ORIGINAL ARTICLE

# The $^{226}$Ra, $^{232}$Th and $^{40}$K contents in the Abakaliki baked shale construction materials and their potential radiological risk to public health, southeastern Nigeria

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Natural radioactivity associated with the Abakaliki baked shale as construction materials was measured using gamma spectrometry to establish radiological risk to public health. From the results, the mean activity concentrations of $^{226}$Ra, $^{232}$Th and $^{40}$K are 38.852 ± 2.829 Bqkg$^{-1}$, 69.589 ± 1.759 Bqkg$^{-1}$ and 557.667 ± 3.002 Bqkg$^{-1}$ respectively, higher than the world average concentrations of 35 Bqkg-1, 45 Bqkg$^{1}$ and 420 Bqkg$^{-1}$ respectively. The radiation hazard indices such as air absorbed dose ranged from 67.22 to 108.7 nGyh$^{-1}$ higher than the world average value of 55 nGyh$^{-1}$ [United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR]. The annual effective dose which ranged from 0.08 to 0.13 mSvy$^{-1}$ are less than the annual effective dose limit [E] of 1.0 mSvy$^{-1}$ for humans. The Radium equivalent activities [Raeq] [146.23 -234.59 Bqkg$^{-1}$] are below UNSCEAR recommended limit of 370 Bqkg$^{1}$. The internal [0.410-0.740] and external [0.393-0.640] radiation hazard indices are less than unity which is the world permissible value. The gamma activity indexes correspond to an activity concentration index of 2 ≤ $I_{\gamma r}$ ≤ 6 proposed by European Commission. The annual gonadal equivalent dose [AGED] average value of 537.53 mSvy$^{-1}$ is higher than the world average value of 300 mSvy$^{-1}$. On average, studied shales satisfy most of the health risk indices such as E and Raeq as well as the radiation hazard indices as their mean values are lower than the permissible limits. The use of these construction materials is free of any health risk related to radiation.

**Key Words:** *Albian, Asu river group, Construction material, Hazard indices, Health risk, Radiological effects*

Environmental problems associated with the extraction of naturally occurring radioactive materials [NORMs] in mines happened during drilling, leaching, handling, storage and transportation of mineral ores or aggregates (1,2). According to United Nations Scientific Committee on the Effects of Atomic Radiation (3) report, the NORM represents a potential internal radiation exposure hazard to both mine workers and members of the public through the inhalation and ingestion of radionuclides. Building raw materials and processed products can vary greatly in radionuclide content depending on the character and the geological origin. The natural radionuclides of concern in terrestrial environment are mainly uranium [$^{238}$U], thorium [$^{226}$Th] and potassium [$^{40}$K] and the radioactive gas radon. According to Environmental Protection Agency (4), radon emanates from the ground as a result of the direct decay of naturally-occurring radium. About 54% of the total external dose received by the public in normal background areas originates from $^{226}$Ra, $^{232}$Th and $^{40}$K (5). The NORMs in building materials can lead to occupants. In homes, the exposure rate depends on the concentrations of the radionuclides in the construction materials which the homes were constructed. Innocent et al. (6), agree that the spread of NORMs contaminate the environment, resulting in potential radiation exposure to humans.

According to UNSCEAR (7), inhalation of radon decay products in quarry sites can lead to high incidences of lung cancer in mine workers. Although there have been extensive studies on the radionuclide concentrations (1,2) in Nigerian mines, radionuclide concentrations have not been subjected to radiological regulatory control and so there is little or no awareness of the radiological hazards on the exposure to NORMs in mining areas such as in the Abakaliki area. Due to the health risks associated with the exposure to NORMs and inhalation of the short-lived decay products of radon, international bodies such as International Commission on Radiological Protection and Environmental Protection Agency (4) adopted strong measures at minimizing risk to such exposure. This is achieved by measuring the activity concentrations of $^{238}$U, $^{232}$Th and $^{40}$K by gamma spectroscopy. In the uranium series, the decay chain segment starting from radium [$^{226}$Ra] is radiologically the most important and, therefore, reference is often made to radium instead of uranium. Tzortzis et al. (8) noted that most rocks such as shale and phosphate rocks usually have relatively high content of this radionuclide. The Abakaliki area in the Benue Trough is underlain predominantly by shales of the Albian Asu River Group (9). The Abakaliki shales are being utilized in the region both as subgrade, aggregates in road and highway constructions and as building foundation materials. Other rocks exposed in the area include sandstone, siltstone, limestone, pyroclastics and diorite (9-11). Most of these rocks are crushed and used as aggregates in major construction projects (Figure 1). Demand for aggregate continues to rise because of new construction projects houses, bridges, railroads and highways. Information on the environmental and health impacts of radiation from the mining of rocks in Nigeria is very sparse. In Abakaliki area of Southeastern Nigeria, only the work on the radionuclide studies and the health risks of processing and constructing with the Abakaliki pyroclastics presently exist. No other research has been conducted on the various construction materials used in the study area and beyond. Based on this, the awareness of the radiological hazards and risks associated with long-term exposure to naturally occurring radioactive materials of most construction materials in the Abakaliki area are still sparse. The aim of this work is therefore, to evaluate the radiological health hazards associated with the use of the Abakaliki baked shales as utilized as construction aggregates by measuring the activity concentrations of $^{238}$U, $^{232}$Th and $^{40}$K in the rocks and determines the radiation hazard indices and the effective dose to the general public. The data generated will provide base line values of exposure to radiation in the area where mining activities are taking place and provide the authorities useful data for the implementation of radiation protection standards for the general population.

