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Assessment of Radionuclides Contents in Soil and Food Samples Due to Civil War in Enugu State, South-Eastern, Nigeria and Its Radiological Hazards

¹Agbelusi, O. I., *¹Ayanlola, P. S., ²Akinlabi, I. A., ³Amusat, T. A., ¹Amuda, D. B., ¹Isola, G. A.

¹Department of Pure and Applied Physics, Ladoke Akintola University of Technology, Ogbomoso, Nigeria. ²Department of Earth Science, Ladoke Akintola University of Technology, Ogbomoso, Nigeria. ³Department of Physical Sciences Education, Emmanuel Alayande University of Education, Oyo, Nigeria

*Corresponding author's email: psayanlola28@lautech.edu.ng

Keywords: Civil war, Enugu State, Food, Nigeria, Radionuclides, Soil. **ABSTRACT** Human activities may influence the distribution of naturally occurring radioactive materials in the environment. This therefore, cause for concern radiological, as human are exposed to these radioactive materials without their consent. This study hereby evaluate the radionuclide contents from natural and artificially occurring radionuclides in soil samples and farm produce in Enugu. Gamma-ray spectroscopy was used to determine the radioactivity concentration level of the environmental radionuclides such as 238 U, 232 Th, and 40 K and manmade radionuclides in soil, cassava, and bitter leaf samples within studied locations in Enugu State, Nigeria. The mean activity concentration of the radionuclides detected in the samples were 1136.74 \pm 27.07, 1118.53 \pm 4.85, 196.104 \pm 0.02 Bqkg⁻¹ for ⁴⁰K, 41.66 \pm 0.04, 22.99 \pm 0.38, Bqkg⁻¹ for ²³⁸U and 36.44 \pm 0.04, 15.89 \pm 0.07, 7.69 \pm 0.02 Bqkg⁻¹for ²³²Th. ¹³⁷Cs ranged from $3.07 \pm 0.01 - 7.81 \pm 0.03$ Bqkg⁻¹. The radioactivity levels and estimated radiological indices were higher than the recommended limits. The values confirmed the reports of different fora on the radiological hazards due to exposure to high radiation in the region assayed.

INTRODUCTION

Radiation has always been an integral part of the human environment, and radiation exposure is a continuing and inescapable phenomenon on Earth. The environment occupied by living things is normally radioactive, and individuals are frequently exposed to radiation from the inestimable beams, characteristic radionuclides in water, air, soil, and man-made radioactivity from aftermaths of post-war activities, clinical applications e.t.c (Ademola *et al.,* 2014). Radionuclides and their decay products from 238 U and 232 Th series together with $40K$ are terrestrial primordial radionuclides that originated from the earth's crust and are the sources of natural radioactivity in the environment (Kessaratikoon and Awaekechi, 2008).

According to Kabore et al. (2017), Naturally Occurring Radioactive Materials (NORMs) are ubiquitously distributed in all living and non-living components of the biosphere. Tettey-Larbi *et al.* (2013) stated that NORMs are found in every constituent of the environment such as air, water, soil, food, and in humans. However, a variety of human activities like civil war, mining, and milling of natural resources may result in Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM) in the products and by-products of those activities

(UNSCEAR, 2000). Artificial radiation results from radioactive materials concentrated by human activities such as medical practices, nuclear reactors, post-war activities, and so on. However, both natural and artificial radiations constitute damage to biological matter.

The presence of radioactive materials and radiation generators is harmful to humans and the environment. However, exposure to radioactive material is very detrimental to the biological system, and as such the interaction alters the genetic makeup of the biological system. Radionuclides detected in plants are originally contained in soil from where they get translocated via the root system to different plants' parts or transported either by direct fallout of radionuclides and re-suspension of contaminated soils followed by deposition on plant leaves or soils within the vicinity of the plants (Noordijik *et al.,* 1992). The study therefore aimed to determine the activity concentration level in soil and food samples collected across Enugu State, South Eastern Nigeria. This study was necessitated because of several media reports on the incidences of cancer and other health challenges in some parts of the study area to which radiation may be one of the factors causing the health issues. This study will go a long way in providing baseline radiometric information which could help in future epidemiological investigation.

