A., Pessina, V., and Dallara, G., (1993). Radionuclide content of building materials and gamma-ray dose rates in dwellings of Rio- Grande Do-Norte Brazil. *Radiation Protection Dosimetry*, 48:199–203
- Mujahid, S. A., Rahim, A., Hussain, S., and Farooq, M., (2008). Measurement of natural radioactivity and radon exhalation rates from different brands of cement used in Pakistan. *Radiation Protection Dosimetry*, doi:10.1093/rpd/ncm49.

- Ndontchueng, M. M., Simo, A., Nguelem, E. J. M., Njinga, R. L., Beyala, J. F., and Kryeziu, D. (2013). Preliminary investigation of Naturally Occurring Radionuclide in some six representative Cement types commonly used in Cameroon as building material. *International Journal of Science and Technology*, 3(10):365–371.
- Ngachin M., Garavaglia M., Giovani C., Kwato, N. M. G., and Nourreddine. A., (2007). Assessment of Natural Radioactivity and associated radiation hazards in some Cameroonian building materials. *Radiation Measurement*, 42:61–67.
- Ogundele, L. T., Ayeku, P. O., Inuyomi, S. O., Ogunsakin, O. M., Oladejo, O. F., and Adejoro, I. A.. (2020). Assessment of Naturally Occurring ^{40}K , ^{232}Th and ^{238}U and their associated Radiological hazard indices in soils used for building in Ondo West Local Government area, Southwestern, Nigeria. *EQA – International Journal of Environmental Quality*, 37:11–21.
- Ononugbo, C. P., Avwiri, G. O., and Tutumeni, G., (2016). Measurement of Natural Radioactivity and Evaluation of Radiation Hazards in Soil of Abua/Odual Districts using Multivariate Statistical Approach. *British Journal of Environmental Sciences*, 4(1):35–48.
- Ozdis, B. E., Ozdis, N. F., Cam, B., and Ozturk, C. (2017). Assessment of Natural Radioactivity in cements used as Building Materials in Turkey. *Journal of Radioanalytical and Nuclear Chemistry*, 31(1):307–316.
- Pantelic, G. K., Todorovic, D. J., Nikolic, J. D., Rajacic, M. M., Jankovic, M. M., and Sarap, N. B., (2015). Measurement of Radioactivity in Building Materials in Serbia. *Journal of Radioanalytical and Nuclear Chemistry*, doi:10.1007/s10967-014-3745-2.
- Raghu, Y., Ravissankar, R., Chandrasekran, A. P., and Venkatraman, V., (2017). Assessment of Natural Radioactivity and Radiological hazards in Building Materials used in the Tiruvannmalali District, Tamilnadu, India, using a Statistical Approach. *Journal of Taibah University of Science*, 11:523–533.
- Ravisankar, R., Vanasundari, K., Suganya, M., Raghu, Y., Rajalakshmi, A., Chandrasekaran, A., Sivakumar, S., Chandramohan, J., Vijayagopal, P., and Venkatraman, B. (2014). Multivariate Statistical Analysis of Radiological data of Building Materials used in Tiruvannamalai, Tamilnadu, India. *Applied Radiation and Isotopes*, 85:114–127.
- Rizzo, S., Brai, M., Basile, S., Bellia, S., and Hauser, S., (2001). Gamma activity and geochemical features of Building Materials: Estimation of gamma dose rate and indoor radon levels in Sicily. *Applied Radiation and Isotopes*, 55:259–265.
- Sam, A. K., and Abbas, N., (2001). Assessment of Radioactivity and the Associated hazards in local and imported cement types used in Sudan. *Radiation Protection Dosimetry*, 93:275–277.
- Senthilkumar, G., Ravisankar, R., Vanasundari, K., Vijayalakshmi, I., Vijayagopal, P., and Jose, M. T., (2013). Assessment of Radioactivity and the Associated hazards in local Cement types used in Tamilnadu, India. *Radiation Physics and Chemistry*, 88:45 – 48.
- Shala, F., Xhixha, G., Xhixha, M. K., and Hasani, F., (2017). Natural Radioactivity in Cement and raw materials used in Albanian Cement industry. *Environmental Earth Science*, 76:670–677.
- Stoulos, S., Manolopoulou, M., and Papastefanou, C., (2003). Assessment of Natural Radiation exposure and radon exhalation from building materials in Greece. *Journal of Environmental Radioactivity*, 69:225 –240.
- Taskin, H., Karavus, M., Ay, P., Topuzoglu, A., Hidiroglu, S., and Karahan, G., (2009). Radionuclide concentrations in soil and lifetime cancer risk due to gamma radioactivity in Kirklareli, Turkey. *Journal of Environmental Radioactivity*, 100(1):49–53.
- Trevisi, R., Risica, S., DAlessandro, M., and Paradiso, D., (2012). Natural radioactivity in building materials in the European Union: a database and an estimate of radiological significance. *Journal of Environmental Radioactivity*, 105:11–20.
- Turhan, S., (2008). Assessment of the Natural Radioactivity and radiological hazards in Turkish cement and its raw materials. *Journal of Environmental Radioactivity*, 99:404–414.

- UNSCEAR, (1993). United Nations Scientific Committee on the Effects of Atomic Radiation. Sources and effects of ionizing radiation, Report on the Effects of Atomic Radiation Report. www.unscear.org/unscear/en/publications/1993.html ISBN: 92-1-142200-0
- UNSCEAR, (2000). United Nations Scientific Committee on the Effects of Atomic Radiation. Exposures from natural radiation sources. www.unscear.org/unscear/en/publications/2000_1.html ISBN: 92-1-142238-8
- UNSCEAR (2008). Sources and effects of ionizing radiation, Report on the Effects of Atomic Radiation Report. www.unscear.org/docs/reports/2008/09-86753_Report_2008_GA_Report_corr2.pdf. ISBN: 978-92-1-142274-0