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Assessment of Natural Occurring Radioactive Materials and Radiological Hazards Exposure in Soil of Ohia in Umuahia South Abia State Nigeria, Using High Purity Germanium (HPGe) Gamma Ray Spectrometry

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ABSTRACT
Evaluation of natural occurring radioactive materials (NORMs) and radiological exposure are important. The main goal of this work was to determine the NORMs level and radiological exposure risk in Umuahia, using High Purity Germanium Gamma Ray Spectrometry at IPEN, Brazil. The radionuclides (RDNs) such as: ^{40}K, ^{226}Ra and ^{232}Th and radiological parametric indices (RPIs) such as: Absorbed Dose Rate in Air (D), Outdoor and Indoor Annual Effective Dose Equivalent (AEDE), Radium Equivalent Activity (Ra_{eq}), Annual Gonadal Equivalent Dose (AGED) and Activity Concentration Index (I) were determined. The mean values of ^{40}K, ^{226}Ra and ^{232}Th in Bq kg^{-1} were: 147, 28, and 31 while the RPIs mean values for D, $AEDE_{out}$, $AEDE_{in}$, AGED, Ra_{eq}, and I **are:** 37 nGy h^{-1}, 0.05 mSv y^{-1}, 0.18 mSv y^{-1}, 260 μSv y^{-1}, 83 Bq kg^{-1} and 0.29 respectively. Mean value of RDNs were found to be lower than 412 Bq kg^{-1}, 32 Bq kg^{-1} and 35 Bq kg^{-1} average value while RPIs were also lower than 59 nGy h^{-1}, 0.08 mSv y^{-1} for $AEDE_{out}$, 0.42 mSv y^{-1}for $AEDE_{in}$, 300 μSv y^{-1} for AGED, 370 Bq kg^{-1} for Ra_{eq}, and 1 mSv y^{-1} respectively global safe limit or mean values. It is concluded that the soil is radiological safe for agriculture, building and construction purposes.

Keywords:
Soil,
Natural Occurring Radioactive Materials (NORMs),
Radiological Hazards Exposure,
High Purity Germanium (HPGe) Detector,
Umuahia South,
Abia State, Nigeria.

INTRODUCTION

Natural Occurring Radioactive Materials (NORMs) are radioactive materials that occurs during the creation of earth (Ibrahim *et al.*, 2021) due to the presence of one or more naturally existent radionuclides. Examples are uranium, thorium; and their radioactive decay outcomes such as radium, radon, lead, polonium, and potassium (EPA, 2021). These radionuclides have been present about 4.5 billion years, since the planted formation, and are spread in soil, rocks, building materials, and sediments from where they can move into the air and water (Lilley, 2013; Ibrahim *et al.*, 2021).

Radiological hazards exposure (RHE) are those variable radiological parameters that are used to determine the approximate exposure risk-index as a result of radionuclides and radiation that may be existing in soil and other material samples during nuclear, atomic and radiological analysis. Examples area: absorbed dose rate in air (D), annual effective dose equivalent (AEDE), activity concentration index (I), yearly gonadal equivalent dose (AGED) and radium equivalent activity (Ra_{eq}) (Onudibia, 2023).

Knowledge of the spreading of radionuclide in the surroundings is advisable for radiation measurement and protection (Adewoyin *et al.*, 2018). The terrestrial radionuclides are unequally distributed in earth and depends on the geological nature, geochemical composition, mineralogy, and organic concentration of the soil (Eke *et al.*, 2024). They are the original generative force of ionizing radiation exposure of human body (Beogo *et al.*, 2022), being responsible for external and inside exposure.

Outside exposure is observed by the reactions of gamma, alpha or beta radiation emitted by the radionuclides around the human system while that of internal exposure takes place when the source of radionuclides is inhaled or ingested and decays occurs inside the living body (Beogo *et al.*, 2022). This natural radiation constitutes the main summation composition of the of annual exposure to the human populace (Addo *et al.*, 2020; Eke *et al.*, 2024**)**.

The awareness of radionuclide/ NORMs spreading within soils is important in order to regulate health jeopardy that may probably affect the population (Alzubaidi *et al*., 2016) since continuous exposure of radiation can cause dangerous radiological effects (UNSCEAR, 2000; Botwe *et al.,* 2017; Adewoyin *et al.,* 2018; Zubair, 2020; Eke *et al.*, 2024). On the other hand, it is believed that low portion radiation may present the hormesis effect and observed in different organic systems such as immunological and hematopoietic systems (UNSCEAR, 2016; Mbonu and Ubong, 2021).

