Radiological health assessment of natural radioactivity in the vicinity of Obajana cement factory, North Central Nigeria

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ABSTRACT

Measurements of activity concentrations of natural radionuclides in and around Obajana cement factory, North Central Nigeria have been carried out in this study to determine the activity levels of natural radionuclides in different environmental matrices in order to assess the radiological health hazards associated with the use of these matrices by the local population. A low-background Pb-shielded gamma spectroscopic counting assembly utilizing NaI (Tl) detector was employed for the measurements. The results show that sediment samples have the highest activity concentrations of all the radionuclides relative to soil, farmland soil, and rock samples. The radium equivalent activity and indoor gamma dose rates together with the corresponding annual effective indoor doses evaluated were found to be lower than their permissible limits. It suffices to say, that contrary to age-long fear of radiation risks to the population in the vicinity of the cement factory, no excessive radiological health hazards either indoors and/or outdoors is envisaged. Therefore, the environmental matrices around the factory could be used without any restrictions.

Key words: Building material, marble rocks, North Central Nigeria, Obajana, sediment, terrestrial radionuclides

Introduction

Naturally occurring radionuclides are known to be present in varying proportions in rocks and soil of different geological formations around the world. Their concentrations and associated external doses in different environments depend on the geology and geographical conditions in such environments. Due to weathering and other environmental processes, radionuclides in rock and soil may accumulate in sediment and dissolve into drinking water, thereby leading to human exposure. Exposure to natural terrestrial radiation in the outdoor environment is predominantly from radionuclides that are present in the upper 30 cm layer of the soil. Human exposure pathways include: Root uptake from contaminated soil, direct ingestion of radionuclides deposited on plant leaves, consumption of water animals, ingestion of contaminated water, inhalation of soil dust, and the direct exposure to gamma ray emitted from primordial radionuclides in the indoor and outdoor environments, etc.

There are approximately 340 naturally occurring nuclides in the earth’s crust. Of these, about 70 are radionuclides. Many of the naturally occurring radionuclides decay into daughters which are radioactive leading to radioactive decay chains. All nuclides in a lineage constitute a decay chain or series. There are four of such radioactive decay chains in nature, namely, uranium, thorium, actinium, and neptunium. Among these radioactive series, only uranium and thorium contribute appreciably to population exposure. The longest lived member of uranium series is $^{238}$U, which constitutes about 99.27% of natural uranium with half-life of $4.47 \times 10^{9}$ years. Its concentration in common rock is about 4.7 ppm and decays through alpha particle emission. The thorium series is headed by $^{232}$Th...
In the present study, the activity which are present at trace levels in all rocks, soil, farmland soil, and sediment samples were collected around Obajana cement factory. The marble rocks analyzed in this study form the main components of Dangote cement, which is utilized as building material in all parts of Nigeria and some neighboring countries. Obajana cement factory is reputed as the largest cement factory in West African sub-region. The location of the study area in Nigeria is shown Figure 1. The samples were collected and processed following standard procedures. Each sample was collected in a polythene bag, carefully labeled at the collection site and brought to the laboratory where each was air dried at room temperature until constant weight was attained. All the rock samples were identified at the Geology Department, University of Ibadan. They were then grounded to powder in order to have the same matrix as the standard samples. Two hundred gram each of the grounded samples were sealed in air tight plastic containers to avoid the escape of $^{220}\text{Ra}$ and $^{226}\text{Rn}$ and were stored for more than 4 weeks before measurement so as to attain secular equilibrium. Gamma spectrometric measurements were made with a low-background Pb shielded 760 mm $\times$ 760 mm NaI (Tl) detector (Model 802 series by Bicron) optically coupled to a photomultiplier tube. The detector is interfaced with a Canberra series 10 plus multi-channelled analyzer (model: 1104). The multi-channelled analyzer (MCA) is a complete system having all functions needed for spectroscopic analysis. The energy resolution (FWHM) of the detector is about 8% at 0.662 MeV. The measuring assembly used in this work consists of the MCA coupled with a 50 $\Omega$ coaxial cable to the detector, which is shielded from background radiation with a 10 cm-thick lead castle with copper lining made by Canberra Inc. It has a positive signal output. Two forms of calibration were carried out in this work. The first, which is known as energy calibration, was to convert channel numbers to $\gamma$-rays energy in MeV. The second, known as efficiency calibration, was to convert the area under a photopeak to concentration of the radionuclides in unit of Bq/kg. The calibration of the detector system to determine the equation relating energy to channel number was done by measuring mixed standard sources of known concentrations of terrestrial radionuclides in marble rock, soil, farmland soil, and sediment around Obajana cement factory, North Central Nigeria have been measured to determine the outdoor radiation exposure and the radiological implications of using the marble as a component of cement manufacturing. The results of studies such as this are of great interest in an environmental radiological protection study, especially for Nigeria, where marble is widely used as a building material. To the best of our understanding, no such study has been carried out in this area. The study, therefore, will form a baseline on which any extended radiological research in this area can be compared.

