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DETERMINATION OF RADIOACTIVITY CONCENTRATION OF NATURAL RADIONUCLIDES AND RADIATION HAZARD INDICES IN SOIL SAMPLES FROM OWERRI, SOUTH EAST NIGERIA.

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ABSTRACT

The determination of radioactivity concentration of natural radionuclides and radiation hazard indices in soil samples from three locations in Owerri, Nigeria was carried out. The levels of naturally occurring radioactivity in the soil samples were evaluated using Gamma Ray Spectrometry. The mean values obtained were 145.2 ± 3.6 ($Bq.Kg^{-1}$), 12.5 ± 1.6($Bq.Kg^{-1}$) and 14.9 ± 0.6($Bq.Kg^{-1}$) for ^{40}K, ^{238}U and ^{232}Th respectively. The Absorbed Dose Rate, External Hazard Index and Internal Hazard Index were evaluated using the activity concentrations of ^{40}K, ^{238}U and ^{232}Th. The results showed that the mean value of radium equivalent activity is 45.0 $Bq.Kg^{-1}$, while the values of absorbed dose rate and annual effective dose equivalent are 21.4nGy h^{-1} and 0.026mSvy^{-1} respectively. *The values of external and internal health hazard indices are 0.12 and 0.15 respectively.* The obtained values were less than the recommended safety limits of 51nGy.h^{-1} and 1Bq.kg^{-1}. The study shows that the risk due to radiation contamination in the study area is low.

KEYWORDS: Activity concentration, Dose, Uranium. Natural radioactivity, Radiological hazards.

INTRODUCTION

Radioactivity is the spontaneous disintegration of unstable nuclei by emitting nuclear particles and energy in the process. The types radiation emitted in the process of radioactivity *are called alpha (α), beta (β) and gamma (γ) radiation.* Natural radioactivity is a source of

continuous exposure to human beings. It is present in the human environment due to the presence of cosmogenic and primordial radionuclides such as ^{40}K, and the radionuclides from the ^{238}U and ^{232}Th and their decay products, which are still present in the Earth's crust, (European Commission, 2015). Cosmogenic radionuclides are produced by the interaction of cosmic- rays with atomic nuclei in the atmosphere, while Primordial radionuclides, appeared on the Earth at the time of formation of the Earth (Radenkovic, M.B. et. al.2009).

Natural radioactivity is common in rocks, soil, sediments and ground and surface waters, and even in building materials and houses with varying degree of concentrations depending on the geographical conditions and geologic formations (Ravisankar et al., 2015; Huang et al., 2015). About 87% of the radiation doses received by humans are from natural radiation sources, which come from the naturally occurring radioactive isotopes of 238U and 232Th and their progeny as well as 40K (Shetty & Narayana, 2010; UNSCEAR, 1993). Other radionuclides of concern are those from the decay of ^{226}Ra and ^{228}Ra. Estimated exposure to natural radiation from naturally occurring radionuclides has become environmental concern to the public and national authorities of many countries because of its deleterious effects on human health. Therefore tremendous efforts are being made to locate and control the sources of natural radiation where economical interest exists and on which legislation must be applied.

Previous studies involving assessment of radioactivity in the environment have been conducted.

Nwankwo, L. I et al.(2016). Assessed the natural radioactivity in soil samples within the basement complex terrain of Tanke district of Ilorin in west central part of Nigeria. The results revealed the presence of potassium, uranium and thorium radio-elements. The activity of the radionuclides range from 252.03 to 494.02 Bq Kg-1 for 40K with a mean of 379.30 ± 31.15 Bq Kg^{-1}, 2.08 to 12.87 Bq Kg-1 for ^{238}U having a mean of 6.81 ± 2.03 Bq Kg^{-1} and ^{232}Th activity ranges from 6.55 to 11.85 Bq Kg^{-1} with a mean of 9.05 ± 2.95 Bq Kg^{-1}. The derived absorbed dose rate ranges from 20.01 to 30.03 nGy h-1 with a mean of 24.89 nGy h^{-1}. Consequently, the annual effective dose received by the population is estimated to range from 0.025 to 0.037 mSv y^{-1} with an average of 0.031 mSv y^{-1}. The radio-activities in soil samples of the area are found to be within global range for average terrestrial ionizing radiation exposure due to radionuclides in the soil. Ajayi, S. O,et al,(2013). Examined the radioactivity of surface soils from Oyo state. The specific acivity concentrations of the radionuclides

