DETERMINATION OF RADIOACTIVITY LEVELS IN SOIL SAMPLES AT CHIKUN ENVIRONMENT OF KADUNA METROPOLIS USING GAMMA RAY SPECTROMETRY

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ABSTRACT
Fourteen (14) soil samples were collected in Chikun Local Government Area of Kaduna metropolis. The radio activity level in samples were detected and analysed by employing 76×76mm NaI (Tl) detector crystal. The average activity of $^{226}$Ra, $^{232}$Th and $^{40}$K were found to be 62.2287Bq/kg, 155.3592 Bq/kg and 459.5646 Bq/kg, respectively. The mean activity of $^{226}$Ra is lower than the world average of 400Bq/kg, while those of $^{232}$Th and $^{40}$K are higher than the world average of 50Bq/kg and 420 Bq/kg, respectively.

Keywords: Radionuclides; soil samples, activity concentration, Chikun Area, Kaduna Metropolis.

INTRODUCTION
Exposure to ionizing radiation originates from two major sources, namely naturally occurring and man-made sources. Naturally occurring radioactivity present on the earth’s crust can be further classified into two distinct categories such as virgin and modified natural sources. Virgin sources of radiation are of cosmogenic or primordial (terrestrial) origin and have existed on the earth since primordial times. Modified natural sources are mainly from activities like mining, usage of fossil fuel, production of fertilizers or usage of natural materials for building constructions. The latter is known as Technologically Enhanced Natural Radiation (TENR).

Nuclear radiation is the largest contributor to the collective world radiation dose rate. The major contribution of high dose from natural radiation in normal background regions arise due to inhalation of radon and its progeny (UNSCEAR, 2000). Gamma radiation from radionuclides, such as $^{40}$K and $^{232}$Th series, and their decay products, represents the main external source of irradiation to the human body. The absorbed dose rate in air from cosmic radiation outdoor or sea level is about 30 nGy-h$^{-1}$ (UNSCEAR, 2000).

External exposures to gamma radiation outdoors arise from terrestrial radio nuclides occurring at trace levels in all rock formations. Therefore, the natural environmental radiation mainly depends on geological and geographical condition (Nagaraja and SaiLith, 2010). These have direct effect by influencing the soil composition and natural radioactivity concentration levels; and hence the level of $\gamma$-absorbed dose received at a locality. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shale and phosphate rocks have relatively high content of radio nuclides (Ramachandra et al, 1995).

Ionizing radiation has always been part of the human environment. Along with natural radioactive sources present in the earth crust and cosmic radiation. Man’s discovery of atomic and nuclear energy with advance in technology, the uses of man produced radiation, especially in the production of elastic power are being continuously multiplied, so are man's chances of exposure to harmful radiation (Agbalaje, 2010).

This research work determines the levels of the primordial radionuclides $^{40}$K, $^{232}$Th and $^{238}$U in the selected sampling sites using a 76×76mm NaI (Tl) detector crystal, primarily for monitoring rapid identification and assessment of radioactivity and absorbed dose rates in air.

MATERIALS AND METHOD
In this research Rados meter (RDS 120) was used for background mapping. The Rados is a versatile gamma radiation detector designed for wide range of applications involving the detection of abnormal or elevated radiation levels. It could also be used to determine background radiation of a place. Its performance and its user friendly interface makes it a perfectly suitable devices for monitoring land detections of radiological hazards.

Monitoring of any release of radioactivity to the environment is important for environmental protection against ionizing radiation. Rapid and accurate methods for the measurements are essential. Many important isotopes in the natural occurring radioactive materials(NORM) and technology enhance natural occurring radioactive materials(TENORM) have some suitable gamma rays, allowing qualitative and quantitative determination of the radio nuclides by high-resolution gamma spectrometry. Measurements of radiation levels and the concentrations of radio nuclides in the environment are accomplished employing appropriate nuclear instruments. Then radioactivity level were detected and analysed employing 76×76mm NaI (Tl) detector crystal optically coupled to a photomultiplier tube (PMT), to determine each of the radioactivity level of these three radio nuclides, there is spectral energy window and energy calibration as presented in table 1 and 2.

Soil Sample Preparation
Each of the soil samples collected were dried and crushed to fine powder with the use of pulveriser. Packaging of the samples into radon-impermeable cylindrical plastic containers which were selected based on the space allocation of the detector vessel which measures 76×76mm in dimension (geometry) was also carried out to prevent $^{222}$Ra from escaping; the packaging in each case was triple sealed.

Determination of Radioactivity Levels in Soil Samples at Chikun Environment of Kaduna Metropolis Using Gamma Ray Spectrometry
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. After the samples were prepared, each empty container was weighed to know the weight of the empty container and then when the sample was sealed into the container it was then weighed again to know the weight of empty container and soil sample, then to get the weight of sample you are to subtract the weight of the empty container from the weight of container + sample. Radon and its short-lived progenies were allowed to reach secular radioactive equilibrium by storing the samples for 30 days at ambient temperature prior to gamma spectrometry measurements.

