Evaluation of Radioactivity Concentration of some selected Mineral Rocks From Mayo-Belwa Local Government Area of Adamawa State, Nigeria

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Abstract
Radiation from natural sources is constantly present around people and their surroundings. Natural Occurring Radioactive Materials (NORM) present in rock, soil and underground water are the major sources of this radiation. In this study, radioactivity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K from Ten (10) different Granite (GN), Gneiss (GS), and Migmatite (MG) rocks samples obtained from Mayo Belwa Local Government Area of Adamawa State were evaluated using a well calibrated and shielded Canberra 3 x 3 inch NaI(Tl) detector at the National Institute of Radiation Protection and Research (NIRPR), University of Ibadan. Rock samples were cleaned, pulverised and placed in the detector for counting, and based on standard expressions, the radionuclide content of the granite rock samples were evaluated. The result shows that the activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K in GN samples varies from 62.44 – 117.67 Bq/kg, 76.59 – 165.58 Bq/kg, and 688.03 – 1472.42 Bq/kg with corresponding mean of 74.59 ± 3.12, 104.41 ± 3.12, and 950.16 ± 3.12 Bq/kg. Activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K in GS samples ranges from 19.23 – 36.49 Bq/kg, 29.06 – 49.42 Bq/kg, and 310 – 924.21 Bq/kg with corresponding mean of 28.1 ± 5.36 Bq/kg, 38.92 ± 6.38 Bq/kg, and 664.21 ± 178.14 Bq/kg. Activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K in MG samples ranges from 32.11 – 74.73 Bq/kg, 40.79 – 105.87 Bq/kg, and 453.34 – 1040.77 Bq/kg with corresponding mean of 50.19 ± 14.35 Bq/kg, 60.50 ± 19.96 Bq/kg, and 714.88 ± 200.37 Bq/kg. The mean activity from this study are higher than the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) global mean of $^{238}$U (32 Bq/kg), $^{232}$Th (45 Bq/kg), and $^{40}$K (420 Bq/kg) in soil and rock samples except for $^{238}$U and $^{232}$Th in GS samples which are lower than the recommended standards. The results signifies that usage of such rocks as building construction raw materials might pose radiological hazards in the long run. Therefore, mineral content of the rock responsible for the high radionuclide concentration should be investigated.

Keywords: Natural Occurring Radioactive Materials, Radioactivity Concentration, Mineral Rocks, Granite, Gneiss, Migmatite.
INTRODUCTION
Man and his environment is consistently exposed to radiation from natural sources (UNSCER, 2000). Natural occurring radioactive materials (NORM) are the major sources of this radiation. NORM are present in rock, soil and underground water. Materials derived from these geological matrices retain these NORM in varying proportions. Radionuclide assay in rocks as environmental material or building materials is important for delineating radiologically safe environment and building materials. Crustal materials contain radioactive isotopes which consist mainly of NORM (\(^{238}\text{U},^{232}\text{Th}\) and \(^{40}\text{K}\)). The wide distribution of these isotopes in rocks and soils contributes majorly to the background radiation dose received by man. High level of radionuclide and the radiation that emanate from them could cause health complications and system malfunction (Ogundare and Olarinoye, 2016, Akpan et al., 2020). Therefore, the level of NORM in environmental samples is important from public health perspective.

Granite, Banded Gneiss, Migmatite are some of the rock species that are found in abundance in Nigeria and mined for the construction of roads, offices, home structures, commercial centres and also as ornament stones. The harvesting of these types of rocks and their subsequent use as building construction raw materials could perturb the background level of radiation from natural radionuclides in the environment. Granite, Banded Gneiss, Migmatite are found in large quantity in the Adamawa State of Nigeria. Consequently, licenced and unlicensed mining companies are active in the area to mine these rock and process them for different mineral extraction and construction purposes.

The radiological impact of the mining of these rocks in large quantity at Mayo-Belwa area of Adamawa State has so far not been investigated. Furthermore, the use of the said rock samples as building / construction materials could elevate indoor radiation dose to man if the radioactivity level of the rocks is above recommended safety limits. Consequently, the radiotoxicity burden associated with these rocks as building materials should be a cause for concern and interest to builders, policy makers and the public. Another common source of radiation exposure source is building material. A great proportion of human population spent more than 50% of their activities indoor. The fact that many common building materials are derived from geological matrices such as rocks, stones, soil, and plants suggest that indoor radiation exposure is largely dependent on the radionuclide content of the geological units they are derived from. Apart from being used as basic building material, some rocks such as banded gneiss, migmatite, and granite due to their attractive nature, strength and abundance are used as flooring material, grave stones, pavement stones, working surface, road construction, landscaping stones and ornamental stones. The presence of such structures in homes, parks, and offices could elevate the radiation burden of human population. It is thus important that such materials be screened for radionuclide content before they are used for such purposes.