## DESCRIPTION OF THE AREA AND GEOLOGICAL SETTING

This study covers the Abakaliki metropolis (Figure 2) which is the capital of the Ebonyi state Nigeria. It is located on latitude 06°15' N and 06°25' N and longitude of 08°00' E and 08°10' E and covers about 420 km$^2$ The administrative status, the academic activities and fertile agricultural soil of the surrounding towns are the most significant reasons for the rapid economic growth in population and the spatial expansion of the metropolis. The climate of the study area is that of tropical rainforest with distinct wet and dry seasons. The dry season lasts from November to March and is usually characterized by periods of dry hot weather while the rainy season begins in April and ends in October. The major Shale Unit referred to as "Abakaliki baked shales" are Albian sediments which constitute part of the Asu-River Group (9,12) outcropping on the Abakaliki Anticlinorium. In southern Benue Trough, the Asu River Group sediments form the oldest Cretaceous

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**Figure 1)** *Photographs of the Abakaliki indurated shales, (a) quarry sites, (b) being processed*

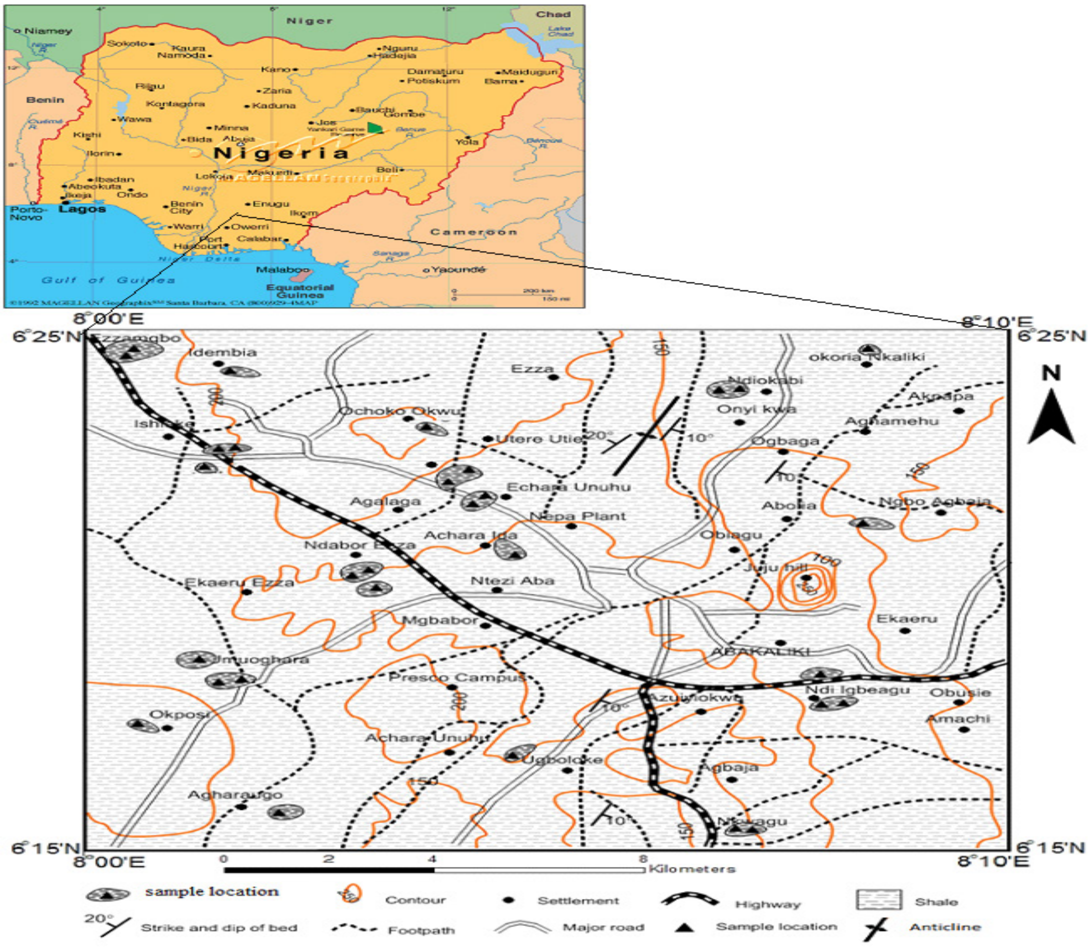


**Figure 2)** *Map of Abakaliki area showing sample locations superimposed by map of Nigeria*

marine sediment deposited during the Albian marine transgression in the Abakaliki area. The shales lie unconformably on the Precambrian basement complex. The Group comprises mainly of olive-brown or bluish grey coloured shales and sandy shales, fine-grained micaceous sandstones and micaceous mudstones with thin limestone bed s around Abakaliki (13). The Unit includes dark grey to black coloured pyritic micaceous shales with thin sandstone and siltstone beds, magnesitic and dolomitic horizons estimated thickness to be 2500 m thick (14). The shales of the Asu River Group consist dominantly of quartz veins, calcite, muscovite, pyrite and kaolinite as well as illite as clay minerals (15). According to Nweke et al. (16), the mineralogical analyses of the Abakaliki shale of Asu River Group of southeastern Nigeria reveals that the principal mineral components are clay minerals [kaolinite and illite], non-clay minerals [quartz and pyrite] with chlorite as secondary components. Previous research results (11,17,18) argue that the Lower Benue rift was filled by Pre-Santonian [Late Aptian to Coniacian] sedimentary rocks which were subjected to regional metamorphism. The induration of the shales in the Abakaliki area is attributed to the baking effect of igneous intrusions (19).