MATERIALS AND METHODS Study Area

Enugu State is one of the states in the Southeastern part of Nigeria where the civil war was waged half a century ago. As a result, these areas may have elevated levels of radioactivity, and associated health risks. The knowledge of radionuclides and radiation levels in the environment is therefore important for assessing the effects of radiation exposure due to terrestrial and extraterrestrial sources.

Figure 1: The map of the study area

Sampling

Soil samples were collected across different communities, by clearing the surface vegetation at each location after which soil auger was used to collect the samples at depths of 10 cm. This was in a bid to determine the presence of $137Cs$ that may be present in the soil (Akinloye *et al.,* 2012). Cassava (*Manihot esculenta*) samples were collected during the harvest period, directly from selected farmlands across the study area to ensure they were site-specific samples. The Bitter-leaf (*Vernonia amygdalina*) samples were handpicked across communities in the civil war zone. A total of seventy-five (75) samples were collected for analysis; soil (25), cassava (*Manihot esculenta*) (25), and bitter leaf (*Vernonia amygdalina*) (25), respectively. At the point of collection, samples were carefully placed in separate polythene bags, labeled, and transferred to the laboratory for further processing. The samples were air-dried, to constant weight, pulverized, and sieved to ensure uniform particle size after which they were sealed tight in containers weighing 200 g each and were kept for a minimum of 28 days for secular equilibrium to take place.

Radioactivity Measurements

After the secular equilibrium was attained, the samples were counted using a well-shielded and -calibrated sodium iodide-based gamma spectrometer detector at the Radiation and Health Physics laboratory, Ladoke Akintola University of Technology, Nigeria. Data acquisition and analysis of gamma-ray spectra were achieved using Theremino software. The activity concentration A (Bqkg⁻¹) of each identified radionuclide in the samples was calculated using Equation 1:

$$
A\left(Bqkg^{-1}\right) = \frac{c_{net}}{\text{Py} \times \text{Exm} \times \text{t}}\tag{1}
$$

where C_{net} is the net peak area, P_{γ} is the absolute gamma-ray emission probability, ε is the obtained full energy peak efficiency for each identified radionuclide, m is sample mass and t is the counting time.

Calculation of radiological hazard indices in the samples

External absorbed dose rate (DR)

The external Gamma Dose Rate (D) for the sediment samples was determined using Equation (2) from the Activity Concentrations at about 1.0 m above ground (Uosif et al., 2014)

$$
D (nGyh^{-1}) = 0.462 A_u + (0.621 A_{Th}) + (0.0417 A_k)
$$

(2)

Where, A_U , A_{Th} and A_K are the activity concentration of ²³⁸U, ²³²Th and ⁴⁰K respectively.

Annual Effective Dose Equivalent (AEDE)

The effective dose in millisieverts per year was calculated using Equation (3)

 $AEDE$ $(mSvyr^{-1}) = D$ $(nGyh^{-1}) \times 8760(hy^{-1}) \times 0.2 \times 10^{-1}$ $0.7 (SvGy^{-1}) \times 10^{-6}$ (3)

Where, 0.7 SvGy^{-1} is the conversion coefficient which transforms the absorbed dose rate in the air to an effective dose, and 10^{-6} is the factor converting nano into milli.

Annual effective dose due to ingestion of food cultivated in the area

The Annual effective dose due to ingestion (D_{ing}) was determined with Equation 4 (UNSCEAR 2000);

 $D (Svvr-1) = A \times CR \times DF$ (4)

Where, A ($Bqkg^{-1}$) is the activity concentration of radionuclide, CR is the consumption rate per year (kgyr-¹), and DF $(SvBq^{-1})$ is the standard dose conversion factor which is equal to $0.28 \mu Sv$ Bq⁻¹ for ²³⁸U, 0.23 μ SvBq⁻¹ for ²³²Th and 0.0062 μ SvBq⁻¹ for ⁴⁰K for the persons who live over 17 years. The food consumption rates for maize, vegetables, fruits, and yams obtained from Food Balance Sheet Nigeria, 2014 were 31.10, 46.70, 59.50, and 100.40 kgyr⁻¹ respectively.

Excess lifetime cancer risk (ELCR)

The excess lifetime cancer risk (ELCR) was calculated using Equation 5:

 $ELCR = AEDE \times DL \times R$ (5) Where, AEDE, is the annual effective dose due to ingestion, DL is the duration of life (70 years) (WHO, 2018) and risk factor R is 0.05 Sv^{-1} which is fatal cancer risk per sievert.