**Gamma Ray Spectrometry**

Gamma spectrometry offers a convenient, direct and non-destructive method for the measurement of the activity of different radionuclides in environmental samples. Gamma spectrometry techniques offer high efficiency (NaI(Tl) detectors) and high resolution semiconductors detection. The technique enables the use of large quantity of samples for counting. Liquid nitrogen cooling is required only when the detectors are in use. Hence, gamma ray spectrometer was employed in the present work to determine the activity concentration of nuclear radionuclides in environmental soil samples.

**Evaluation of Radioactivity of Samples**

The analysis was carried out using a 76×76mm NaI(Tl) detector crystal optically coupled to a photomultiplier tube (PMT). The assembly has a preamble incorporated into it and a 1 kilovolt external source. The detector is enclosed in a 6 cm lead shield to block the radiation. The data acquisition is made by Canberra nuclear (Ibeau, 1999).

\[
c(Bq.kg^{-1}) = \frac{C_{n}}{f_{k}}
\]

Where \( C \) is the activity concentration of the radionuclides in the sample, \( C_{k} \) is Calibration factor of the detecting system, \( C_{n} \) is count rate (counts per second) and \( f_{k} \) is the Calibration Factors

\[
Counts\ per\ second\ (cps) = \frac{net\ count}{lifetime}
\]

**Calibration and Efficiency Determinations**

Calibration of the system for energy and efficiency were done using two calibration point source; Cs-137 and Co-60. These were done with the amplifier gain that gives 72% energy resolution for the 66.16 keV of Cs-137 and counted for 30 mins.

**Backgrounds Readings**

The background count rate was done for 29000 seconds (10hrs 50mins) and Table 1 shows the spectral energy window used in the analysis.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Gamma Energy (keV)</th>
<th>Energy Window (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ra−226</td>
<td>1764.000</td>
<td>1620−1820</td>
</tr>
<tr>
<td>Th−232</td>
<td>2814.500</td>
<td>2480 − 2820</td>
</tr>
<tr>
<td>K−40</td>
<td>1460.000</td>
<td>1380 − 1550</td>
</tr>
</tbody>
</table>

**Spectra Analysis**

The computer based multi-channel analyser system with emulsion software (MAESTRO-32) was used for spectra acquisition. Based on two point energy calibration as set for the operation, the prominent peaks were identified in a bench-mark spectrum and the appropriate regions of interest were set up. These peaks, which are characteristic of typical environment spectral are:

- The 295, 352, 607, 1120 and 1765 keV peaks in the Uranium series,
- The 238, 510, 583, 911, 965 and 2615 keV peaks in the Thorium series and
- The 1460 keV peak of potassium.

The set energy bands define the peaks of where the left and right channel markers are representative of the Compton continuum. Detector’s specific calibration factors (efficiency) were applied to convert from net count rate to concentration. Only peaks with reasonable γ-ray emission probability were considered.

**Calculation of Activities Concentration**

Following spectrum analysis, calculation of count rates for each detected photo peaks and radiological concentration (activities per unit mass or specific activities) of detected radionuclides depends on the establishment of secular equilibrium was reached between \(^{238}U\) and their decay products. The \(^{232}Th\) was determined from the average concentration of \(^{204}Tl\) and \(^{227}Ac\), and that of \(^{238}U\) was determined from the average concentration of \(^{214}Pb\) and \(^{208}Bi\) decay products. Thus, accurate radionuclide concentration of \(^{232}Th\) and \(^{238}U\) were determined, whereas a true measurement of \(^{40}K\) concentration was made.

**Table 2 Energy Calibration for Quantitative Spectral Analysis**

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Calibration Factors</th>
<th>Conversion Factors(Bq/kg)</th>
<th>Detection Limits</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10^3cps/ppm</td>
<td>10^3cps/ppm</td>
<td></td>
</tr>
<tr>
<td>40K</td>
<td>0.026</td>
<td>6.431</td>
<td>545.4</td>
</tr>
<tr>
<td>85Ra</td>
<td>10.500</td>
<td>8.632</td>
<td>12200</td>
</tr>
<tr>
<td>223Th</td>
<td>3.612</td>
<td>8.768</td>
<td>22.7</td>
</tr>
</tbody>
</table>

**Determination of Radioactivity Levels in Soil Samples at Chikun Environment of Kaduna Metropolis Using Gamma Ray Spectrometry**
The specific activity \((Bq \, kg^{-1})A_{Ei}\) of a nuclide and for a peak at energy \(E\), is given by
\[
A_{Ei} = \frac{N_{Ei}}{\varepsilon_E \cdot t \cdot \gamma_{yd}}
\]
(3)

Where \(N_{Ei}\) is the net peak area of a peak energy \(E\)
\(\varepsilon_E\) is the detection efficiency at energy \(N\)
\(t\) is the counting live-time
\(\gamma_{yd}\) is the gamma ray yield per disintegration of specific nuclide for a transition of energy \(E\)