Screening level for radioactivity in building materials have been recommended by different regulatory bodies such as the European Union, International Atomic Energy Agency (IAEA), United State Environmental Protection Agency (USEPA), and the National Council for Radiation Protection (NCRP). These bodies have set limits for radionuclide content of different categories of building materials (UNSCEAR, 2000: Sas et al., 2016; Singovszka et al., 2018) with which materials can be screened before use. It is uncommon in Nigeria to check building materials for radioactive content. As a result, various domestic and foreign materials are utilized in building without consideration for the added radiotoxicity they pose to the environment. This may result in radiation-related health risks. Because radiation-induced
health problems are not uncommon, early diagnosis is challenging and frequently poorly managed. Therefore, checking the radionuclide content of commonly used building materials is a crucial part of quality control in the construction sector. Furthermore, there is a scarcity of published information on the radioactive concentrations of granite, banded gneiss, and migmatite from Nigeria, hence the need for this study. A study of primordial radionuclide concentration in some granite rocks collected from quarry sites in Abuja Nigeria using NaI (Tl) detector shows that the mean activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K are all higher than the recommended United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) global mean of $^{238}$U (32 Bq/kg), $^{232}$Th (45 Bq/kg), and $^{40}$K (420 Bq/kg) in soil and rock samples (Shittu et al. 2015).

MATERIALS AND METHODS

Description of Study Area.

Adamawa State is located in the Northeastern Zone of Nigeria, bounded by latitudes 7° and 11° N and longitudes 11° and 14° E and occupies a land area of about 36,917 km$^2$. Mayo Belwa is a Local Government Area in Adamawa State with a total land area of 1768 km$^2$. It lies between latitude 9° 45' – 9° 43' N and Longitude 13°05' – 14° 12' E of the Greenwich meridian (Adebayo and Tukur, 1999). Figure 1.1 show the map of Nigeria, Adamawa State, Mayo Belwa and environs while figure 2 shows the geological map of study area showing the concentrations of of granite, banded gneiss, and migmatite rocks respectively.

Figure 1: Map of Nigeria indicating the study area.
Materials Used for sample Collection
For the purpose of this study, the following materials were used during sample collection:- hammer, disposable hand gloves, nose mask, plastic containers, sieve, masking tape and marker.

Sample Collection.
Three rock species commonly found and mined in the study area for various purposes were collected from quarry sites and mine pits across Mayo Belwa area using systematic random sampling techniques. Ten samples each of banded Gneiss, Granite, and Migmatite were collected for gamma spectrometry analysis. The samples were collected for the period of seven days. A hammer was used to break the rocks into smaller sizes. The rocks were marked with a masking tape with a sample ID using a white board marker. The marked rock samples were transported to the laboratory for preparation and further analysis. Figure 3(a-c) shows the sample plate collected from the study area for each category of rock samples.
Sample Preparation and Analysis
At the laboratory, the rocks were cleaned and washed with distilled water to remove attached debris which could be sources of contaminations. The washed rocks were then sun-dried for six hours to remove water molecules remaining on the rock surface after washing. The dried rocks were crushed, ground and sieved through a 500 µm mesh size sieve to homogenise the pulverised samples. 500 g each of the homogenised samples were weighed using a digital weighing balance and poured into a well-marked plastic container for proper identification. The container was made airtight by applying candle wax and tape on the lid to prevent cross contamination and gaseous exchange between the sample and the environment. The well-sealed samples in the containers were kept for 32 days to attain secular radioactive equilibrium between $^{238}$U, $^{232}$Th, $^{226}$Ra and its short live progenies ($^{222}$Rn, $^{214}$Pb, $^{226}$Rn).

A Canberra 3 X 3 inch NaI(Tl) detector at the National Institute of Radiation Protection and Research (NIRPR), University of Ibadan, Ibadan was used for γ-rays spectrometry samples to determined the activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K from Ten (10) different Granite, Gneiss, and Migmatite rocks samples. The detector is housed in a 5 cm thick cylindrical lead shield with internal diameter of 24 cm and height of 60 cm. The lead shield is also lined with layers of cadmium and Plexiglas of 3 mm thick each as part of the set-up shield to lockout external background radiation that may come from other sources.