Radium Equivalent Activity (Raeq)

It is mathematically calculated by Equation (6) to address the levels of ²³⁸U, ²³²Th, and ⁴⁰K thus taking into account the radiological risks associated with them (Agbalagba and Onoja, 2011);

 $Raeq = Au + (1.43 A_{TH}) + (0.077 A_k)$ (6)

Where, A_U , A_{Th} and A_K are the activity concentration of ²³⁸U, ²³²Th, and ⁴⁰K respectively.

Radiation Hazard Indices

These indices are used to estimate the level of gamma radiation hazard associated with the natural radionuclide in samples. The external radiation hazard (H_{ext}) and the internal radiation hazard (H_{int}) was calculated using Equation 7a and b respectively:

$$
H_{ext} = (A_U/370) + (A_{Th}/259) + (A_K/4810)
$$
 (7a)
\n
$$
H_{int} = (A_U/185) + (A_{Th}/259) + (A_K/4810)
$$
 (7b)

Where, A_U , A_{Th} and A_K are the radioactivity concentration in Bqkg⁻¹ of ²³⁸U, ²³²Th, and ⁴⁰K respectively. Hin should be less than unity for the radiation hazard to be negligible. Internal exposure to radon is very hazardous which can lead to respiratory diseases like asthma (Tufail *et al.,* 2007; Orosun *et al.,* 2018).

RESULTS AND DISCUSSION

The activity concentrations of the radionuclides in Tables 1, 2, and 3 showed that the values of soil samples for ⁴⁰K, fall in the range of 269.53 ± 2.07 to $2559.87 \pm$ 19.7 Bqkg-1 , having a mean value of 1136.74±27.07 Bqkg⁻¹, The concentrations of ²³⁸U ranged from 8.96 \pm 0.44 to 85.02 ± 19.38 Bqkg⁻¹ while the mean value was recorded to be 41.66 ± 0.04 Bqkg⁻¹. The activity concentrations of ²³²Th ranged from 8.25 \pm 0.04 to 82.26 ± 1.56 Bakg⁻¹ having a mean value of 36.44 ± 0.04 Bqkg⁻¹, with ¹³⁷Cs ranging from 3.21 \pm 0.01 – 6.48 \pm 0.03 Bqkg-1 . The activity concentrations of cassava samples for 40 K, were found to range from 239.58 \pm 4.85 to 2275.44 \pm 14.96 Bqkg⁻¹ having a mean value of 1118.53 \pm 4.85 Bqkg⁻¹. The concentrations of ²³⁸U ranged from 5.26 \pm 0.38 to 52.4 \pm 4.69 Bqkg⁻¹ recording a mean value of 22.99 ± 0.38 Bqkg⁻¹. The activity concentrations of ²³²Th were observed to lie in the range of 4.79 \pm 0.07 to 36.51 \pm 1.44 Bqkg⁻¹ having a mean value of 15.89 ± 0.07 Bqkg⁻¹ with ¹³⁷Cs ranging from 3.07 ± 0.01 to 7.66 ± 0.03 Bqkg⁻¹. The activity concentrations in bitter leaf samples values for 40 K in Enugu were between 44.33 ± 0.02 to 759.51 ± 6.89 Bqkg⁻¹ with a mean value of 196.10 ± 0.02 Bqkg⁻¹. The activity concentrations of ²³⁸U ranged from 10.17 ± 0.06 to 19.99 \pm 1.18 Bqkg⁻¹ while the mean value was recorded to be 13.78±0.06 Bqkg-1 . The concentrations of ²³²Th ranged from $5.01 \pm 0.02 - 13.09 \pm 0.07$ Bqkg⁻¹ with a mean value of 7.69 ± 0.02 Bqkg⁻¹ and ¹³⁷Cs ranged from $3.85 \pm 0.01 - 7.81 \pm 0.03$ Bqkg^{-1.40}K displayed the highest activity. This is expected because $40K$ is a naturally occurring radionuclide that abounds in the earth's crust. The mean activity concentration of k-40 and U-238 obtained for the soil samples of the study area is higher than the recommended value (UNSCEAR, 2000) and in agreement with the global trend on the distribution of natural radionuclides in the soil as reported in literature. The detection of Cs-137 in the study area also conformed to the findings of Akinloye et al. (2012). The concentration of K-40 recorded in this study is very high while the U-238 and Th-232 low when compared to others in literature (Table 2b).This high value recorded for the K-40 can be attributed to the use of NPK fertilizers on the farmland from which the cassava samples were collected.