## METHODOLOGY

### Sampling and testing

A total of 15 Abakaliki baked shale samples were collected from different mining areas and taken to the laboratory at the Centre for Energy Research and Training, Ahmadu Bello University, Zaria where the samples were subjected to gamma spectrometry analysis. Each sample was dried and crushed to fine powder using a pulverizer. Both the sample preparation and analysis adopted followed the procedures as outlined in Girigisu et al. (20) the samples were packaged into radon-impermeable cylindrical plastic containers in the detector vessel which measures 7.6 cm by 7.6 cm. To prevent radon-222 escaping, the packaging in each case was triple sealed. The sealing process included smearing of the inner rim of each container lid with vaseline jelly, filling the lid assembly gap with candle wax to block the gaps between lid and container and tight-sealing lid-container with masking adhesive tape. Radon and its short-lived radionuclides were allowed to reach secular radioactive equilibrium by storing the samples for 30 days prior to gamma spectroscopy. The analysis was carried out using a 76 x 76 mm sodium iodide detector crystal optically coupled to a photomultiplier tube. The assembly had a preamplifier incorporated into it and a 1 kilovolt external source. The detector was enclosed in a 6 cm lead shield with cadmium and copper sheets. This arrangement was aimed at minimizing the effects of background and scattered radiation. The data acquisition software adopted was Maestro by Camberra Nuclear Products. The samples were measured for a period of 29000 seconds, for each sample. The peak area of each of the energy in the spectrum was used to compute the activity concentrations in each sample by the use of Equation 1.

$$C = C_n/C_{fk} \quad [1]$$

Where

- C = activity concentration of the radionuclides in the sample given in $BqKg^{-1}$
- $C_n$ = count rate [counts per second]; Count per second [cps] = Net Count/Live Time
- $C_{fk}$ = Calibration factor of the detecting system

Calibration of the system for energy and efficiency was done with two calibration point sources, Cs-137 and Co-60. These were done with the amplifier gain that gives 72% energy resolution for the $66.16K_{ev}$ of Cs-137 and counted for 30 minutes (Table 1).

The standards used to check for the calibration performed above are the International Atomic Energy Agency [IAEA] gamma Spectrometric reference materials with the IDs $RGK^{-1}$ for $^{40}K$, $RGU^{-1}$ for $^{226}Ra$ [Bi-214 peak] and RTG -1 for $^{232}Th$ [Ti-208].

**Calculation of radiological effects**

To assess the radiation hazards associated with the Abakaliki baked shale samples, 8 hazard indices were employed in this study (21-23).

Radium equivalent activity

The gamma-ray radiation hazards due to the specified radionuclides Ra, Th and K were assessed by radiation hazard index using the so called the radium equivalent activity, [Raeq] (21,22). The radium equivalent activity is given by Equation 2 as:

$$R_{aeq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad [2]$$

Representative level index

According to Alam et al. (24), another radiation hazard index called the representative level index, Iyr was calculated using Equation 3.

$$I_{yr} = \left(\frac{A_{Ra}}{200Bq/Kg} + \frac{A_{Th}}{100Bq/Kg} + \frac{A_K}{1500Bq/Kg}\right) \quad [3]$$

Where ARa, ATh and AK are the activity concentrations of 226Ra, 232Th and 40K, respectively, in Bq/kg. Manigandan et al. (25) stated that to satisfy the dose criteria, the value of the representative gamma index should be ≤1 which corresponds to an annual effective dose of ≤1 mSv (26).

Air absorbed dose rate

The total air absorbed dose rate, D [nanogray per hour, $nGyh^{-1}$] due to the activity concentrations of $^{238}Ra$, $^{232}Th$ and $^{40}K$ [Bq/kg] were mathematically calculated using the Equation 4 according to reference of Beck et al. (27) and UNSCEAR (5)

$$D = 0.427 A_{Ra} + A_{Th}\, 0.622 + A_K\, 0.0432 \quad [4]$$

Where $A_{Ra}$ is the activity concentration of $^{2268}Ra$, $A_{Th}$ is the activity concentration of $^{232}Th$ and $A_K$ is the activity concentration of $^{40}K$ in the samples.

Annual effective dose

The annual effective dose rate, E [millisievert per year, $mSvy^{-1}$] from outdoor gamma radiation can be estimated by taking into account the conversion coefficient from the absorbed dose in air to the effective dose [0.7 $SvGy^{-1}$] and an outdoor occupancy factor of 0.2 received by adults and an average value of 4.8 h spent in the mining area every day for a year. Under these assumptions, the annual effective dose equivalent can be calculated by using Equation 5 (7).

E = D [nGh-1] x 8760 [h] x 0.2 x 0.7 [SvGy-1] x 10-6 [5]

External hazard index

The external hazard index is used to limit the external gamma-radiation dose from building materials. The external hazard index [Hex] according to Berekta et al. (22) was calculated from Equation 6.

$$H_{ex} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \le 1 \quad [6]$$

Where $A_{Ra}$, $A_{Th}$ and $A_K$ are the activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$, respectively.

Internal hazard index

Radon and its short-lived products are also hazardous to the respiratory organs. So internal exposure to radon and its short-lived products is quantified by internal hazard index and is expressed by Beretka et al. (22) in Equation 7 as follows:

$$H_{in} = \left(\frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \le 1 \quad [7]$$

Where $H_{in}$ is the internal hazard index and $A_{Ra}$, $A_{Th}$ and $A_K$ are the activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$, respectively.

Annual gonadal equivalent dose

According to UNSCEAR (5), the gonads, the active bone marrow and the bone surface cells are considered organs of interest. However, the annual gonadal equivalent dose [AGED] for the residents in the study area due to the specific activities of $^{226}Ra$, $^{232}Th$ and $^{40}K$ was calculated using Equation 8 given by Arafa (28) as:

AGED = 3.09ARa + 4.18ATh + 0.314A [8]

Where $A_{Ra}$, $A_{Th}$ and $A_K$ are the activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$, respectively

Alpha index

Alpha-indexes [$I_\alpha$] have been developed (21) to assess the excess alpha radiation due to the radon inhalation originating from building materials. The alpha-indexes were determined using Equation 9 below:

$$I_a = \frac{A_{Ra}}{200Bq/Kg} \quad [9]$$

When the 226Ra activity concentration [$A_{Ra}$] of building material exceeds the value of 200 $Bqkg^{-1}$, the radon exhalation from this material could cause indoor radon concentration exceeding 200 $Bqm^{-3}$. The recommended exemption level and recommended upper level for the $226_{Ra}$ activity concentrations in building materials are 100 and 200 $Bqkg^{-1}$, respectively (29). This upper level is similar to and in agreement with the action level given by the ICRP (30) and by the European Commission (31).