Materials and Methods

Sample collection and measurement

In the present study, marble rocks, soil, farmland soil, and sediment samples were collected around Obajana cement factory. The marble rocks analyzed in this study form the main components of Dangote cement, which is utilized as building material in all parts of Nigeria and some neighboring countries. Obajana cement factory is reputed as the largest cement factory in West African sub-region. The location of the study area in Nigeria is shown Figure 1. The samples were collected and processed following standard procedures. Each sample was collected in a polythene bag, carefully labeled at the collection site and brought to the laboratory where each was air dried at room temperature until constant weight was attained. All the rock samples were identified at the Geology Department, University of Ibadan. They were then grounded to powder in order to have the same matrix as the standard samples. Two hundred gram each of the grounded samples were sealed in air tight plastic containers to avoid the escape of $^{220}\text{Ra}$ and $^{226}\text{Rn}$ and were stored for more than 4 weeks before measurement so as to attain secular equilibrium. Gamma spectrometric measurements were made with a low-background Pb shielded 760 mm $\times$ 760 mm NaI (Tl) detector (Model 802 series by Bicron) optically coupled to a photomultiplier tube. The detector is interfaced with a Canberra series 10 plus multi-channelled analyzer (model: 1104). The multi-channelled analyzer (MCA) is a complete system having all functions needed for spectroscopic analysis. The energy resolution (FWHM) of the detector is about 8% at 0.662 MeV. The measuring assembly used in this work consists of the MCA coupled with a 50 $\Omega$ coaxial cable to the detector, which is shielded from background radiation with a 10 cm-thick lead castle with copper lining made by Canberra Inc. It has a positive signal output. Two forms of calibration were carried out in this work. The first, which is known as energy calibration, was to convert channel numbers to $\gamma$-rays energy in MeV. The second, known as efficiency calibration, was to convert the area under a photopeak to concentration of the radionuclides in unit of Bq/kg. The calibration of the detector system to determine the equation relating energy to channel number was done by measuring mixed standard sources of known concentrations of terrestrial radionuclides in marble rock, soil, farmland soil, and sediment around Obajana cement factory, North Central Nigeria have been measured to determine the outdoor radiation exposure and the radiological implications of using the marble as a component of cement manufacturing. The results of studies such as this are of great interest in an environmental radiological protection study, especially for Nigeria, where marble is widely used as a building material. To the best of our understanding, no such study has been carried out in this area. The study, therefore, will form a baseline on which any extended radiological research in this area can be compared.

Materials and Methods

Sample collection and measurement

In the present study, marble rocks, soil, farmland soil, and sediment samples were collected around Obajana
radionuclides with well-defined energies within the energy range of interest. The detection efficiency calibration and the quality control of the system were carried out using standard soil and sediment samples. The standard soil sample for gamma source was prepared from Rockydne laboratories, Canoga Park, California, USA which is traceable to a mixed standard gamma source (ENV 984084) by Analytics Inc., Atlanta Georgia, USA. The reference soil sample is certified to have activity concentrations of 578.40 Bq/kg, 20.90 Bq/kg, and 10.47 Bq/kg for $^{40}$K, $^{238}$U, and $^{232}$Th, respectively. International Atomic Energy Agency (IAEA)-certified reference sediment sample (IAEA-Sediment-315) of mass 100 g obtained from the Analytical Quality Control Services (AQCS), IAEA Agency’s Laboratories, Seibendorf and Vienna, Austria was used to determine the activity concentrations of the sediment samples. The reference samples have similar matrices to the analyzed samples and were counted at the same geometry. The quality control parameters of the detector are reported elsewhere.[13]