This study was conducted, in order to examine the activity concentrations of natural radionuclides including its radiological exposure of soils from the Umuokwom Ohia Umuahia South, Abia State Nigeria. Main study area is approximately 31/4 km from clay mining site. Soils generally exhibits relatively high radionuclides amount, it is important to investigate NORMs in the present research domain because of the agriculture and building activities, and moreover, it a dwelling area and proximity to place of worship. In order to arrive at this aim, the following steps were carried out: determination of the radionuclides activity concentration, RHE, contour map analysis and correlation between the RHE and radionuclides.

MATERIALS AND METHODS

Study area

The research was carried out in Umuahia South, South East, Nigeria. Precisely at Umuokwom Ohia. Umuahia South is one of the local governments cited Umuahia Abia State. It has a Longitude of 5° 31’ 42” and Latitude of 7° 27’ 21”. The research domain and samples points are depicted in Figure 1. The mean area and population are 140 km² and 138,570, respectively. Umuokwom Ohia is along Port Harcourt and Enugu Road, approximately 9 km from Umuahia main town and approximately 46 km to Aba, Enyimba metropolis of Abia state. Generally, Umuahia has the following weather and climatic features: average temperature of 26.4 °C, 2333 mm of precipitation yearly and vegetation of tropical rainforest including paramount natural vegetation which is common in Southern Nigeria (Onudibia *et al.*, 2023).

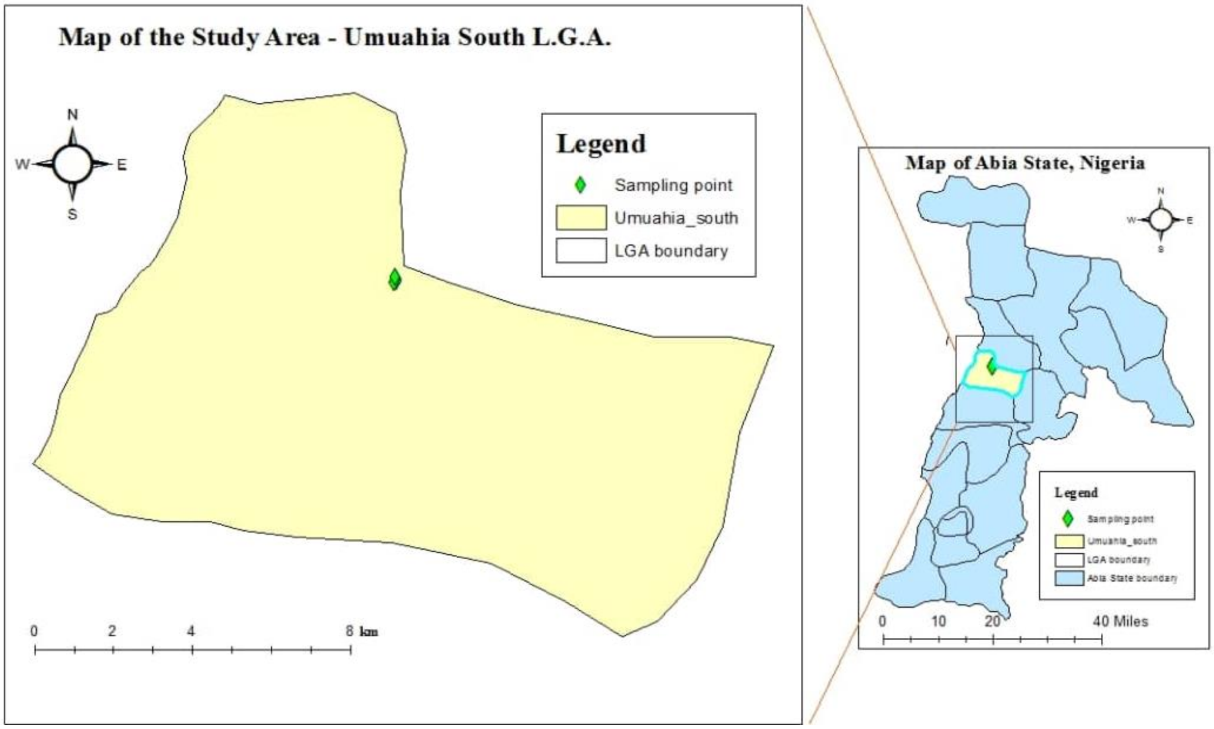

Figure 1: Map of Abia State showing Umuahia South L.G.A. Sampling

Five (5) soil samples were picked using a soil auger sampler at a depth of 5-15 cm. For each of the sampling point, about 1 kg to 1.5 kg soil samples were taken. The soil samples were transferred into identified plastic bags and properly tied (Onudibia, 2023; Onudibia *et al.*, 2023).