## RESULTS AND DISCUSSION

The results of natural radionuclides concentrations measured on the Abakaliki baked shales are presented in Table 2. The results indicate that the activity concentration of $^{226}Ra$ ranged from 21.205 ± 1.159 to 57.034 ± 4.867 $Bqkg^{-1}$ with an average value of 38.852 ± 2.829 $Bqkg^{-1}$. The highest activity concentration of $^{226}Ra$ was detected in Sample SML5 with the value of 52.03 ± 4.86 $Bqkg^{-1}$. The activity concentration of $^{232}Th$ ranged from 53.820 ± 1.026 to 97.834 ± 2.281 $Bqkg^{-1}$ with an average value of 69.589 ± 1.759 $Bqkg^{-1}$. The highest activity concentration of $^{232}Th$ was detected in Sample SML4 [97.83 ± 2.28 $Bqkg^{-1}$].The activity concentration of $^{40}K$ ranged from 401.555 ± 1.711 to 722.706 ± 4.124 $Bqkg^{-1}$ with an average value of 557.667 ± 3.002 $Bqkg^{-1}$while the highest activity concentration of $^{40}K$ was detected also

**TABLE 1**

**Spectra energy windows, calibration factors and detection limits**

| Radionuclide | Gamma Energy (Kev) | Energy window (Kev) | Calibration Factors (cps/Bq/kg) | Detection limits (Bq/kg) |
|---|---|---|---|---|
| $^{226}$Ra | 1764.0 | 1620-1820 | 8.632 | 3.84 |
| $^{232}$Th | 2614.5 | 2480-2820 | 8.768 | 9.08 |
| $^{40}$K0 | 1460.0 | 1380-1550 | 0.032 | 14.54 |

in Sample SML4 with the value 722.71 ± 4.07 $Bqkg^{-1}$. In general, the activity concentrations indicate that $^{40}K > ^{232}Th > ^{226}Ra$. These degrees of association among the radionuclides may be because radium and thorium decay series come from the same origin and exist together in nature, whereas potassium is from a different origin (32). The errors as noted in the table include the statistical uncertainty in the peak area, calibration and counting errors. The UNSCEAR (5) recommended standard indicate that the world average activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$ are 35 $Bqkg^{-1}$, 45 $Bqkg^{-1}$ and 420 $Bqkg^{-1}$ respectively. The mean activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in the Abakaliki baked shales are 38.852 ± 2.829 $Bqkg^{-1}$, 69.589 ± 1.759 $Bqkg^{-1}$ and 557.667 ± 3.002 $Bqkg^{-1}$ respectively, is higher than the world average activity concentrations. This may be due to the high density dust generated from the mining processes and other related practices which may raise the possibility of exposure to NORMs and radon gas. Long exposure to any building materials with high uranium and thorium concentrations, according to Agency for Toxic Substance and Disease Registry (33), can cause several health effects such as chronic lung diseases, kidney cancer and necrosis of the mouth. However, areas or building materials dominated by with these concentrations should often be monitored. Workers or dwellers should also be examined especially when constantly exposed to such materials.

The activity concentrations were used to estimate several radiological parameters that served to qualify and quantify the radiological hazard associated with the studied construction materials. The absorbed dose rate due to primordial radionuclides ranged from 66.22 to 108.70 $nGyh^{-1}$ with an average value of 83.95 $nGyh^{-1}$ (Table 3). The world average total air absorbed dose rate according to UNSCEAR (5) is 55 $nGyh^{-1}$. The high concentration of absorbed dose rate could be attributed to the high uranium and thorium concentrations, according to Agency for Toxic Substance and Disease Registry (33). The annual effective dose ranged from 0.08 to 0.13 $mSvy^{-1}$ with an average value of 0.10 $mSvy^{-1}$. The annual effective doses is less than the annual dose limit 1.0 $mSvy^{-1}$ for the public as reported in the Nigeria Basic Ionizing Radiation Regulations (34). The AGED values ranged from 468.96 to 756.52 $mSvy^{-1}$ with an average value of 586.03 $mSvy^{-1}$. The AGED average value obtained is very high when compared with the annual gonadal equivalent dose world average value of 300 $mSvy^{-1}$ (7). The high concentrations of AGED in the studied baked shale are highly depended on the uranium and thorium concentrations, according to ATSDR (33). In most locations, the AGED values doubled when compared with the world average value which implies that the annual gonadal equivalent dose may pose threat to the bone marrow and the bone surface cells of the workers in the quarries. The radium equivalent activity values in the shales ranged from 146.23 to 234.59 $Bqkg^{-1}$ with an average value of 181.30 $Bqkg^{-1}$. The activity index provides a useful guideline in regulating the safety standard dwellings. UNSCEAR (5) and Girigisu et al. (20) recommended that Radium equivalent activity in building materials must be below 370 $Bqkg^{-1}$. Radium causes bone weakening, cranial and nasal tumors. Other diseases caused by radioactivity exposure according to Taskin et al. (35) include lung cancer, pancreas, hepatic, bone, skin, kidney cancers, cataracts, sterility, atrophy of the kidney and leukemia. However, according to Sam et al. (36), the use of a material whose concentration exceeds 370 $Bqkg^{-1}$ is discouraged to avoid radiation hazards. The calculated values of alpha-indexes ranged from 0.106 to 0.285 with average value of 0.194. The values of the alpha index in the baked shale samples are below the recommended limit, $I_\alpha<1$ (37), therefore, radon inhalation from the building material samples under investigation are not as large as to restrict their use in construction.