RESULTS AND DISCUSSION

Data Presentation and Analysis
Figure 4 shows the analysis of the mean activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K from Granite, Gneiss, and Migmatite rocks samples obtained from the study area.
From figure 4, the result shows that the activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K in Granite samples varies from 62.44 – 117.67 Bq/kg, 76.59 – 165.58 Bq/kg, and 688.03 – 1472.42 Bq/kg with corresponding mean of 74.59 ± 3.12, 104.41 ± 3.12, and 950.16 ± 3.12 Bq/kg. Activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K in Gneiss samples ranges from 19.23 – 36.49 Bq/kg, 29.06 – 49.42 Bq/kg, and 310 – 924.21 Bq/kg with corresponding mean of 28.1 ± 5.36 Bq/kg, 38.92 ± 6.38 Bq/kg, and 664.21 ±178.14 Bq/kg. Activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K in Migmantite samples ranges from 32.11 – 74.73 Bq/kg, 40.79 – 105.87 Bq/kg, and 453.34 – 1040.77 Bq/kg with corresponding mean of 50.19 ± 14.35 Bq/kg, 60.50 ± 19.96 Bq/kg, and 714.88 ± 200.37 Bq/kg. The mean activity of the radionuclides are higher than the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) global mean of $^{238}$U (32 Bq/kg), $^{232}$Th (45 Bq/kg), and $^{40}$K (420 Bq/kg) in soil and rock samples except for $^{238}$U and $^{232}$Th in Gneiss samples which are lower than the recommended standards as shown in figure 5, while figure 6 shows the activity concentration ratio of $^{232}$Th and $^{238}$U from the rock samples.
Discussion
The activity concentration vary according to the geological specification of the rocks. Due to the large ionic radius of U and Th, they often exist in late crystalline magmas and the resulting solution which prevents them from being available in crystalline silicate (Maxwell et al., 2015). Consequently, both elemental species are essentially found in pegmatites and granites. More so, based on the formation mechanisms, radionuclides are found in higher quantities in igneous rocks containing intrusive content (Maxwell et al., 2015; Langmuir, 1978).

The result also shows that the mean activity concentration of \(^{238}\text{U}\), \(^{232}\text{Th}\), and \(^{40}\text{K}\) in granite is higher compare to those of the other two rock samples. The higher specific activity could be attributed to the uranium and thorium rich minerals in the granite rock samples and followed by metamorphic rocks (migmatite). It should also be understood that migmatite represents the transition between metamorphic to igneous rock cycle, consequently the higher activity concentration compared to gneiss. The trend of the activity concentration in the rocks, \((\text{A})_{\text{granite}} < (\text{A})_{\text{migmatite}} < (\text{A})_{\text{banded gneiss}}\) is consistent with the conclusion of Adabanija et al. (2020) when they studied the activity and radiogenic heat associated with rocks samples collected from Okene area of Nigeria.

The uranium mobilisation relative to thorium can be quantified using the ratio of \(^{232}\text{Th} / {^{238}\text{U}}\) in most rocks varies within 1 - 3.5 in the absence of uranium enrichment / depletion (Plant et al., 2003). The range and mean value of the ratio in the three species of rocks is presented in Figure 6. The mean value of \(^{232}\text{Th} / {^{238}\text{U}}\) is 1.41, 1.39, and 1.21 in granite, banded gneiss, and migmatite respectively. These values suggest uranium depletion in the rocks which is relatively least in mimatite and granite the most. The mean value for granite and banded gneiss is found to be lower compared to corresponding rock species value of 2.65 and 2.74 for granite and banded gneiss according to the findings by Adabanija et al. (2020) in Okene area Nigeria.
The activity concentration obtained from this study was compared with previous literature and the comparison result is presented in Table 1.

Table 1. Comparison of rock activity concentration in rocks in similar studies.

