Analysis of Natural Radionuclides and Evaluation of Radiation Hazard Indices in Soil Samples From Benue State, Nigeria

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Abstract:- The study analyzed the specific activity concentrations in surface soil samples in Benue State, Nigeria and the potential radiological hazards indices using a gamma ray spectrometer fitted with NaI(Ti) detector. The naturally occurring radionuclides Radium, Thorium and Potassium contents were determined for nine different locations within the study area. The average activity concentrations of these radionuclides were found to be 39.10 ± 2.67 , 29.44 ± 0.99 and 113.02 ± 2.78 Bg/kg for samples from zone A; 41.86 \pm 3.03, 8.14 \pm 0.35 and 404.24 \pm 5.92 for samples from Zone B and 22.48 ± 1.92, 8.93 ± 0.25 and 164.58 ± 2.06 for samples from zone C. The Raeg values averaged 77.2, 75.8 and 48.0 Bq/kg. These are less than the recommended maximum permissible value of 370 Bq/Kg. The average absorbed dose rates had calculated values of 40.5, 39.6 and 22.6 nGy/h respectively and are well below the global value of 55nGy/h and the annual effective dose rates had average values of 5.0, 4.9 and 2.8 mSv/y which are above the 1 mSv/y recommended. The average Hex and Hin values were below unity. The study established the presence of natural radionuclides, ²²⁶Ra, ²³²Th and ⁴⁰K in soils within the study area and has also provided information for regulatory agencies to use as a guideline for estimation and monitoring environmental radioactivity.

Keywords:- Natural radioactivity, Radiation hazards, Gamma Spectroscopy, Activity concentrations, Radium equivalent.

I. INTRODUCTION

Benue State is one of the north central area states in Nigeria and is divided into three (3) senatorial zones A, B and C. The state is unquestionably one of the most important in the country in terms of the production of crops for the sustenance of its population. It is a rich agriculture region known for the production of oranges, mangoes, soya beans, yams, sweet potatoes, cassava, guinea corn, rice, groundnuts, sesame, tomatoes, millet and many others on its soils (Government of Benue State, 2017). However, it is well known fact that traces of naturally occurring radionuclides can be found in the atmosphere, soil, water and the human bodies (Sharama, Sharama & Virk, 2011). These radionuclides can be inhaled and ingested on a daily basis since they have been present on earth right from creation. The presence of naturally occurring radionuclides in soil results in internal and external exposure to human beings (El-Taher & Abdel Halim, 2014). Basically, radionuclides which are found in nature are basically divided in two distinguishable parts, either Cosmogenic or Terrestrial origin (El-Taher & Al-Zahrani, 2014). The common radionuclides that irradiate the human body due to external exposure primarily by gamma radiation are ²³⁸U, ²³⁵U and ²³²Th and their subsequent radioactive decay products and ⁴⁰K (Alashrah & El-Taher, 2016; Knoll, 2000).

The basic aim of this work therefore is to determine the specific activity concentrations of naturally occurring radionuclides; radium, thorium and potassium and the associated radiation hazards in soil samples from Benue State.

II. MATERIALS AND METHODS

Soil samples were collected from nine (9) locations from a vertical depth of 15-30cm and surface diameter of about 15cm from the study area. The collected samples were transferred into well coded plastic bags, numbered for sample location and then the bags sealed in order to avoid cross contamination of the samples and were taken to the research centre for measurement of natural radioactivity. The soil sampling locations and their codes are summarized in Table 1.

| Sample code | s Sampling Area | Local Govt. Are | ea Zones |
|-------------|-----------------|-----------------|----------|
| S1 | Ikov | Ushongo | А |
| S2 | Mbake | Konshisha | А |
| S3 | Jato-Aka | Kwande | А |
| S4 | Binev | Buruku | В |
| S5 | Mbakwen | Gboko | В |
| S6 | Mbagwen | Makurdi | В |
| S7 | Umogidi | Oturkpo | С |
| S8 | Oraha | Oju | С |
| S9 | Eke-Akpa | Ogbadibo | С |

Table 1:- Soil Sample Codes, sampling area with locality and zones from Benue State

A. Sample preparation

At the Centre, samples were taken to the laboratory where they were dried at 100°C for 24hours in an electric oven to remove moisture. They were then ground into fine powder using a pulverizer and sieved through a 100-micron mesh to obtain a homogenized sample with uniform grain size for measurements. The samples were then packed into radon-impermeable cylindrical plastic containers based on the space allocation of the detector vessel which measured 7.6cm by 7.6cm in dimension. The samples were stored for 30 days to allow radon and its short-lived progenies to reach secular radioactive equilibrium prior to gamma spectroscopy measurements.