The values of representative level index which estimate the level of gamma radioactivity associated with different concentrations of certain specific radionuclides ranged from 1.058 to 1.722 with an average value of 1.327. This gamma index is also used to correlate the annual dose rate due to the excess external gamma radiation caused by superficial material. Thus, the activity concentration index should be used only as a screening tool for identifying materials that might be of concern when used in construction. There are some discussions on the dose criteria for gamma index levels for construction materials (25,26). According to European Commission (31) and Taskin et al. (35), values of representative level index $I_{\gamma r} \leq 0.5$ corresponds to an annual effective dose criterion of 0.3 $mSvy^{-1}$, whereas $2 \leq I_{\gamma r} \leq 6$ corresponds to an annual effective dose criterion of 1 mSvy-1. However, it is recommended that construction materials with $I_{\gamma r} > 6$ should totally be avoided for use as building material, since these values correspond to an annual effective dose higher than 1 mSv/y which is maximum permissible limit. The values of gamma activity index of the baked shale correspond to an activity concentration index of $2 \leq I_{\gamma r} \leq 6$ proposed by European Commission (31) for materials used in construction. On this basis, the use of the baked shale as construction materials may not likely lead to respiratory diseases such as asthma and cancer and external diseases such as erythema, skin cancer and cataracts (38-41).

The external radiation hazard index values ranged from 0.393 to 0.640 with an average value of 0.495 less than unity [1] which is the world maximum permissible value (7). According to Krieger (21) and Beretka et al. (22), for the radiation hazard to be negligible, the value of external radiation hazard index must be less than unity and this corresponds to the upper limit of Radium equivalent activity [370 $BqKg^{-1}$]. The internal radiation hazard index values ranged from 0.410 to 0.740 with an average value of 0.595. Internal exposures to radon are very hazardous and could cause respiratory diseases like asthma and cancer. According to Beretka et al. (22), internal radiation hazard index should also be less than unity for the radiation hazard to be negligible. However, the external radiation hazard index, internal radiation hazard index and radium equivalent activity for the baked shale satisfy the internationally acceptable limit and are free from the radiation hazards, suggesting that there may not be any inhalation of the short-lived decay products of radon when used as construction.

Comparison of the results of this work with published data from similar investigations in other rock types and the world averages are presented in Tables 4 and 5. The average value of radium equivalent activity for baked

**TABLE 2**

**The results of the activity concentrations of the indurated shale samples**

| Sample ID | Location | $^{226}Ra$ (Bq/Kg) | Error | $^{232}Th$ (Bq/Kg) | Error | $^{40}K$ (Bq/Kg) | Error | $^{226}Ra+^{232}Th+^{40}K$ (Bq/Kg) |
|---|---|---|---|---|---|---|---|---|
| **SML1** | Nkwagu | 49.247 | 2.897 | 81.870 | 2.281 | 664.075 | 0.001 | 795.192 |
| **SML2** | Agbaja | 34.762 | 4.287 | 56.328 | 1.482 | 401.555 | 0.001 | 795.192 |
| **SML3** | Unuhu | 36.269 | 4.867 | 68.415 | 1.026 | 520.684 | 0.003 | 625.368 |
| **SML4** | Iyiokwu | 39.050 | 2.550 | 97.834 | 2.166 | 722.706 | 0.003 | 859.590 |
| **SML5** | Ndiechi | 52.028 | 2.202 | 64.196 | 1.847 | 557.543 | 0.003 | 673.767 |
| **SML6** | Obusie | 21.205 | 1.159 | 64.196 | 1.824 | 582.582 | 0.002 | 667.983 |
| **SML7** | Ebonyi River | 35.110 | 1.506 | 57.127 | 1.938 | 488.647 | 0.002 | 580.884 |
| **SML8** | Ezeagu | 32.908 | 3.476 | 53.820 | 1.060 | 478.383 | 0.001 | 565.111 |
| **SML9** | Agbaja | 46.255 | 2.898 | 80.873 | 2.281 | 644.072 | 0.001 | 771.200 |
| **SML10** | Akpatakpa | 38.863 | 4.288 | 66.333 | 1.467 | 421.581 | 0.001 | 526.777 |
| **SML11** | Juju Hill | 38.675 | 4.867 | 61.423 | 1.226 | 575.682 | 0.003 | 675.780 |
| **SML12** | Obeagu Aba | 39.053 | 2.550 | 89.834 | 2.166 | 712.712 | 0.003 | 841.599 |
| **SML13** | Abaofia | 57.034 | 2.208 | 74.225 | 1.847 | 554.543 | 0.003 | 685.802 |
| **SML14** | Ogbaga | 24.214 | 1.176 | 68.225 | 1.843 | 543.583 | 0.002 | 636.022 |
| **SML15** | Aghamehu | 38.112 | 1.506 | 59.135 | 1.938 | 497.651 | 0.002 | 594.898 |
| ***Min*** | | 21.205 | 1.159 | 53.820 | 1.026 | 401.555 | 0.001 | 526.777 |
| ***Max*** | | 57.034 | 4.867 | 97.834 | 2.281 | 722.706 | 0.003 | 795.195 |
| ***Mean*** | | 38.852 | 2.829 | 69.589 | 1.759 | 557.667 | 0.001 | 666.108 |
| **UNSCEAR (2000)[a]** | | 35.0 | | 45.0 | | 420.0 | | 500.0 |