The detection limit of any measuring system measures the operating capability of such system without the influence of the sample. The detection limit (DL), which is required to estimate the minimum detection level for appropriate determination of radionuclides in each sample were obtained using the expression[14,15] (Kitto et al., 2006, Jibiri and Emelue, 2008):

$$DL(Bq \, kg^{-1}) = \frac{1.96 \left[ B + SD_B \right]}{k \times \varepsilon \times m} \quad (1)$$

where $SD_B$ is the estimated standard error of the net background count rate in the spectrum of the radionuclide, $T$ is the sample counting time (s), $\varepsilon$ is the counting efficiency (cps/Bq), $m$ is the mass of the sample, $k$ is the factor that converts cps to Bq, and 1.96 represents a 95% confidence level. The detector characteristics and the minimum detectable activities evaluated in this study are presented in Table 1. Each sample was counted for 10 hours. This counting time was sufficient enough to produce strong peaks at gamma emitting energies used to evaluate the radionuclides considered in this study. Assuming secular equilibrium, respectively, in the uranium and thorium decay chains, $^{226}$Ra was determined by means of its progeny photo peak, $^{214}$Bi (1760 keV), and $^{232}$Th was analyzes by means of its progeny photo peak, $^{208}$Tl (2615 keV).[16] The 1460.75 keV gamma-ray transition line was used to measure the activity concentrations of $^{40}$K.[17] The activity concentrations in the samples were calculated according to the following expression:

$$A = \frac{C}{x \times \varepsilon \times M \times T} \quad (2)$$

where $C$ is the count rate of gamma rays, $\varepsilon$ is the detectors efficiency of the specific $\gamma$-ray, $P_x$ is the absolute transition probability of $\gamma$-decay, $M$ is the mass of the sample in kg, and $T$ is the counting time in seconds. The activity concentrations obtained for the measured radionuclides are expressed in Bq/kg per dry weight.

**Assessment of radiological hazard parameters**

The ultimate use of the measured activity concentrations in any material is to assess the radiological hazards incurred by man either outdoor or indoor. Outdoor exposure is considered for the population working on the mining site and adjoining farmlands as well as those living in the immediate environment, while the indoor exposure is for those living in cement houses made from marble rocks used in the Obajana cement factory. The radiation hazards of the radionuclides uniformly distributed in marble rock used as a major component of Dangote cement was assessed through various radiation hazard indices, which include radium equivalent activity, outdoor dose rates, indoor dose rates, and annual effective dose rates.

**Radium equivalent activity**

The radium equivalent activity is a widely used radiation hazard index to estimate the suitability of any material to be utilized as a component of building construction. It is calculated 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-ray dose rate.[10,11] The formula for calculating radium equivalent activity ($Ra_{eq}$) is given by Berekta and Matthew[18] as:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (3)$$

where $A_{Ra}$, $A_{Th}$, and $A_{K}$ are the activity concentrations of $^{226}$Ra (in equilibrium with $^{222}$U), $^{232}$Th, and $^{40}$K, respectively. Radium equivalent activity is directly related to the external and internal gamma dose due to radon and its progenies.[20] In evaluating the external and internal hazard indices, a critical comparison between radium equivalent activity ($Ra_{eq}$), external hazard index ($H_{ex}$), gamma index ($I_{\gamma}$), and internal hazard index ($H_{in}$) shows that all the indices are multiples of each other and convey the same information.[3] They are related in such a way that if $Ra_{eq} \leq 370$ Bq/kg, then $H_{ex} \leq 1$,

| Nuclides | Energy (keV) | $\gamma$-yield | Detection efficiency | Background count (cps) | Detection limits (Bq/kg) |
|----------|--------------|----------------|---------------------|------------------------|-------------------------|
| $^{40}$K | 1460.75      | 0.107          | 1.76 x 10$^{-2}$    | 1859 ± 7.6             | 17.3                    |
| $^{238}$U | 1760.0       | 0.159          | 1.45 x 10$^{-2}$    | 1135 ± 79.6            | 4.2                     |
| $^{232}$Th | 2615.0       | 0.358          | 0.95 x 10$^{-2}$    | 1148 ± 8.6             | 5.2                     |

Table 1: Detection characteristics and minimum detectable limits of the NaI (TI) detector used in this study

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\[ I_1 \leq 1, \quad H_2 \leq 1. \] Therefore, in this study, only radium equivalent activity was selected to represent the others.