The total uncertainty \((\sigma_{tot})\) and weighted systematic activity value is composed of counting statistic \((\sigma_{st})\) and weighted systematic errors \((\sigma_{sys})\) calculated by the following formula (Paramesh et al., 2003)
\[
\sigma_{tot} = \sqrt{\sigma_{st}^2 + \frac{1}{3} \sum \sigma_{sys}^2}
\]
(4)

RESULTS AND DISCUSSION
The results of radio activity levels in Fourteen (14) soil samples within Chikun Local Government Area of Kaduna metropolis were determined. Samples contain a number of radio nuclides originating from both natural and man-made sources. Distribution of radio nuclides varies from place to place that is because of the different activities within the sample area. Activity concentration of some of the radio nuclides such as \(^{226}\)Ra, \(^{232}\)Th and \(^{40}\)K were determined by direct counting methods employing the nuclear instruments. The results obtained are presented in Table 3. From the tables there was no significant difference of the result from the survey meter and the result obtained from Centre for Energy Research and Training (CER) Zaria.

Fig.1 is the graphical representation of the result from background radiation readings and gamma spectrometry analysis obtained from soil samples in Chikun L.G.A.

Table 3: Result of Background Radiation Reading at Chikun L.G.A

<table>
<thead>
<tr>
<th>Code</th>
<th>Mean of (\mu Sv/hr)</th>
<th>Error (\mu Sv/hr)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.11</td>
<td>0.03</td>
</tr>
<tr>
<td>2</td>
<td>0.12</td>
<td>0.04</td>
</tr>
<tr>
<td>3</td>
<td>0.13</td>
<td>0.05</td>
</tr>
<tr>
<td>4</td>
<td>0.14</td>
<td>0.06</td>
</tr>
<tr>
<td>5</td>
<td>0.15</td>
<td>0.07</td>
</tr>
<tr>
<td>6</td>
<td>0.16</td>
<td>0.08</td>
</tr>
<tr>
<td>7</td>
<td>0.17</td>
<td>0.09</td>
</tr>
<tr>
<td>8</td>
<td>0.18</td>
<td>0.10</td>
</tr>
</tbody>
</table>

Table 4: The Result Obtain from Soil Samples in Chikun L.G.A

<table>
<thead>
<tr>
<th>Code</th>
<th>(^{226})Ra</th>
<th>(^{232})Th</th>
<th>(^{40})K</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.1401</td>
<td>0.2401</td>
<td>0.4001</td>
</tr>
<tr>
<td>2</td>
<td>0.2402</td>
<td>0.3402</td>
<td>0.5402</td>
</tr>
<tr>
<td>3</td>
<td>0.3403</td>
<td>0.4403</td>
<td>0.6403</td>
</tr>
<tr>
<td>4</td>
<td>0.4404</td>
<td>0.5404</td>
<td>0.7404</td>
</tr>
<tr>
<td>5</td>
<td>0.5405</td>
<td>0.6405</td>
<td>0.8405</td>
</tr>
</tbody>
</table>

Fig.1 is the chart for the background radiation reading obtained in Chikun L.G.A. It shows a minimum background reading at SBR3 (0.110 ± 0.006) \(\mu Sv/hr\) and Rido2 (0.11 ± 0.006) \(\mu Sv/hr\) while KRPC1 (0.190 ± 0.008) \(\mu Sv/hr\) as the maximum background reading.
Fig 2: Chart showing Result for $^{40}$K, $^{226}$Ra and $^{232}$Th.

Fig. 2 is the chart showing the result obtained from Gamma spectrometry done in Centre for Energy Research and Training (CERT) Zaria, on the soil samples. Thus, the result shows that K-40 is more abundant among the three radio nuclides and is peaked at KRPC$_1$(889.7356 Bq/kg$^{-1}$) and KRPC$_2$ (765.3188 Bq/kg$^{-1}$) and is minimum at SBR$_1$(29.3935 Bq/kg$^{-1}$). Th-232 is the second most, which is peaked at KRPC$_3$(288.5975 Bq/kg$^{-1}$) and is minimum at GN$_1$(22.1209 Bq/kg$^{-1}$). Ra-226 is the least among the three Radio nuclides and also the minimum is at RIDO$_2$(15.9907 Bq/kg$^{-1}$).

CONCLUSION

When exposed to radiation the body absorbs some dose which causes chemical reactions to occur which may alter the normal function of the body. From the obtained result, the average exposure is 0.15 mSv and average activity of $^{226}$Ra, $^{232}$Th and $^{40}$K were found to be 62.2827 Bq/kg$^{-1}$, 155.3592 Bq/kg$^{-1}$ and 459.5646 Bq/kg$^{-1}$, respectively. Though the mean activity of $^{226}$Ra (62.2827 Bq/kg$^{-1}$) is lower than the world average of 400 Bq/kg$^{-1}$, mean activity concentration of $^{232}$Th (155.3592 Bq/kg$^{-1}$) is higher than the world average of 50 Bq/kg and the activity concentration of $^{40}$K (459.5646 Bq/kg$^{-1}$) is also higher than the world average of 420 Bq/kg$^{-1}$.

REFERENCES