Calculation of specific activities concentration, hazard indices and dose parameters

The data acquisition was carried out using a 7.6×7.6 cm NaI (TI) detector crystal optically coupled to a photomultiplier tube (PMT) and consisted of a preamplifier incorporated into it and a 1kilovolt external electricity source. The detector was enclosed in a 6cm lead shielded with cadmium and copper sheets. This arrangement was aimed at minimizing the effects of background and scattered radiation.

The samples were placed in the detector cavity and measured for a period of 29000 seconds, for each radionuclide. The peak area of each energy in the spectrum was used to compute the specific activity concentrations in the sample as well as other radiological hazards.

> Calculating the Specific Activity Concentrations

The calculation for the specific activity concentrations was done using the procedure given by Lalit, Shukala, Ramachandran, & Mishra (1982), Kolo, Baba-Kutigi, Olarinoye & Sharifat (2012), Abbady, Uosif & El-Taher (2005):

$$A_{s}(Bq/kg) = \frac{ca}{\epsilon PrMs}$$
(1)

where C_a is the net gamma counting rate (counts per second), ε is the detector efficiency of the specific γ -ray, P_r the absolute transition probability of Gamma-decay and M_s the mass of the sample (kg).

Radium Equivalent Activity (Bq/kg)

The index, Ra_{eq} was calculated using the formula given below based on the assumption that 370Bq/kg of ²²⁶Ra, 259Bq/kg of ²³²Th and 4810Bq/kg of ⁴⁰K produce the same gamma-ray dose rate (Beretka & Mathew, 1985):

$$Ra_{eq} (Bq/kg) = A_{Ra} + 1.43A_{Th} + 0.077A_k$$
(2)

Where A_{Ra} , A_{Th} and A_k are the activity concentrations (Bq/kg dry weight) of 226 Ra, 232 Th and 40 K respectively.

> Absorbed Dose rate and Annual Effective Dose rate

The specific activity concentrations of 226 Ra, 232 Th and 40 K were converted into doses by applying the factors 0.462, 0.604 and 0.0417 for radium, thorium and potassium respectively (UNSCEAR.2000). These factors were used to calculate the total absorbed gamma dose rate in air at 1m above the ground level using the following equation: Absorbed dose rate, D (nGy/h) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_k (3)

The annual effective dose rate (AEDR) is obtained to test the health effect of those absorbed dose rates. To estimate annual effective dose rates, one has to take into account of (i) the conversion coefficient from absorbed dose rate in air to effective dose rate and (ii) the outdoor occupancy factor. The annual effective dose rates are determined using (UNSCEAR. 2000) as follows:

Annual effective dose rate = $D(nGy/h) \times 8760h \times 0.2 \times 0.7$ Sv/Gy x 10⁻⁶ (4)

\succ External and internal hazard index (H_{ex} , H_{in})

The external (H_{ex}) and internal (H_{in}) hazard index are calculated from the equations (UNSCEAR, 2000; Righi & Bruzzi, 2006):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \le 1$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \le 1$$
(5)

In (5), values of external and internal hazard index must be ≤ 1 , which corresponds to the upper limit of Ra_{eq} (370Bq/kg), in order to keep the radiation hazard in check.

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III. RESULT PRESENTATION AND DISCUSSION

The measured specific activity concentrations, radium equivalent activity (Ra_{eq}), absolved dose rates (D), annual effective dose rates (AEDR) and external and internal hazard index (H_{ex} , H_{in}) of naturally occurring radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K in the soil samples are shown in Table 2, 3, 4 and 5.

| | Specific Activity concentration (Bq/Kg) | | | |
|---------------|---|-------------------|-------------------|--|
| Zone A | 226 Ra | ²³² Th | 40 K | |
| S1 | 42.67±2.912 | 11.09±1.69 | 214.29±4.08 | |
| S2 | 55.42±2.72 | 7.04±0.16 | 74.33±3.32 | |
| S3 | 19.09±2.36 | 70.18±1.14 | 50.46±0.91 | |
| Average Value | 39.10± 2.67 | 29.44 ± 0.99 | 113.02 ± 2.78 | |

Table 2:- The specific activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in soil samples from Zone A

| | Specific Activity concentration (Bq/Kg) | | | |
|---------------|---|-------------------|-------------|--|
| Zone B | ²²⁶ Ra | ²³² Th | 40 K | |
| S4 | 54.42±2.84 | 9.24±0.47 | 468.39±6.11 | |
| S5 | 15.74±3.55 | 0.43±0.43 | 273.29±6.54 | |
| S6 | 55.42±2.72 | 7.04±0.16 | 471.01±5.09 | |
| Average Value | 41.86±3.03 | 8.14±0.35 | 404.23±5.92 | |