**Outdoor dose rate**

The outdoor absorbed dose rates in air at a height of 1 m above the ground surface to which the workers in the mining area and farmlands were exposed are estimated based on the formula provided by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). \[ \text{BDL} \] The estimation assumed a uniform distribution of the natural occurring radionuclides in the rock and soil samples. The following formula was used for the calculations. \[ 21,22 \]

\[ D_{\text{out}} = 0.462A_{\text{Ra}} + 0.604A_{\text{Th}} + 0.0417A_{\text{K}} \left( \text{nGy h}^{-1} \right) \] (4)

where, \( A_{\text{Ra}}, A_{\text{Th}}, \) and \( A_{\text{K}} \) are the activity concentrations of \( ^{226}\text{Ra}, ^{232}\text{Th}, \) and \( ^{40}\text{K} \), respectively.

**Indoor dose rate**

The marble rock from the study area is used predominantly for the production of cement, which is distributed for use as a building material in every part of the country. Therefore, the indoor dose rate in a typical masonry building of standard room \( 4 \times 5 \times 2.8 \) m\(^3\) dimensions with wall thickness of 20 cm and density of the structure \( 2350 \) kg m\(^{-3}\) was calculated using data and formula provided by UNSCEAR\[^{[21]}\] and European Commission\[^{[22]}\] for building materials as applied by several researchers. \[^{[1,24‑26]}\]

The formula is given as:

\[ D_{\text{in}} = 0.92A_{\text{Ra}} + 1.1A_{\text{Th}} + 0.80A_{\text{K}} \left( \text{nGy h}^{-1} \right) \] (5)

where, \( A_{\text{Ra}}, A_{\text{Th}}, \) and \( A_{\text{K}} \) are respectively activity concentrations of \( ^{226}\text{Ra}, ^{232}\text{Th}, \) and \( ^{40}\text{K} \).

**Annual effective doses**

The severity of any radiological hazard is estimated based on the annual radiation dose received by a person working or living in the radiation environment. \[^{[1]}\]

To estimate the annual effective dose both outdoor and indoor, the following must be taken into account: (1) The conversion coefficient from absorbed dose in air to effective dose and (2) the outdoor and indoor occupancy factors.\[^{[9]}\] Using the dose rate data obtained from the concentrations of natural radionuclides in marble rock and soil samples, adopting the 0.7 Sv/Gy conversion factor from absorbed dose rates in air to effective dose received by adults and considering that people in Nigeria, on the average, spent \( \sim 20\% \) of their time outdoor, and \( \sim 80\% \) indoor, the outdoor and indoor effective doses were calculated as follows:

\[ \text{AED}_{\text{out}} (\text{SV}) = D_{\text{out}} \left( \text{nGy h}^{-1} \right) \times 24 \times 365 \times 24 \times 365 \times 0.7 \text{ Sv Gy}^{-1} \times 0.2 \] (6)

\[ \text{AED}_{\text{in}} (\text{SV}) = D_{\text{in}} \left( \text{nGy h}^{-1} \right) \times 24 \times 365 \times 0.7 \text{ Sv Gy}^{-1} \times 0.8 \] (7)