Samples Preparation

The samples were kept in natural air–dried room temperature for about two weeks. Furthermore, the samples were transported to the laboratory and all water samples were removed at a temperature of 105 °C within 3-4 hours till all the samples were dried to a steady weight, at the central lab of Federal University Wukari (FUW). Soil samples were grinded using mortar and pestle. A sieve of 2 mm pore size mesh was used to get constant particle sizes, as recommended by (IAEA, 1989; Onudibia *et al.*, 2023). The final homogeneous soil samples were fled to Nuclear and Energy Research Institute (IPEN), Sao Paulo Brazil for analysis.

Analysis of Samples/Analytical Technique

The radionuclides ^{40}K, ^{226}Ra, and ^{228}Ra were ascertained using p-type High Purity Germanium Detectors (HPGe), Canberra (USA) with relative efficiency 20%, and resolution 2.00 keV at 1.33MeV.

The activity concentration of the radionuclides was ascertained using the following energies: ^{40}K was ascertained using the 1460 keV gamma-ray peak, for ^{226}Ra, the 351 keV (214Pb) and 609 keV (214Bi) energies were used, and for ^{228}Ra, the 911 keV and 968 keV photopeaks from ^{228}Ac were used.

All samples were counted for 86,400 seconds, the background was counted for 172,800 seconds while the certified reference materials (CRM) RG-U, RG-Th and RG-K, from IAEA, were counted for 7,200 seconds. Activity concentrations were determined through punctual calibration comparing the intensity of peak samples with the respective peaks of the standard reference material.

Activity Concentration

The activity concentration of the radionuclides was calculated applying Gilmore and Hemingway (1995), the expression is given in Equation (1).

$$\text{Ac} = \frac{N_c e^{\lambda t_0}}{\varepsilon F_c I_\gamma M t_c} \quad (1)$$

where Ac is the Activity concentration of the radionuclide, N_c is number of counts at a given energy, subtracted from the BG, λ is the decay constant, t_c is the time of counting, t_0 is the time difference between sampling and start of count, M is the sample mass; is the efficiency, I_γ is the emission probability and F_c is the correction factor.

Radiological Parameters Indices (RPIs)

The radiological parameters are used for ascertaining the risk of radiation for the individuals based in the calculated activity concentrations of the natural occurring radionuclides.

Absorbed Dose Rate in Air (D)

It is used in measuring the exposure that enables ascertaining the quantity of radiation acquired by the individuals due to the concentrations of the radionuclides ^{226}Ra, ^{232}Th and ^{40}K in sample material. The absorbed dose rate D (nGyh^{-1}) in the air helps quantification of the quantity of radiation absorbed by a body at 1 m above the ground due to ^{226}Ra, ^{232}Th and ^{40}K (UNSCEAR, 2000; Onudibia, 2023; Eke *et al.*, 2024). The D was computed with Equation 2.

$$D = 0.0417K_{At} + 0.462\text{Ra}_{At} + 0.604Th_{At} \quad (2)$$

where D is the absorbed dose in nG$y\,h^{-1}$, 0.0417, 0.462, and 0.604 are the conversion constants and K_{At}, Ra_{At} and Th_{At} are the activity-concentrations of ^{40}K, ^{226}Ra and ^{232}Th all in Bqkg^{-1} respectively.