Table 3:- The specific activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in soilfrom Zone B

| | Specific Activity concentration (Bq/Kg) | | | |
|--------------------|---|--------------------------------|-----------------|--|
| Zone C | ²²⁶ Ra | ²³² Th | ⁴⁰ K | |
| S7 | 36.76±3.32 | 5.58±0.24 | 43.12±0.91 | |
| S8 | 4.52±1.68 | 6.33±0.47 | 93.53±1.93 | |
| S9 | 26.17±0.76 | 14.86±0.04 | 357.11±3.33 | |
| Average Value | 22.48±1.92 | 8.93±0.25 | 164.58±2.06 | |
| Table 4. The speci | fig activity concentrations of 226D | 232Th and 40K in soil complete | from Zono C | |

Table 4:- The specific activity concentrations of 226 Ra, 232 Th and 40 K in soil samples from Zone C

| Soil samples | Raeq(Bq/kg) | Absorbed dose Rate | AEDR | Hex | Hin |
|---------------|-------------|--------------------|------|------|------|
| | | (D) | | | |
| S1 | 37.03 | 35.35 | 4.34 | 0.20 | 0.32 |
| S2 | 71.21 | 32.95 | 4.04 | 0.19 | 0.34 |
| S3 | 123.34 | 53.32 | 6.54 | 0.33 | 0.38 |
| Average value | 77.2 | 40.5 | 5.0 | 0.24 | 0.35 |
| S 4 | 103.70 | 50.26 | 6.16 | 0.28 | 0.43 |
| S5 | 22.00 | 18.93 | 2.32 | 0.10 | 0.14 |
| S 6 | 101.75 | 49.50 | 6.07 | 0.27 | 0.43 |
| Average value | 75.8 | 39.6 | 4.9 | 0.22 | 0.33 |
| S7 | 48.06 | 22.15 | 2.72 | 0.13 | 0.22 |
| S8 | 20.77 | 9.81 | 1.20 | 0.06 | 0.07 |
| S9 | 74.92 | 35.96 | 4.41 | 0.20 | 0.27 |
| Average value | 48.0 | 22.6 | 2.8 | 0.13 | 0.19 |

 Table 5:- Absorbed dose rate, annual effective dose rate, radium equivalent activity, index of external and internal radiation hazard for soil samples.

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The measured specific activity concentrations of naturally occurring radionuclides $^{226}\mbox{Ra},\,^{232}\mbox{Th}$ and $^{40}\mbox{K}$ in the soil samples are shown in Tables 2, 3 and 4. They were found to have average values of 39.10 ± 2.67 , 29.44 ± 0.99 and 113.02 ± 2.78 Bq/kg for samples from zone A; $41.86 \pm$ 3.03, 8.14 \pm 0.35 and 404.24 \pm 5.92 for samples from Zone B, and 22.48 \pm 1.92, 8.93 \pm 0.25 and 164.58 \pm 2.06 for samples from zone C respectively.

The radionuclides activity concentration values in the soil samples were higher than the worldwide weighted mean of 33Bq/kg for ²²⁶Ra for zone A and B (UNSCEAR, 2000), but less than the worldwide weighted mean for zone C. Also, ²³²Th and ⁴⁰K mean activity concentrations were

less than the 45Bq/kg for ²³²Th and 420Bq/kg for ⁴⁰K worldwide weighted mean in zones A, B and C of the study area (UNSCEAR, 2000). The difference in the specific activities for samples from zone A and B could be attributed partly to the geological structure of the area, see figure (1).

Also from table (5) it can be noticed that, Ra_{eq} had values averaged 77.2. 75.8 and 48.0 Bq/kg. It can be seen that the Raeq values for all soil samples measured were less than the recommended maximum permissible value of 370 Bq/Kg (UNSCEAR, 2000).



Fig 1:- The specific activity concentration of ²²⁶Ra, ²³²Th and ⁴⁰K for soil samples from the three Zones

For the absorbed dose rates and annual effective dose rates (table 5), result show that the average values were 40.5, 39.6 and 22.6 nGy/h and 5.0, 4.9 and 2.8 mSv/y respectively. These values of absorbed dose rates within the study area are under the limit of the global value of 55 nGy/h (UNSCEAR, 2000), while those for the annual effective dose rates are above the 1 mSv/y recommended by (UNSCEAR, 2000). This could be explained by the origin of the radionuclides as well as by the weathering of rocks within the area.

The values of H_{in} and H_{ex} were averaged between (0.35) and (0.24) and from (0.33) and (0.22) and 0.19 and 0.13 respectively for the area studied and are below the recommended value of unity (1) given by UNSCEAR, 2000.

IV. CONCLUSIONS

Based on the results obtained from the analysis of the samples within the study area, it is therefore concluded that;

- The soils from the study area contain radionuclides with \geq their specific activity concentrations higher in Zone A and B, comparable to the world set limits.
- \geq The radium equivalent activity is within the acceptable limits.

- > The absorbed dose rates are within the limits of 55nGy/h, while the annual effective dose rates are above 1mSv/y.
- The hazard indices measured are less than unity (1).

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