Assessment of natural radioactivity levels in the Ajali Formation, Enugu, South Eastern Nigeria

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Abstract. Activity concentrations of the radionuclides (238U, 232Th and 40K) were analysed using high resolution co-axial HPGe gamma ray spectrometer system to know the implication where groundwater is sourced from Enugu, South-East Nigeria. The activity concentration of 238U ranges from 37 ± 4 to 74 ± 6 Bq kg\(^{-1}\) with the highest value of 74 ± 6 Bq kg\(^{-1}\) noted in the thin coal sample. The 232Th activity level in the rock samples ranges from 58 ± 5 to 85 ± 7 Bq kg\(^{-1}\) with the higher value of 89 ± 7 Bq kg\(^{-1}\) reported in the coal sample. For 40K, the activity concentration varies from 140 ± 19 to 293 ± 25 Bq kg\(^{-1}\) with the highest value of 293 ± 25 Bq kg\(^{-1}\) reported in coal samples whereas lowest value of 140 ± 19 Bq kg\(^{-1}\) was noted in Ajali sandstone in the study area (Enugu State). The higher activity levels of 238U, 232Th and 40K reported in coal samples may be due to the closer clusters of weathered surface of Ajali sandstone that overlays the thin coal layer with an escarpment being spread out, implying higher permeability towards deeper direction. All the values are within the recommended level when compared with United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000). Further research on groundwater activity concentration and rock geochemistry is required within the study area. This will allow for a comprehensive conclusion to be drawn on the level of exposures to the inhabitants relying on groundwater for consumption.

1. Introduction
The activity concentrations of 238U, 232Th and 40K usually detected in groundwater results from the available activity concentrations of 238U and 232Th of aquifer bearing formations as well as other decay products found in rocks beneath. Conversely, the contamination of groundwater within the region occurs when there is a reaction between groundwater, soil and bedrock. However, this process appears not to be the only source of dissolved minerals finding its way into groundwater. According to [1], the other factors contributing to deposition of these dissolved minerals includes: the chemical composition of the water, the rate at which weathering of the subsurface rock takes place, the redox conditions as...
well as the residence time of groundwater in subsurface water bearing formation. However, by which ever means a reaction occur, there is a release of dissolved mineral components which is based on the mineralogical and geochemical composition of the subsurface [2,3]; and can result in a threat to the health of those that consume it unknowingly. Therefore, it is necessary to conduct an environmental assessment of borehole sites to clear public concerns on the possible risk of consuming contaminated water. Therefore, this present study is conducted to investigate the activity levels of natural occurring radionuclides where water is drawn for the consumption of many within the study area.

2. Geology and geographical location of the study area
Figure 1 shows that the area under study lies within the Cretaceous Anambra Basin of South-Eastern region of Nigeria. It occurs as a roughly triangular feature sandwiched between the Benue Trough to the North, East and the Niger Delta to the South. The Nkporo shales, Enugu shales, Afikpo sandstone and Owelli sandstone together constitute the Nkporo Group [4,5].

![Geologic map of the study area.](image)

3. Materials and methods

3.1. Lithological rock samples for this study
Lithologically, four samples were collected in the study area (close to mile 9 expressway, Enugu) between the range of 15 to 250 m below ground level where most of the groundwater resources that serve the entire state is tapped. The Ajali Formation consists essentially of sand with minor occurrences of silt/clay heteroliths. They are generally classified as quartz arenites, bearing quartz as the dominant mineral with feldspars virtually absent. The Ajali Formation is sandwiched between two coal bearing formations, being overlain by the Nsukka Formation and in turn overlays the Mamu Formation. The stratigraphical disposition of the samples is presented in table 1.
Table 1. The lithological rock samples and depths collected for the present study.

| Rock Samples   | Layer Sample Code | Depth (m) |
|----------------|-------------------|-----------|
| Nsukka Formation | L1               | 0-15      |
| Ajali Formation  | L2               | 15-200    |
| Mamu Formation   | L3               | 215-250   |

3.2. Preparation of the samples and method of sampling
Prior to the analysis, four (4) rock samples were harvested from the study area and dried to remove moist under room temperature of 25-29°C for a few days and thereafter, these samples were sealed in a plastic sock before it was transported to the Nuclear Laboratory, of the Universiti Teknologi Malaysia. Second, rock samples were crushed to powder and passed through 250 µm Sieve mesh using a Sieve shaker. Thereafter, the fine samples which passed through the shaker were homogenized and then weighed using electronic balance of ± 0.01 g precision. After that, the prepared samples were collected into a 500 mL Marinelli beakers and labelled appropriately and sealed to prevent mixing up of the samples as well as escape of radionuclides. Lastly, the samples were kept for four weeks before any analysis took place to achieve secular equilibrium between radium and its progeny [6-8].

3.2.1. Determination of γ- spectroscopy. This was achieved using high purity germanium (HPGe) gamma ray spectroscopy, which had a counting efficiency of 20% and resolution of (FWHM) 1.8 keV for 1332 keV gamma ray emission of ⁶⁰Co. In addition, other experimental process was carried out according to procedures outlined in the literature [9-11].