### Results and Discussion

#### Activity concentrations

The radioactivity levels of \(^{40}\text{K}, ^{226}\text{Ra}, \) and \(^{232}\text{Th}\) were measured in four different environmental matrices around Obajana cement factory to ascertain the radiological health hazards incurred by the population within the vicinity of the study area. The results of the measurements showing the range and geometric mean (GM) activity concentrations presented as GM (GSD) for \(^{40}\text{K}, ^{226}\text{Ra}, \) and \(^{232}\text{Th}\) are presented in Table 2. It can be seen from the results that the geometric mean GM (GSD) activity concentrations of \(^{40}\text{K}\) ranged from \( 89.6 (1.8) \) Bq/kg in marble rock to \( 1608.1 (1.4) \) Bq/kg in sediment. The geometric mean activity concentrations of \(^{226}\text{Ra}\) also ranged from \( 11.3 (1.8) \) Bq/kg in marble rock to \( 63.4 (1.2) \) Bq/kg in sediment and those of \(^{232}\text{Th}\) followed the same trend with minimum geometric mean concentration of \( 8.0 (1.7) \) Bq/kg in marble rock and maximum geometric mean concentration of \( 27.4 (1.7) \) Bq/kg in sediment. The radioactive levels of \(^{40}\text{K}\) are seen to be higher than those of \(^{226}\text{Ra}\) and \(^{232}\text{Th}\) at all the sampling points as well as in all the sampled matrices.

From the table, it is also clear that sediment has the highest concentrations of the three radionuclides among the sampled matrices with a range of \( 1000.6-2361.3 \) Bq/kg for \(^{40}\text{K}\), \( 41.8-88.2 \) Bq/kg for \(^{226}\text{Ra}\), as well as BLD–124.2 Bq/kg for \(^{232}\text{Th}\). The increasing order of concentrations for the three radionuclides in all the samples is as follows: Rock < farmland soil < soil < sediment. The mean concentrations of the three radionuclides are slightly lower in farmland soil than those in soil around the cement factory. This might be as a result of leaching, washing away or plant-uptakes of the radionuclides in the farmlands due to continuous tilling of the farmland soil. Also, the farmlands are owned by peasants who seldom apply fertilizer which could increase the radionuclides.

#### Table 2: Ranges and geometric mean GM (GSD) values of activity concentration of terrestrial radionuclides in marble rock, soil, farmland soil, and sediment samples

| Radionuclides | Marble (Bq/kg) | Soil (Bq/kg) | Farmland soil (Bq/kg) | Sediment (Bq/kg) |
|---------------|---------------|--------------|-----------------------|------------------|
| \(^{40}\text{K}\) | 30.7–560.9    | 135.0–630.2  | 129.2–211.7           | 1000.6–2361.3    |
| \(^{226}\text{Ra}\) | BDL–22.5     | 18.0–45.3    | 22.8–31.5             | 41.8–88.1       |
| \(^{232}\text{Th}\) | BDL–36.7     | BDL–103.9    | BDL–124.2             | 27.4 (3.5)      |

| \(^{40}\text{K}\) | GM (GSD) | GM (GSD) | GM (GSD) | GM (GSD) |
|---------------|----------|----------|----------|----------|
| \(^{226}\text{Ra}\) | 89.6 (1.8) | 257.6 (1.5) | 157.0 (1.2) | 1608.1 (1.4) |
| \(^{232}\text{Th}\) | 11.3 (1.8) | 28.1 (1.2) | 26.0 (1.1) | 63.4 (1.2) |

BDL: Below detection limit, GM: Geometric mean, GSD: Geometric standard deviation

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load in the soil. The high concentrations of radionuclides recorded in the sediment samples as compared to other samples might not be a product of the immediate natural environment but the accumulation of leached material into the water body as the river flows along different geological media or as a result of effluents from the cement factory. A comprehensive result of the activity concentration levels of $^{40}\text{K}$, $^{226}\text{Ra}$, and $^{232}\text{Th}$ in marble rock samples is shown in Table 3. The activity concentrations of the analyzed marble rocks are generally low when compared with other samples within the study area and a few order of magnitude lower than the worldwide average concentrations of $^{40}\text{K}$ (500 Bq/kg), $^{226}\text{Ra}$ (50 Bq/kg), and $^{232}\text{Th}$ (50 Bq/kg) for normal background radiation environment.[18] The low concentration of the radionuclides found in marble rock samples in this study is comparable to the situation for other marble/limestone rocks around the world [Table 4].