<table>
<thead>
<tr>
<th>Location (sample type)</th>
<th>Activity Concentration (Bq/kg)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$^{238}$U</td>
<td>$^{232}$Th</td>
</tr>
<tr>
<td>Mayo Belwa (granite)</td>
<td>74.59</td>
<td>104.41</td>
</tr>
<tr>
<td>Mayo Belwa (banded gneiss)</td>
<td>28.1</td>
<td>38.92</td>
</tr>
<tr>
<td>Mayo Belwa (migmatites)</td>
<td>50.19</td>
<td>60.50</td>
</tr>
<tr>
<td>Minna, Nigeria (granite)</td>
<td>27</td>
<td>48</td>
</tr>
<tr>
<td>Okene (granite)</td>
<td>58.14</td>
<td>27.3</td>
</tr>
<tr>
<td>Okene (granite)</td>
<td>28.47</td>
<td>23.91</td>
</tr>
<tr>
<td>Okene (banded gneiss)</td>
<td>31.5</td>
<td>44.28</td>
</tr>
<tr>
<td>Asa, Nigeria (granite)</td>
<td>11.51</td>
<td>15.42</td>
</tr>
<tr>
<td>Abuja (granite)</td>
<td>74.74</td>
<td>199.23</td>
</tr>
<tr>
<td>Malasia (granite)</td>
<td>67</td>
<td>85</td>
</tr>
<tr>
<td>China (migmatite)</td>
<td>34.8</td>
<td>19.9</td>
</tr>
<tr>
<td>European Union (granite)</td>
<td>78</td>
<td>89</td>
</tr>
<tr>
<td>USA</td>
<td>57</td>
<td>69</td>
</tr>
<tr>
<td>Global Mean (granite)</td>
<td>81</td>
<td>105</td>
</tr>
<tr>
<td>World average (soil)</td>
<td>32</td>
<td>45</td>
</tr>
</tbody>
</table>

Table 1 shows that the radionuclide concentration in the present study varies widely with those in previous studies. The mean activity concentration of $^{238}$U and $^{232}$Th, in granite obtained from this study is greater than that obtained in Okene, Minna, and Asa all in Nigeria. This could be attributed to the presence of uranium and thorium bearing minerals in the granites found in the study area. More so, Adamawa area of Nigeria has been recorded to host uranium deposits in the Mika and Mayo Lope area of Adamawa state (Oshin and Rahaman, 1986). The proximity of the study area to these areas suggests that such deposits could extend to the study area. The extent of such influence would have to be investigated. The concentration was however found to be lower compared to granites collected in Abuja Nigeria, European Union and the global average in granite. The concentration of $^{40}$K, $^{238}$U and $^{232}$Th, in the examined granite was also found to be below world average in soil. Figure 5 provides a qualitative description of the total activity level of Mayo Belwa area based on the collected rocks in comparison to the mean global level. It is obvious that the global mean activities of $^{238}$U (32 Bq/kg), $^{232}$Th (45 Bq/kg), and $^{40}$K (420 Bq/kg) in soil and rock samples is less than that of the study area (51, 68, 776 Bq/kg respectively). The study area could thus be described as area of relatively high activity concentration. The variation in the activity concentration in the study area and those obtained in different geological composition is mainly due to the differences in the geological definition and minerals embedded in the rocks.

CONCLUSION
Nigeria is blessed with abundant natural resources hosted in many rock species found across the country. This has led to the exploitations of rocks in many areas as well as using the rocks for building construction. These often led to the redistribution of radiation in the human habitat. The Mayo Belwa axis of Adamawa State is a host to different outcrops of Granite,
Gneiss and Migmatite. These rocks are mined for a lot reasons of which the quest for building materials is one. The amount of natural radionuclide presents in these three species of rocks (Banded Gneiss, Granite, and Migmatite) collected from the Mayo Belwa area of Adamawa State has been quantified in this research. Ten samples each of these rocks were assayed for their NORM (\(^{238}\)U, \(^{232}\)Th, and \(^{40}\)K) content via a gamma spectrometry process based on a well calibrated NaI detector. The trend of the specific activities of the radioisotopes were found to be in the trend: \(^{40}\)K > \(^{232}\)Th > \(^{238}\)U in most of the samples. The trend was attributed to the natural abundance of the isotopes in undisturbed environment. The variations of radionuclide distributions in the rock samples were attributed to the differences in the geological definition and host minerals in the rocks. The concentration of \(^{40}\)K, \(^{238}\)U and \(^{232}\)Th, in examined granite rocks was found to be below world average in granite stones. The radionuclide distribution in the rocks were above world average level in soils. Therefore, it is highly recommended that the mineral content of the rock responsible for the high radionuclide concentration should be investigated.

REFERENCES