[a] Data from the United Nations Scientific Committee on the Effects of Atomic Radiations (7)

**TABLE 3**

**The radiological hazard indexes for the indurated shale samples**

| Sample ID | Location | $Ra_{eq}$ Bq/Kg | D $nGyh^{-1}$ | E $mSvy^{-1}$ | $H_{ex}$ | $H_{in}$ | AGED $mSvy^{-1}$ | Iα | Iγ | $I_{AU}$ | ELCR $\times 10^{-3}$ |
|---|---|---|---|---|---|---|---|---|---|---|---|
| **SML1** | Nkwagu | 217.45 | 100.6 | 0.12 | 0.587 | 0.410 | 702.92 | 0.246 | 1.592 | 1.49 | 0.396 |
| **SML2** | Agbaja | 146.23 | 67.22 | 0.08 | 0.393 | 0.490 | 468.96 | 0.174 | 1.058 | 1.03 | 0.264 |
| **SML3** | Unuhu | 174.20 | 80.54 | 0.09 | 0.470 | 0.570 | 561.55 | 0.181 | 1.267 | 1.20 | 0.297 |
| **SML4** | Iyiokwu | 234.59 | 108.7 | 0.13 | 0.640 | 0.740 | 756.52 | 0.195 | 1.722 | 1.60 | 0.429 |
| **SML5** | Ndiechi | 186.75 | 86.24 | 0.11 | 0.505 | 0.646 | 604.15 | 0.260 | 1.360 | 1.30 | 0.363 |
| **SML6** | Obusie | 157.86 | 74.16 | 0.10 | 0.424 | 0.481 | 516.78 | 0.106 | 1.153 | 1.03 | 0.330 |
| **SML7** | Ebonyi | 154.44 | 71.63 | 0.10 | 0.418 | 0.511 | 500.73 | 0.176 | 1.129 | 1.05 | 0.330 |
| **SML8** | Ezeagu | 146.71 | 68.21 | 0.08 | 0.396 | 0.485 | 476.87 | 0.165 | 1.076 | 1.02 | 0.264 |
| **SML9** | Agbaja | 165.73 | 97.88 | 0.12 | 0.569 | 0.694 | 683.41 | 0.231 | 1.546 | 1.46 | 0.396 |
| **SML10** | Akpatakpa | 175.98 | 76.07 | 0.09 | 0.454 | 0.554 | 529.74 | 0.194 | 1.200 | 1.19 | 0.297 |
| **SML11** | Juju Hill | 170.84 | 79.59 | 0.10 | 0.459 | 0.569 | 557.02 | 0.193 | 1.254 | 1.15 | 0.330 |
| **SML12** | Obeagu | 222.39 | 103.3 | 0.13 | 0.601 | 0.706 | 719.97 | 0.195 | 1.633 | 1.50 | 0.429 |
| **SML13** | Abaofia | 205.87 | 94.47 | 0.12 | 0.556 | 0.710 | 660.63 | 0.285 | 1.492 | 1.47 | 0.396 |
| **SML14** | Ogbaga | 163.63 | 76.26 | 0.10 | 0.441 | 0.507 | 530.69 | 0.121 | 1.205 | 1.08 | 0.330 |
| **SML15** | Aghamehu | 160.99 | 74.55 | 0.09 | 0.433 | 0.543 | 521.20 | 0.191 | 1.176 | 1.10 | 0.297 |
| ***Min*** | | 146.23 | 67.22 | 0.08 | 0.393 | 0.410 | 468.96 | 0.106 | 1.058 | 1.03 | 0.264 |
| ***Max*** | | 234.59 | 108.7 | 0.13 | 0.640 | 0.740 | 756.52 | 0.285 | 1.722 | 1.50 | 0.429 |
| ***Mean*** | | 181.30 | 83.95 | 0.10 | 0.495 | 0.595 | 586.03 | 0.194 | 1.327 | 1.24 | 0.343 |
| **UNSCEAR*** | | 370.0 | 55.0 | ≤1.0[a] | ≤1.0 | ≤1.0 | 300.0 | <1.0 | ≤6[b] | - | 0.290[b] |

**Note- Data from the United Nations Scientific Committee on the Effects of Atomic Radiations (40); [a]data from NBIRR (34); [b]data from Taskin et al. (35)**

**TABLE 4**

**Radiation hazard parameters of some rock types and the indurated shale samples**

| Type of rock | **Number of samples** | **D ($nGyh^{-1}$)** | **Representative level index** | **Radium equilvalent $BqKg^{-1}$** |
|---|---|---|---|---|
| Esana Shale[a] | 5 | 58.00 (36.9-103.8) | 0.89 (0.57-1.60) | 131.0 (82.9-235.8) |
| Tarawan chalk[a] | 5 | 103.00 (51.6-168.5) | 1.59 (0.79-2.59) | 210.0 (109-339.6) |
| Dakha shale[a] | 9 | 78.00 (27.6-94.0) | 1.20 (0.43-1.46) | 172.0 (63.7-211.2) |
| Quseir Shale[a] | 10 | 125.00 (60.0-197.0) | 1.90 (0.93-3.00) | 275.0 (134.0-438.0) |
| Nubai Sandstone[a] | 10 | 142.00 (104-252.0) | 2.19 (1.6-3.90) | 304.0 (224.0-550.0) |
| Abakaliki indurated shale[b] | 15 | 83.95 (66.2-108.7) | 1.32 (1.05-1.72) | 181.3 (146.2-234.6) |

**Note- The results in parentheses correspond to the minimum and maximum values of the parameters; [a] Adapted from the work of Abbady (41); [b] This study**