**Pearson correlation analysis**

The Pearson’s correlation matrix, showing the interactions among the three radionuclides in the various environmental samples, is presented in Table 5. As seen from the table, there exist strong significant positive interactions between $^{40}\text{K}_\text{rock}$ and $^{226}\text{Ra}_\text{rock}$ ($r^2 = 0.996$), $^{40}\text{K}_\text{rock}$ and $^{232}\text{Th}_\text{rock}$ ($r^2 = 0.981$), $^{226}\text{Ra}_\text{rock}$ and $^{232}\text{Th}_\text{rock}$ ($r^2 = 0.984$) at 0.01 significant level. Strong negative correlation is observed between $^{40}\text{K}_\text{rock}$ and $^{226}\text{Ra}_\text{sediment}$ ($r^2 = 0.973$), $^{226}\text{Ra}_\text{rock}$ and $^{226}\text{Ra}_\text{sediment}$ ($r^2 = 0.954$) and $^{232}\text{Th}_\text{rock}$ and $^{226}\text{Ra}_\text{sediment}$ ($r^2 = 0.955$) at 0.01 and 0.05 significant levels. Strong positive relationships also exist between $^{40}\text{K}_\text{sediment}$ and $^{226}\text{Ra}_\text{sediment}$ ($r^2 = 0.935$), $^{40}\text{K}_\text{sediment}$ and $^{232}\text{Th}_\text{sediment}$ ($r^2 = 0.907$) and $^{226}\text{Ra}_\text{sediment}$ and $^{232}\text{Th}_\text{sediment}$ ($r^2 = 0.959$) at 0.01 level of significance. For farmland soil samples, strong interaction only exists between $^{40}\text{K}_\text{farm}$ and $^{226}\text{Ra}_\text{farm}$ ($r^2 = 0.996$) at 0.01 significant level. Positive correlations found among the three radionuclides in rock and sediment samples suggest that these radionuclides originated from the same source for individual environmental matrix. The strong negative interactions between the radionuclides in rock and soil samples may imply similar behavior but different sources. Poor interactions were, however, observed between rock and sediment, rock, and farmland soil as well as sediment and farmland soil suggesting different sources and distributions of these radionuclides. These observations further confirm that the higher concentrations of the three radionuclides observed in sediment samples may be due to anthropogenic sources such as the waste generated in the cement factory.

**Outdoor and indoor exposures**

In order to assess the exposure to radiation in the outdoor and indoor environment within the study area, the radium equivalent activity, outdoor and indoor dose rates were calculated using equations 2–4. The radium equivalent activity evaluated in marble rock samples are presented in Table 3. For the rock samples, the value of $\text{Ra}_{eq}$ ranges from 10.9–115.1 Bq/kg with a geometric mean of 31.3 (1.5) Bq/kg. When compared with the maximum permissible level for

### Table 3: Activity concentrations, radium equivalent activity, and dose rate obtained in marble rock samples from Obajana

| S/N | Activity concentrations (Bq/kg) | Radium equivalent activity (Bq/kg) | Dose rate (nGy/h) |
|-----|--------------------------------|-----------------------------------|------------------|
|     | $^{40}\text{K}$              | $^{226}\text{Ra}$                | $^{232}\text{Th}$ | $\text{Ra}_{eq}$ | $D_{\text{outdoor}}$ | $D_{\text{indoor}}$ |
| 1   | 50.7±2.0                      | 14.1±0.8                         | BDL              | 18.0            | 8.6              | 17.0              |
| 2   | 85.5±2.5                      | 5.9±0.4                          | 22.7             | 30.8            | 4.5              | 27.6              |
| 3   | 130.7±3.1                     | 9.1±0.6                          | 6.5±0.5          | 15.7            | 7.3              | 13.6              |
| 4   | 76.8±2.3                      | 9.4±0.6                          | 25.8             | 11.8            | 22.3             |
| 5   | 560.9±6.4                     | 19.4±0.9                         | 115.0            | 54.5            | 103.0            |
| 6   | 117.8±2.9                     | 8.5±0.6                          | 36.8             | 16.9            | 32.0             |
| 7   | 107.0±2.8                     | 8.7±0.6                          | 16.9             | 8.5             | 16.6             |
| 8   | 67.0±2.2                      | 8.5±0.6                          | 18.8             | 8.9             | 17.1             |
| 9   | 111.8±2.9                     | 12.6±0.8                         | 27.8             | 13.3            | 25.6             |
| 10  | 100.2±2.7                     | 11.5±0.7                         | 29.8             | 14.0            | 26.7             |
| 11  | 102.9±2.8                     | 16.2±0.9                         | 53.6             | 24.5            | 46.7             |
| 12  | 37.9±1.9                      | 7.8±0.6                          | 36.1             | 16.5            | 31.9             |
| 13  | 92.7±2.6                      | 8.9±0.6                          | 33.3             | 15.5            | 29.6             |
| 14  | 140.3±3.2                     | 13.7±0.8                         | 28.7             | 13.9            | 27.0             |
| 15  | 30.7±3.1                      | 19.4±0.9                         | 52.4             | 23.3            | 44.3             |
| 16  | 82.4±2.5                      | 4.5±0.5                          | BDL              | 10.9            | 5.5              | 10.7              |
| 17  | 69.9±2.3                      | 6.6±0.5                          | 23.6             | 10.9            | 20.9             |
| 18  | 83.0±2.5                      | 6.2±0.5                          | 36.1             | 16.8            | 32.7             |
| 19  | 93.4±2.6                      | 15.0±0.8                         | 26.2             | 12.5            | 24.3             |
| GM (GSD) | 89.6 (1.8) | 11.3 (1.8) | 8.0 (1.7) | 31.3 (1.5) | 14.6 (1.5) | 28.0 (1.5) |