ranged from 1±0.4 Bq kg^{-1} for 137Cs to 1190±30 Bq kg^{-1} for ^{40}K. The es□mated outdoor absorbed dose rates in air varied from 52 nGy h^{-1} in Egbeda (a rural area) to 414 nGy h^{-1} in Eruwa (also a rural area). The mean annual outdoor effective dose equivalent for the urban areas in the state was 0.1 mSv y^{-1} while that of rural areas was 0.3 mSv y^{-1} with a standard deviation of 0.02 mSv y-1 and 0.3 mSv y-1 respectively. The mean for the study area was 0.2 mSv y-1.

Nwaka, B.U.et al,(2014) . Investigated t*he natural radiation of soil samples in* Owerri, Imo state, Nigeria using NaI(Tl) gamma ray spectrometer. The mean values obtained were 167.2 ± 10.5 ()**,** 19.7 ± 1.9 () and 18.1 ± 3.3 () for 40K, 226Ra and 232Th respectively. These values were used to evaluate the radiological health hazard indices using standard analytical methods. The results showed that the mean value of radium equivalent activity is 58.5, while the values of absorbed dose rate(D) and annual effective dose equivalent (E) are 27.nGyh^{-1} and 132.78mSvy^{-1} respectively. The values of external and internal health hazard indices are 0.16 and 0.21 respectively.

M.O. Isinkaye, M.O. (2015). Determined the distributions of naturally occurring radionuclides 226Ra, 232Th and 40K in sediments of Oguta Lake, Nigeria were determined using gamma ray spectrometry in order to assess the radiological health hazards and excess lifetime cancer risks associated with the use of the sediments. The mean activity concentrations of 226Ra, 232Th and 40K were found to be 47.89 ± 18.67 Bq kg_1, 55.37 ± 32.74 Bq kg_1 and 1023 ± 474 Bq kg_1, respectively. The results of the radiological indices and dose rates obtained in this study were all higher than their worldwide mean values but lower than their maximum recommended limits indicating that the use of the sediments as building materials do not constitute any excessive radiological hazards.

In this paper the results of the measurements of ^{40}K, ^{238}U and ^{232}Th concentrations in soil samples collected from Owerri are presented. The calculations of the absorbed dose, annual effective dose and external hazard index are also presented and discussed.

MATERIALS AND METHODS

Study area

The study area is the urban area of Owerri, Imo State, South East Nigeria and its environs. Owerri is the capital city of Imo State, South East Nigeria. Owerri with a population of about

150,000 situates between 5^0 20'N, 6^0 55'E in the south-western corner and 5^0 34'N, 7^0 08'E in the north-eastern corner.