**TABLE 5**

**Comparison of mean values of some radiological indices of the present study with those of other parts of the world**

| Location | Air absorbed dose ($nGyh^{-1}$) | Annual effective dose ($mSvy^{-1}$) | Radium equivalent $BqKg^{-1}$ |
|---|---|---|---|
| **Western Ghats, India** | 91.54 | - | 208 |
| **Northern Pakistan** | 87.47 | 0.11 | 190 |
| **Saudi Arabia** | 35.2 | 0.04 | 68.1 |
| **Tehran city, Iran** | 69.1 | 0.08 | 143.6 |
| **Eastern Sichuan, China** | 60 | 0.07 | 130 |
| **Niger Delta, Nigeria** | 30 | 0.04 | 76 |
| **West Bank, Palestine** | 88.2 | 0.11 | 185.8 |
| **Kuantan, Malaysia** | 11.16 | 0.01 | 24.92 |
| **This study[a]** | 83.95 | 0.10 | 181.3 |

**Note- [a]Data from this study**

shale and the other rock types were found to be below the internationally accepted value of less than 370 $Bqkg^{-1}$ for materials that will be used in building of dwellings. The average value of representative level index for all the rock types are however higher than the internationally accepted value of 1 $Bqkg^{-1}$ (5) except for the Esana Shale with Iyr average value of 0.89 which corresponds to an annual effective dose criterion of 1 $mSvy^{-1}$. The average values of gamma absorbed dose rates of the baked shale [83.95 $nGyh^{-1}$] and the other rock types as shown in Table 4. 23 are higher than the estimate of average global terrestrial radiation of 55 $nGyh^{-1}$ (5). In terms of air absorbed dose rate the Abakaliki baked shale with air absorbed dose rate of 83.95 $nGyh^{-1}$ with other shales such as Esana shale [58.0 $nGyh^{-1}$] and Dakha shale [78.0 $nGyh^{-1}$] referred to in this study have values higher than internationally accepted limit of 55 $nGyh^{-1}$ (Table 4).

## CONCLUSION

The mean activity concentrations of $^{226}Ra$, $^{232}Th$ and $^{40}K$ in the Abakaliki baked shales are 37.57 ± 2.86 $Bqkg^{-1}$, 67.97 ± 1.70 $Bqkg^{-1}$ and 552.02 ± 2.92 $Bqkg^{-1}$ respectively, higher than the world average activity concentrations. The activity concentrations which indicate that $^{40}K > ^{232}Th > ^{226}Ra$ were used to estimate several radiological parameters that qualify and quantify the radiological hazard associated with the studied construction materials the low alpha index also satisfied the internationally acceptable limit. The annual gonadal equivalent dose values are doubled when compared with the world average permissible limit of ≤ 300 $mSvy^{-1}$ indicating unsafe situation, as relates to radiation. The radiation hazard indices such as annual effective dose rate, the external radiation hazard index, internal radiation hazard index and radium equivalent activity for Abakaliki baked shale meet international standards on radiological protection, as only D and AGED failed the standards. Therefore, the studied materials are free from radiation hazards which suggest probable absence of inhalation of the short-lived decay products of radon from the baked shales used as construction material in the region. The results of this study have provided strong basis for the assessment of exposure of humans in the region to radiological health hazards and implementation of radiation protection standards by the concerned authorities. However, the acceptability of Abakaliki baked shale as construction materials or any other materials should henceforth not only be based on the determination of the level of natural radioactivity but on the radiological indices on the building materials and other possible pathways which usually confirm whether the materials are free of any health risk related to radiation.

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## REFERENCES

1. Abdulkarim MS, Umar S. An investigation of natural radioactivity around gold mining sites in Birnin Gwari North Western Nigeria. Res J Physical Sci 2013;1:20-23.

2. Ademola JA, Ademonehin S. Radioactivity concentrations and dose assessment for bitumen and soil samples around bituminous deposit in Ondo state, Nigeria. Radiopro 2010;45:359-368.

3. UNSCEAR. Source and Effects of Ionizing Radiation. Report to General Assembly, with Scientific Annexes. United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations, New York. 2010:19-220.

4. Environmental Protection Agency [EPA]. United States. Ionizing radiation Fact book. EPA. Office of Radiation and Indoor Air. 2007;EPA-402-F-06-061.

5. United Nation Scientific Committee on the Effects of Atomic Radiation [UNSCEAR]. Sources, Effects and Risks of Ionizing Radiation. United Nations, New York, Annex A, B.1988.

6. Innocent AJ, Onimisi MY, Jonah SA. Evaluation of naturally occurring radionuclide materials in soilsamples collected from some mining sites in Zamfara State, Nigeria. Br J Appl Sci Technol 2013;3:684-92.

7. UNSCEAR. Sources and Effects of Ionizing Radiation. United Nations, New York, Annex B. 2000;1:83-157.

8. Tzortzis M, Svoukis E, Tsertos HA. Comprehensive study of natural gamma radioactivity levels and associated dose rates from surface soils in Cyprus. Radiat Prot Dosim J 2004;109:217-24.

9. Reyment RA. Aspects of the Geology of Nigeria. University Press: Ibadan, Nigeria. 1965:154.

10. Ofoegbu CO, Amajor LC. A geochemical comparison of the pyroclastic rocks from Abakaliki and Ezillo, southeastern Benue Trough, Nigeria. J Min Geol 1987;23:45-52.

11. Obiora SC, Umeji AC. Petrographic evidence for regional burial metamorphism of the sedimentary rocks in the lower Benue Rift. J Afr Earth Sci. 2004;38:269-77.

12. Ojoh KA. The Southern part of Benue Trough, Nigeria Cretaceous stratigraphy, Basin analysis, paleo-oceanography and the aerodynamic evolution of the Equatorial domain of the South Atlantic NAPE Bulletin 1992;7:67–74.

13. Simpson A. The Nigerian coalfield. The geology of parts of Onitsha, Owerri and Benue provinces. Bulletin Geological Survey Nigeria 1954;24:85.