BDL: Below detection limit, GM: Geometric mean, GSD: Geometric standard deviation

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materials to be used for building construction, the mean value of radium equivalent activity obtained in this study is several orders of magnitude lower than 370 Bq/kg. This indicates that the Obajana marble rock is safe for use as a component of building materials. The mean radium equivalent activity obtained in this study for marble rock is also lower than values obtained in many locations around the world [Table 4]. The geometric mean outdoor dose obtained for rock, soil, and farmland soil samples are 14.6 nGyh⁻¹, 46.4 nGyh⁻¹, and 40.0 nGyh⁻¹, respectively. All these values are lower than the mean outdoor dose of 59 nGyh⁻¹ estimated for areas with normal background radiation levels around the world by UNSCEAR. These mean absorbed doses correspond to annual biological effective outdoor doses (using equation 4) of 0.018 mSv/y, 0.057 mSv/y and 0.049 mSv/y, respectively for marble rock, soil and farmland soil and these are all lower than 0.07 mSv estimated as world-wide average annual outdoor effective dose. The gamma dose incurred indoor by the populace due to the activity concentration levels in the marble rocks has been estimated using equation 4 and presented in Table 3. The indoor dose ranged from 10.7 nGyh⁻¹ to 103.1 nGyh⁻¹ with a geometric mean value of 28.0 nGyh⁻¹, which corresponds to 0.137 mSv mean indoor effective dose (equation 6) per year. These values are far lower than the average world-wide values of 84 nGyh⁻¹ and 0.41 mSv, respectively for indoor dose rate and annual effective indoor dose reported in UNSCEAR Reports. The indoor and outdoor doses incur from marble rocks are always generally low compared with other rock types; this is due to low concentrations of naturally occurring radionuclides in marble rocks around the globe.

### Conclusion

Measurements of activity concentrations of terrestrial radionuclides in marble rock, soil, farmland soil, and sediment samples collected around the Obajana cement factory have been carried out to ascertain the variations of activity levels of these radionuclides in different environmental samples and to assess the radiological health hazards associated with their use in building construction. The results show that sediment samples possess the highest concentrations of all the examined radionuclides in comparison to other environmental matrices. The activity concentrations in marble rock samples analyzed in this study are generally low. In particular, the activity concentration of Ra and Th in marble rock samples are relatively lower when compared with values obtained in marble rocks from other locations around the world and several orders of magnitude lower than the world average values for building materials. Strong positive relationships