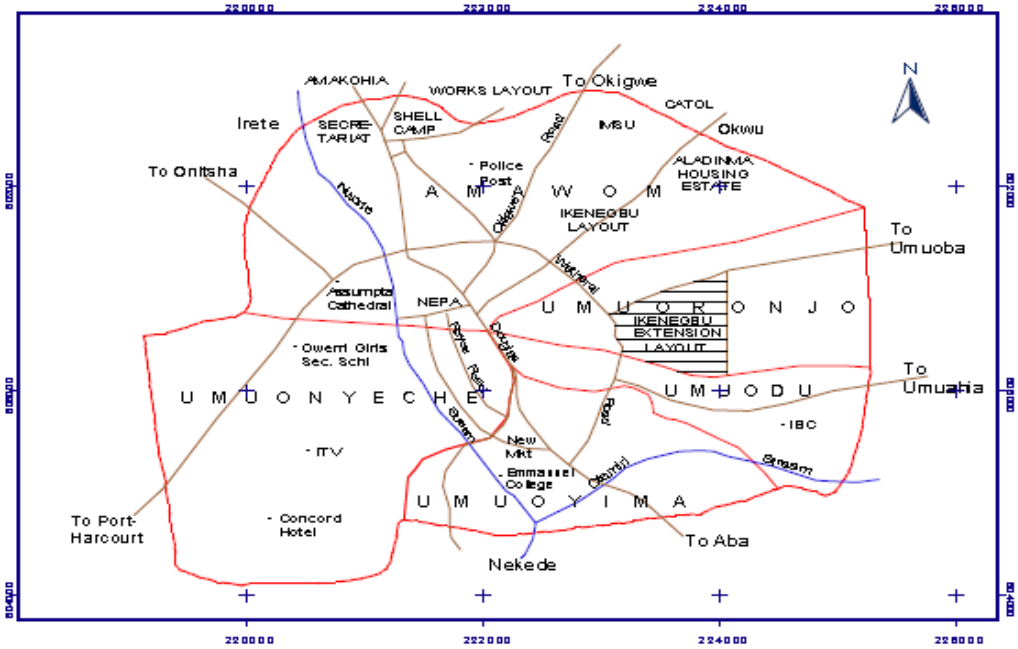

Figure 1. Map of Owerri Urban

Sample collection and preparation

The soil samples were collected at a depth of about 50 cm at the various locations. The names of the sampling stations are given in Table 1. The samples were processed following the standard procedures. Soil samples were well mixed after removing extraneous materials such as roots, pieces of stones, gravel and plant materials such as litter, roots, etc. Samples were weighed and then dried in an oven at 105°C overnight in order to remove any available moisture. After drying, the samples were crushed and sieved with a mesh having holes each of diameter of 0.2 mm. Sieved samples were weighed and a mass of 200 g of each sample was placed in a plastic container. The containers were sealed for thirty (30) days to allow for ^{238}U and its short-lived progenies to reach secular radioactive equilibrium (Veiga et al, 2006). The counting time for the samples was 36,000 seconds. Each sample was counted for 36,000 seconds to reduce the statistical uncertainty. The gamma ray spectrometer used in this analysis consists of a 7.6 cm by7.6 cm NaI (Tl) detector enclosed in a 5 cm thick lead shield to reduce background radiation. The setup was coupled to a Multichannel Analyzer (MCA) employed for obtaining the data and analysis of gamma spectra. The gamma spectrometry detector was calibrated before it was used for analysis. The spectrum was measured and the

area under the photopeaks computed using the algorithm of the Multichannel Analyzer (MCA). The transition lines of 1.460 MeV of 40K, 1764.5 KeV of 214Bi and 2614.7KeV of 208Ti were used to determine the concentrations of ^{40}K, ^{238}U and ^{232}Th respectively.

Table 1. Location of the soil samples in the study area.

S/N	Location of Samples	Sample code
1	Port Harcourt Road	X_1
2	Amakohia	X_2
3	Nekede	X_3

Calculations

The Activity Concentration

The specific activity concentration A_C (i.e. $A_C(K)$,$A_C(U)$ and $A_C(Th)$ for ^{40}K, ^{238}U and ^{232}Th respectively) in soil samples were computed using the relation (Baykara et al 2009).

$$A_C = \frac{C}{\varepsilon P_\gamma M_s} \qquad (1)$$

Where A_C is the activity concentration of the radionuclide in the sample given in Bq kg^{-1}, C is the count rate under the corresponding peak, ε is the detector efficiency at the specific γ-ray energy, P_γ is the absolute transition probability of the specific γ-ray and *Ms* is the mass of the sample (*kg).*

The Radium Equivalent Activity (Ra_{eq})

The radium equivalent activity, Ra_{eq} (Mahur et al., 2008) was calculated according to Eq. (2). The radium equivalent concept allows a single index or number to describe the gamma output from different mixtures of ^{238}U, ^{232}Th and ^{40}K in sediments samples from different locations.

$$Ra_{eq} = A_U + 1.429\, A_{Th} + 0.077\, A_K \qquad (2)$$

Where A_U, A_{Th} and A_K are the specific activity concentrations of ^{238}U, ^{232}Th and ^{40}K (Bq kg^{-1}), respectively.

Absorbed gamma dose rate (D $nGyh^{-1}$)

This is the amount of radiation energy absorbed or deposited per unit mass of substance. The absorbed gamma dose rates due to gamma radiations in air at about 1meter above the ground surface for the uniform distribution of the naturally occurring radionuclides (^{238}U, ^{232}Th and ^{40}K) were calculated based on the guidelines provided by UNSCEAR, 2000. The

conversion factors used to compute the absorbed gamma dose rate in air per unit activity concentration in Bq kg^{-1} corresponds to 0.429 nGy h^{-1} for ^{238}U, 0.666 nGy h^{-1} for ^{232}Th and 0.042 nGy h^{-1} for ^{40}K. Therefore D can be calculated as follows:

$$D(nGy\ h^{-1}) = 0.429A_U + 0.666\ A_{Th} + 0.042A_K \quad (3)$$

where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in $Bqkg^{-1}$, respectively.