Table 2a: Activity Concentrations of Radionuclides in Cassava Samples in Enugu

Table 2b: Comparison of radionuclides concentrations (Bqkg−1) of ⁴⁰K, ²²⁶Ra, ²³²Th in Cassava

Table 3: Activity Concentrations of Radionuclides in Bitter leaf Samples in Enugu

Radiological Hazard Indices for soil samples

Table 4 presents the radiological parameters for the soil samples. The absorbed dose rate was recorded and observed to fall between 31.23 and 172.18 $nGyh^{-1}$ with a mean value of 88.54 $nGyh^{-1}$. The annual effective dose equivalent recorded an interval of 0.038 and 0.211

mSvy−1 with a mean value of 0.11 mSvy-1 . It is the tissue-weighted sum of the equivalent doses in all specified tissues and organs of the body and represents the stochastic health risks to the whole body. It takes into account the type of radiation and the nature of each organ or tissue being irradiated, and enables summation of organ doses due to varying levels and types of radiation, both internal and external, to produce an overall calculated effective dose. This combines both internal and external exposures (Orosun, *et al*., 2016). The calculated radium equivalent activities of soil samples ranged between 65.99 to 343.99 Bqkg⁻¹ with a mean value of 176.69 Bqkg⁻¹. The values of radium equivalent indices for all examined soil samples have a value lower than 370 Bqkg⁻¹ recommended by UNSCEAR (2000). Similarly, table 4 shows the external

and internal hazard indices which were calculated and estimated to be 0.178 to 0.929 Bqkg-1 and 0.211 to 1.09 Bqkg⁻¹ with mean values of 0.49 and 0.59 Bqkg⁻¹ respectively. The Excess Lifetime cancer risk (ELCR) ranged between 0.133 to 0.738 $(x10^{-3})$ with mean values of 0.38 $(x 10⁻³)$ which implies that there is a possibility of persons in the investigated area having cancer over a lifetime exposure to low-level background radiation dose as the values are higher than the world permissible limit of 0.29 x 10⁻³.

Radiological Hazard Indices for food samples

Tables 5 and 6 presents the radiological parameters calculated for Cassava (*Manihot esculenta*) and Bitter leaf (*Vernonia amygdalina*), which are known to be the most common staple foods in the South Eastern part of Nigeria. The values obtained for the annual effective dose equivalent were observed to be higher than the UNSCEAR permissible limit of 1.1 mSvy⁻¹, ranging between 2.98 to 24.73 and 0.82 to 10.87 $mSvy^{-1}$ with mean values of 12.03 and 4.02 mSvy⁻¹ for cassava and

bitter leaf respectively. Also, the calculated value for the excess lifetime cancer risk (ELCR) varied between 1.03×10^{-3} and 5.69×10^{-3} with a mean value of 2.93×10^{-3} and 1.03×10^{-3} and 5.7×10^{-3} with a mean value of 2.86×10^{-3} When compared with the recommended safe limit of 2.9×10-3 by UNSCEAR, it could be inferred that the ELCR values may determine the probability of being casino genic and the likes, such as skin cancer and cataracts.

Table 6: Estimated Radiological Parameters for Bitter Leaf Samples in Enugu

CONCLUSION

The radionuclide contents of soil and food samples have been analyzed utilizing gamma spectrometry. The results obtained shown high values of ⁴⁰K making it the dominant radionuclide in the study areas. The results also indicated the presence of $137Cs$ in two or three villages which could be due to the aged post-war activities in the study area, though with low concentration. The calculated radium equivalent, External hazard index (Hex), and internal hazard index (H_{in}) were found below the international reference limits of 370 Bqkg-1 and 1.00 Bqkg-1 respectively, thereby showing that the use of soil for building purposes may not pose any radiological hazard risks to the dwellers. The annual effective dose equivalent (AEDE), and excess lifetime cancer risk for the food samples were all calculated and it suffices to say that the food samples may not be radiologically safe for consumption since they are above the world permissible values. It is, therefore recommended that appropriate measures should be taken to protect the populace from adverse health implications. It is also recommended that such study be routinely carried out in the study area.

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