### Table 4: Comparison of activity concentrations of ⁴⁰K, ²²⁳Ra, ²³²Th and radium equivalent activity (Raeq) in marble rock samples with values from other locations around the world

| Location       | ⁴⁰K (Bq/kg) | ²²³Ra (Bq/kg) | ²³²Th (Bq/kg) | Raeq (Bq/kg) |
|----------------|------------|---------------|---------------|--------------|
| China          | 113.20     | 24.20         | 20.20         |              |
| Egypt          | 19.30      | 20.40         | 4.40          | 48.78        |
| Italy          | 13.50      | 13.10         | 6.00          | 32.68        |
| Malaysia       | 243.3      | 19.0          | 16.5          | 61.3         |
| Jordan         | 93.3       | 19.3          | 11.9          | 43.4         |
| Pakistan       | 57.0       | 33.0          | 32.0          | 66.0         |
| Islamabad      | 13.80      | 14.32         | 2.05          | 34.41        |
| Margalla hills | 98.0       | 29.0          | 32.0          | 82.0         |
| Serbia         | 31.0       | 37.0          | 19.2          |              |
| Turkey         | 788.8      | 20.3          | 9.3           | 95.3         |
| Ankara         | 5.4        | 4.9           | 9.8           |              |
| Nigeria        | 89.6       | 11.3          | 8.0           | 31.3         |

### Table 5: Pearson’s correlation matrix showing the interactions among the radionuclides in different environmental matrices

|            | ⁴⁰Krock       | ²²³Rarock     | ²³²Throck     | ⁴⁰Ksed | ²²³Rased       | ²³²Thsed       | ⁴⁰Kfarm | ²²³Rafarm     | ²³²Thfarm     | ⁴⁰Kperm | ²²³Rperm     | ²³²Thperm     | ²³²K | ²³²Ra | ²³²Th       |
|------------|---------------|---------------|---------------|--------|---------------|---------------|---------|---------------|---------------|---------|---------------|---------------|------|------|-------------|
| ⁴⁰Krock    | 1             | 0.996**       | 0.981**       | 0.486  | -0.156        | -0.0753       | 0.616   | -0.073**      | 0.618          | -0.379  | -0.370**      | -0.372**       |      |      |             |
| ²²³Rarock  | 0.996**       | 1             | -0.973**      | -0.156  | -0.055        | -0.0721       | 0.616   | 0.592          | 0.523          | -0.035  | -0.347        | -0.506         |      |      |             |
| ²³²Throck  | 0.981**       | -0.973**      | 1             | -0.156  | -0.055        | -0.0721       | 0.616   | 0.592          | 0.523          | -0.035  | -0.347        | -0.506         |      |      |             |
| ⁴⁰Ksed     | 0.486         | -0.156        | -0.973**      | 1       | 0.450         | 0.605          | 0.616   | 0.592          | 0.523          | -0.035  | -0.347        | -0.506         | 0.486| 0.616| 0.486       |
| ²²³Rased   | -0.156        | -0.055        | -0.131        | 0.450   | 1             | 0.479          | 0.616   | 0.592          | 0.523          | -0.035  | -0.347        | -0.506         |      |      |             |
| ²³²Thsed   | -0.0753       | -0.0721       | -0.087        | 0.605   | 0.479         | 1             | 0.616   | 0.592          | 0.523          | -0.035  | -0.347        | -0.506         |      |      |             |
| ⁴⁰Kfarm    | 0.616         | 0.605         | 0.616         | 0.616   | 0.450         | 0.479          | 1       | 0.486          | 0.616          | -0.035  | -0.347        | -0.506         |      |      |             |
| ²²³Rafarm  | 0.618         | 0.592         | 0.592         | 0.618   | 0.450         | 0.479          | 0.616   | 1              | 0.486          | -0.035  | -0.347        | -0.506         |      |      |             |
| ²³²Thfarm  | 0.523         | 0.492         | 0.564         | 0.801   | 0.500         | 0.421          | 0.616   | 0.486          | 1              | 0.025   | 0.214         | 0.336          |      |      |             |

**Correlation is significant at 0.01 level (two-tailed), *Correlation is significant at 0.05 level (two-tailed)**
were observed among the three radionuclides for rock and sediment samples, respectively, suggesting a similar source for individual matrix. Poor inter-relationships were observed for the radionuclides between rock and sediment, soil, and sediment as well as sediment and farmland soil indicating different sources and behaviors. All the radiological parameters considered in this study have values which are much lower than their permissible limits. It can be concluded, therefore, that Obajana marble rock and other environmental matrices considered in this study do not pose any excessive radiological health hazards, indoors or outdoors and as such could be used without any restrictions.

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