2.7. Annual effective dose rate (AEDR)

The annual effective dose rate (AEDR) in $mSvy^{-1}$ resulting from the absorbed dose values (*D*) was calculated using the following formula (UNSCEAR, 2000; Ravisankar et al. 2015).

Ann. Eff. dose rate($mSvy^{-1}$) = $D(nGyh^{-1})$ ×8760h y^{-1} × 0.7 × 10^3 mSv

10^9 nGy × 0.2

$$AEDR = D \times 1.23 \times 10^{-3} \quad (4)$$

External and internal hazard indices (H_{ex}) and (H_{in})

Beretka et al.,(1985), defined two other indices that represent external and internal radiation hazards. The external and internal hazard index is obtained from Ra_{eq} expression through the supposition that its allowed maximum value (equal to unity) corresponds to the upper limit of Ra_{eq}(370 Bq kg^{-1}). The external hazard index(H_{ex}) and internal hazard index (H_{in}) can then be defined as:

$$H_{ex} = \left(\frac{Au}{370}\right) + \left(\frac{ATh}{259}\right) + \left(\frac{Ak}{4810}\right) \quad (5)$$

$$H_{in} = \left(\frac{Au}{185}\right) + \left(\frac{ATh}{259}\right) + \left(\frac{Ak}{4810}\right) \quad (6)$$

where A_U, A_{Th} and A_K are the activity concentrations of ^{238}U, ^{232}Th and ^{40}K, respectively. This index value must be less than unity in order to keep the radiation hazard to be insignificant.

Table 2. The activity concentrations of radionuclides in soil samples collected from the study area.

S/N	Sample code	^{40}K ($Bq{\cdot}kg^{-1}$)	^{238}U($Bq{\cdot}kg^{-1}$)	^{232}Th($Bq{\cdot}kg^{-1}$)
1	X_1	143.1± 3.0	11.6± 1.3	14.7± 0.5
2	X_2	141.2± 1.4	12.5± 1.6	16.4± 1.0
3	X_3	151.3± 6.3	13.4± 2.0	13.6± 0.4
Mean		145.2± 3.6	12.5± 1.6	14.9± 0.6

Table 3. The radium equivalent (Ra_{eq}), the absorbed dose rate (D), the annual effective dose rate (AEDR), external (H_{ex}) and internal (H_{in}) hazard index of the soil samples collected from the study area.

S/N	Sample code	Ra_{eq} ($Bq \cdot kg^{-1}$)	*D* ($nGy \cdot h^{-1}$)	(AEDR) ($mSv\ y^{-1}$)	H_{ex}	H_{in}
1	X_1	43.63	20.78	0.025	0.12	0.14
2	X_2	46.81	22.22	0.027	0.13	0.16
3	X_3	44.48	21.16	0.026	0.12	0.15
Mean		45.00	21.40	0.026	0.12	0.15

Table 4. Comparison of the results of the present study with similar published data.

Region	$^{238}U(Bq \cdot kg^{-1})$	$^{232}Th(Bq \cdot kg^{-1})$	$^{40}K(Bq \cdot kg^{-1})$	Reference
Egypt	5 – 64 (17)	2 – 96 (18)	29 – 650 (320)	UNSCEAR (2000)
USA	8 – 160 (40)	4 – 130 (35)	100 – 700 (370)	UNSCEAR (2000)
Greece	1 – 240 (25)	1 – 190 (21)	12 – 1570 (360)	UNSCEAR (2000)
India	7 – 81 (29)	14 – 160 (64)	38 – 760 (400)	"
Japan	6 – 98 (33)	2 – 88 (28)	15 – 990 (310)	"
Spain	6 – 250 (32)	2 – 210 (33)	25 – 1650 (470)	"
Nigeria	9 – 18 (14)	1 – 38 (19)	712 – 1098 (896)	Okeyode et al (2010)
Saudi Arabia	11 – 30 (15)	7 – 25 (11)	98 – 320 (225)	Alaamer (2008)
Nigeria	11 – 13 (12)	13 – 16 (14)	143 – 151 (145)	Present Study
World Average	17 -60 (35)	11 – 64 (30)	140 -850 (400)	UNSCEAR (2000)