3.3. Calculation of the Concentration of ²³⁸U, ²³²Th and ⁴⁰K
As a result of the low life-time of the radionuclides within the disintegration series of ²³⁸U and ²³²Th, the ²³⁸U concentration were gotten from the average concentrations of ²¹⁴Pb at 352 keV and ²¹⁴Bi at 609 keV in the sample, while that of ²³²Th were obtained from the average concentrations of ²⁰⁸Tl at 583 keV and ²²⁸Ac at 911 kev decay products [6,9-11].

4. Results and discussions
The activity concentrations measured from the samples are presented in table 2. The results show the various activity levels of ²³⁸U, ²³²Th and ⁴⁰K for the four (4) samples collected.

Table 2. Activity concentration of naturally occurring radionuclides in the lithological units of subsurface formation in the coal mining area, Nigeria.

| Rock Samples  | Activity Concentrations of samples measured in Bq kg⁻¹ |
|---------------|------------------------------------------------------|
|               | ²³⁸U   | ²³²Th   | ⁴⁰K     |
| Nsukka Formation | 37 ± 4 | 63 ± 6 | 207 ± 27 |
| Ajali Formation   | 47 ± 5 | 58 ± 5 | 140 ± 19 |
| Mamu Formation    | 57 ± 6 | 85 ± 7 | 214 ± 28 |

4.1. Activity concentrations of the naturally occurring radionuclides present in the samples
The activity concentration of ²³⁸U from the samples obtained between Nsukka Formation to Mamu Formations below ground level varied from 37 ± 4 to 74 ± 6 Bq kg⁻¹ as presented in table 2. The highest value noted in thin coal bed sample with a value of 74 ± 6 Bq kg⁻¹ while the lowest value of 37 ± 4 Bq kg⁻¹ was observed in sample obtained from the Nsukka Formation. Furthermore, from table 2, the activity concentration of ²³²Th in sampled gotten from the subsurface in the Nsukka Formation to
Mamu Formations varied from $58 \pm 5$ to $85 \pm 7$ Bq kg$^{-1}$. The highest value of $89 \pm 7$ Bq kg$^{-1}$ was recorded in coal bed sample, while the lowest value of $58 \pm 5$ Bq kg$^{-1}$ was observed in the Ajali sandstone samples. In addition, the activity concentration of $^{40}\text{K}$ in the rock samples varied from $140 \pm 19$ to $293 \pm 25$ Bq kg$^{-1}$ with the highest value of $293 \pm 25$ Bq kg$^{-1}$ noted in Mamu Formation, whereas the lowest value of $140 \pm 19$ Bq kg$^{-1}$ reported in Ajali Formation. The corresponding results obtained in the present study showed that the activity concentrations of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ are within the range and in agreement with the world standard values (table 3). However, the values obtained from this study were higher than those obtained from maiganga coal area, Nigeria [12] by factors of 7.82, 7.75 and 9.05 for Potassium-40, Thorium-232 and Uranium-238, respectively. These higher values may be due to the intercalation of naturally occurring radionuclides concentrations in Ajali sandstone and thin slim coal formation that overlies aquifer. Significantly, this may have elevated the concentration $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ in the samples.

Table 3. Comparism of activity concentration of samples obtained from some states in Nigeria and world standard.

| Region/ Country          | $^{232}\text{Th}$ (Bq kg$^{-1}$) | $^{238}\text{U}$ (Bq kg$^{-1}$) | $^{40}\text{K}$ (Bq kg$^{-1}$) |
|-------------------------|----------------------------------|---------------------------------|--------------------------------|
|                         | Range  | Mean  | Range  | Mean  | Range  | Mean  |
| Enugu, South Eastern    | 58-89  | 54    | 37-74  | 74    | 140-293| 214   |
| Nigeria$^a$             |        |       |        |       |        |       |
| Dei-De1, Abuja, North   | 45-98  | 67    | 18-37  | 30    | 119-750| 830   |
| Central Nigeria$^b$     |        |       |        |       |        |       |
| Kubwa, Abuja, North     | 32-84  | 61    | 15-52  | 34    | 236-1195| 573   |
| Central, Nigeria$^b$    |        |       |        |       |        |       |
| Ikogosi-Ekiti, South    | 1-108  | 82    | 4-111  | 58    | 40-2437| 1203  |
| western Nigeria$^c$     |        |       |        |       |        |       |
| World$^d$               | 7-50   | 45    | 16-116 | 33    | 100-700| 420   |

| a- present study        |        |       |        |       |        |       |
| b- [6,7]                |        |       |        |       |        |       |
| c- [13]                 |        |       |        |       |        |       |
| d- [14]                 |        |       |        |       |        |       |

5. Conclusions

The activity concentrations of naturally occurring radionuclides obtained from the subsurface layers within Ninth Mile Ajali sandstone water bearing formation identified thin coal bed as the major radioactivity source from the U-series. In addition, this study reveals that these factors: 7.83 for Potassium-40, 7.75 for Thorium-232 and 9.05 for Uranium-238 were higher than those obtained by Kolo et al studies from coal formation area in Nigeria [12]. Geologically, these higher values obtained from this present study could be attributed to the interbedding of Ajali sandstone that overlies the thin coal formation from which groundwater is tapped. Significantly, the results obtained from this study clearly indicate that the higher activity levels of radionuclides noted in water bearing formation may pose health risk of contamination on the groundwater due to water-rock interaction. Further work on groundwater analysis for both radioactive and heavy metal contents in the water is required.

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