14. Agumanu AE. The Abakaliki and Ebonyi Formations: subdivisions of the Albian Asu River Group in the southern Benue Trough, Nigeria. J Afr Earth Sci 1989;9:195-207.

15. Ehinola OA, Abimbola AF. Preliminary assessment of major and trace elements content in the middle Cretaceous black shales of the Abakaliki fold belt, Southeastern Nigeria Nafta 2002;53:323–6.

16. Nweke OM, Okogbue CO. The potential of cement stabilized shale quarry dust for possible use as road foundation material. Int J Geo-Eng 2017;8:1-14.

17. Benkhelil J. Cretaceous deformation, magmatism and metamorphism in the lower Benue Trough, Nigeria. Geological Journal 1987;22:467-93.

18. Obiora SC, Charan SN. Geochemistry of Regionally metamorphosed sedimentary rocks from the Lower Benue Rift: Implications for provenance and tectonic setting of the Benue Rift sedimentary suite. S Afr J Geol 2011;1:25-40.

19. Cratchley C, Jones GP. An interpretation of the geology and gravity anormalies of the Benue Valley, Nigeria. Overseas geol. Surv Geophys Paper 1, Geological Magazine, 1965;118:59-67.

20. Girigisu S, Ibeanu IGE, Adeyemo DJ, et al. Assessment of Radiological Levels in Soils from Artisanal Gold Mining Exercises at Awwal, Kebbi State, Nigeria. Res J Appl Sci Eng Technol 2014;7:2899-904.

21. Krieger R. Radioactivity of construction materials. Betonwerk Fertigteil Techn. 1981;47:468.

22. Beretka I, Mathew PI. Natural radioactivity of Australian building materials, waste and by-products. Health Phys. 1985;48:87–95.

23. Zarie KA, Mugren KSA. Measurement of natural radioactivity and assessment of radiation hazard in soil samples from Tayma area [KSA]. Isotope and Rad Res 2010;42:1- 9.

24. Alam MN, Chowdhury MI, Kamal M, et al. The 226Ra, 232Th and 40K activates in beach sand minerals and beach soils of Cox's Bazar, Bangladesh. J Environ Radioactivity 1999;46:243–50.

25. Manigandan P, Chandar Shekar B. Evaluation of radionuclidesin the terrestrial environment of Western Ghats. J Radiat Res Appl Sci 2014;7:310- 6.

26. Ravisankar R, Vanasundari K, Suganya M, et al. Multivariate statistical analysis of radiological data of building materials used in Tiruvannamalai, Tamilnadu, India. Appl Radiat Isot 2014;85:114–27.

27. Beck HL, Decompo J, Gologak J. In-situ Ge[Li] and NaI[Tl] gamma ray spectrometry. Health and Safety Laboratory AEC, Report HASL 258, New York. 1972.

28. Arafa W. Specific activity and hazards of granite samples collected from the eastern desert of Egypt. J Environ Radioact 2004;75:315-22.

29. Naturally occurring radiation in Nordic countries recommendation", In: Akerblom G, Mjones L, Annanmaki M, et al. [Ed.]. The Flag-Book Series, The Radiation Protection Authorities in Denmark, Finland, Norway and Sweden, Reykjavik. 2000.

30. International Commission on Radiological Protection, ICRP. Radiation Protection: 1990 Recommendations of the International Commission on Radiological Protection. Pergamon Press, Oxford. 1990.

31. European Commission. Radiation Protection 112-radiological protection principles concerning the natural radioactivity of building materials, Directorate-General Environment. Nuclear safety and civil Protection. 1999.

32. Tanaskovic I, Golobocanin D, Miljevic N. Multivariate statistical analysis of hydrochemical and radiological data of Serbian spa waters. J Geochem Explor 2012;112:226–234.

33. ATSDR [Agency for Toxic Substances and Disease Registry]. Case studies in environmental medicine. Radon toxicity. Public Health Service, U.S. Depart-ment of Health and Human Services, Atlanta. 1992.

34. Nigeria Basic Ionizing Radiation Regulations [NBIRR]. Established by Nigerian Nuclear Regulatory Authority [NNRA] 2003:85.

35. Taskin H, Karavus M, Topuzoglu PA, et al. Radionuclide concentrations in soil and lifetime cancer risk due to the gamma radioactivity in Kirklareli, Turkey. J Environ Radioact 2009;100:49-53.

36. Sam AK, Abbas N. Assessment of radioactivity and associated hazards in local and imported cement types used in Sudan. Radiat. Prot. Dosim 2010;88:225-260.

37. Raghu Y, Harikrishnan N, Chandrasekaran A, et al. Assessment of natural radioactivity and associated radiation hazards in some building

materials used in Kilpenathur, Tiruvannamalai Dist, Tamilnadu, India. African J Basic & Appl Sci 2015;7:16-25.

38. Avwiri GO, Osimobi JC, Agbalagba EO. Evaluation of radiation hazard indices and excess lifetime cancer risk due to natural radioactivity in soil profile of Udi and Ezeagu local government areas of Enugu State, Nigeria. Comprehensive J Environ. Earth Sci 2012;1:1-10.

39. Ademola AK, Bello AK, Adejumobi AC. Determination of natural radioactivity and hazard in soil samples in and around gold mining area in Itagunmodi, south-western, Nigeria. J Radiat Res Appl Sci 2014;7:249-55.

40. United Nation Scientific Committee on the Effects of Atomic Radiation. (UNSCEAR). Radiation exposure from natural sources of radiation. New York: United Nation. 1993.

41. Abbady AGE. Estimation of radiation hazard indices from sedimentary rocks in Upper Egypt, Technical note. Appl Radiat Isot 2004;60):111–4.