RESULTS AND DISCUSSION

The results of analysis of activity concentration of ^{238}U, ^{232}Th, ^{40}K radionuclides in soil samples for different locations of the study area are presented in Table 2, while the calculations of the radium equivalent (Ra_{eq}), the absorbed dose rate (D), the annual effective dose rate (AEDR), external (H_{ex}) and internal (H_{in}) hazard index of the soil samples collected from the study area are presented in Table 3. The activity of ^{238}U range from 11.6 to 13.4Bq kg^{-1} with a mean of 12.5Bq kg^{-1}; ^{232}Th activity from 13.6 to 16.4 Bq kg^{-1} with a mean of 14.9 Bq kg^{-1} and ^{40}K range between 141.2 to 151.3Bq kg^{-1} with a mean of 145.2Bq kg^{-1} for the soil samples. The radium equivalent activity (Ra_{eq}) calculated for the soil samples vary from 43.63$Bq \cdot kg^{-1}$ to 46.81$Bq \cdot kg^{-1}$ with a mean value of 45.00 $Bq \cdot kg^{-1}$. It is inferred that for all the soil samples analyzed, the radium equivalent activity value is well within and less the permissible limits of 370 $Bq \cdot kg^{-1}$. The calculated absorbed dose rate varied from 20.78 to 22.22 $nGy \cdot h^{-1}$, with a mean value of 21.40 $nGy \cdot h^{-1}$. The mean value of 21.40 nGy·h–1 represents 39% of the world average outdoor exposure due to terrestrial gamma radiation (55 $nGy \cdot h^{-1}$, according to UNSCEAR, 2000). The annual effective dose rates calculated vary from 0.025 to 0.027 mSv y^{-1} with a mean value of 0.026 mSv y^{-1}. The results shows that the soil samples X_1(Port Harcourt Road) has the lowest dose rate of 20.78 nGy h^{-1} (0.025 mSv y^{-}

^{1}) while sample X_2 (Amakohia) has the highest dose rates of 22.22 nGy h^{-1} (0.027 mSv y^{-1}). The mean annual effective dose calculated for the area are within the permissive dose limit of 1 mSv y^{-1}.

The external hazard index (H_{ex}) calculated in the present study ranges from 0.12 to 0.13 with a mean of 0.12. The mean value of the external hazard index is less than the recommended value. . The calculated internal hazard index (H_{in}) of the sediment samples varies from 0.14 to 0.16 with a mean of 0.15.

The recommended value of internal radiation hazard index is less than 1. Therefore, these areas may not pose radiological risks to the inhabitants owing to harmful effects of ionizing radiation from the natural radionuclides in sediments.

Comparison of the results with other countries.

The concentration of ^{238}U, ^{232}Th and ^{40}K observed in the present study with those reported for the normal background regions of other countries are given in Table 4. From the table, it shows that the concentrations observed in soils of present study are comparable to those reported for worldwide range and average values (UNSCEAR, 2000) and also to the values reported for European, Asian, South and North American countries (Okeyode et al,2010 and Alaamer, 2008). The values obtained in this work fall within the ranges but are lower than the global average values.

CONCLUSION

The radioactivity concentrations of ^{238}U, ^{40}K and ^{232}Th and the associated radiation hazard levels in soil samples in the 3 different locations of the study area were investigated using gamma ray spectrometer. The mean values obtained were 12.5 ± 1.6(Bq.Kg^{-1}), 145.2 ± 3.6 (Bq.Kg^{-1}) and 14.9 ± 0.6(Bq.Kg^{-1}) for ^{238}U,^{40}K and ^{232}Th respectively. The results of the mean activity concentrations are lower than the world average values. It is concluded that no harmful radiation effects were posed to the population who live in the study area. The radium equivalent (Ra_{eq}), the absorbed dose rate (D), the annual effective dose rate (AEDR), external (H_{ex}) and internal (H_{in}) hazard index values obtained from the study area were lower than the average national and world recommended safety limits. Therefore, it is an indication that the study area is safe for human activity.

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