Annual Effective Dose Equivalent (AEDE)

The annual effective dose equivalent is used to determine the human being exposed of a certain dose rate to determine its radiological impact (Isinkaye *et al.*, 2018). The annual recommended value for the public by ICRP is 1 mSv (Araromi *et al.*, 2016). It was computed according to depicted in Equation 3.

$$\text{AEDE} = \text{D} \times OF_t \times \text{C}F_t \quad (3)$$

where D is the calculated dose rate in unit of nGy h^{-1}, OF_t is the occupancy factor, which is given as 0.2 if the individuals exhausted 20% of their duration outdoors and 0.8 if they exhausted 80% indoor (Taqi *et al.*, 2018; Adagunodo *et al*., 2018), while the conversion factor for absorbed dose in air to external effective dose in adults is given as 0.7 Sv/Gy and CF_t is the conversion factor (Adagunodo *et al*., 2018). Hence, the AEDE in mSv y^{-1} for outdoor was calculated using (Taqi *et al*., 2018; Nkuba and Nyanda, 2017) the expression given in Equation 4.

$$AEDE_{OUT} = \text{D} \times 0.7 \times 0.2 \times 8760 \times 10^{-6} \quad (4)$$

While the indoor AEDE was calculated using the Equation 5

$$AEDE_{IN} = \text{D} \times 0.7 \times 0.8 \times 8760 \times 10^{-6} \quad (5)$$

Radium Equivalent Activity (Ra_{eq})

Radium equivalent activity is one of the index that has long applied to give the specific activity of ^{40}K, ^{232}Th and ^{226}Ra by a sole amount which considers the hazard of radiation that relates them to the same dose (Araromi *et al.*, 2016). It is one of the commonly used hazard indices in radiation protection assessment for construction materials (Isinkaye *et al.*, 2018), given in Equation 6 where it is supposed that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produces the resemblance gamma dose rate (Adewoyin *et al.*, 2018)

$$\text{Ra}_{eq} = 0.077\text{K}_{At} + \text{Ra}_{At} + 1.43\text{Th}_{At} \quad (6)$$

where K_{At} is the meane activity concentration of ^{40}K (Bq kg^{-1}), Ra_{At} is the average activity concentration of ^{226}Ra, Th_{At} is the mean activity concentration of ^{232}Th.

Annual Gonadal Equivalent Dose (AGED)

Since, the gonads, bone surface cells and active bone marrow are the most vital organs of significant interest, it is imperative to take note of the annual gonadal equivalent dose for the human activity and the populace. It is computed in (μSvy^{-1}) from the specific activities of ^{226}Ra, ^{232}Th and ^{40}K using the expression as given Equation 7 (Ajekiigbe *et al.*, 2017; Onudibia, 2023; Onudibia *et al.*, 2023).

$$AGDE = 0.314K_{At} + 3.09Ra_{At} + 4.18Th_{At} \quad (7)$$

where K_{At}, Ra_{At} and Th_{At} are the activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th, respectively.

Activity Concentration Index (I)

The activity concentration index (I) is based on the fact that since a single radionuclide cannot put up to the dose, it is applicable to make findings of the concentration of the radionuclides to represent a single index that is stands to show whether the annual dose, due to the excess external gamma radiation in a building in more than 1 mSv y^{-1} (Rocznik *et al.*, 2023). It is calculated by equation 8.

$$I = \frac{K_{At}}{3000} + \frac{Ra_{At}}{300} + \frac{Th_{At}}{200} \quad (8)$$

where K_{At}, Ra_{At} and Th_{At} are the activity concentrations of ^{40}K, ^{226}Ra, and ^{232}Th, respectively.

RESULTS AND DISCUSSION

Activity Concentration of Natural Occurring Radionuclides (NORs)

Table 1 presents the obtained mean and individual values of natural occurring radionuclides (NORs) activity concentration for ^{40}K, ^{226}Ra and ^{232}Th in the study area for the five sampling points and the global average value according to UNSCEAR (2008). These NORs ranged from 27±2 to 206±15, 19±1 to 37±3 and 24±4 to 49±7 Bq kg^{-1}, with mean value 147±11, 28±2 and 31±5 Bq kg^{-1} see Figure 2. The mean activity concentration for ^{226}Ra and ^{232}Th in the soil samples, collected from the study area, are very close to the global mean value. Only ^{40}K presented values was less the half of the reported average, probably due to the whitening regime observed in the region (Ugbede, 2020).

Tables 2 shows the comparison of this work with literature reports, for the analyzed NORs. Reported values for ^{40}K in Nigeria varies from 31 to 195 Bq kg^{-1}, close to the values found in this study but below the ones seen in other nations (Tarbool *et al.*, 2022, Al-Ghamdi, 2019 and Alzubaidi, 2016). For ^{226}Ra and ^{232}Th, it was only the region of Owerri metropolis in Imo that observed values in the reported literature were lower than present work. Compared with other countries, the value found in this work are in the range of the observed values.

Table 1: Activity Concentration of NORs of Soil Umuokwom Ohia in Umuahia, (Bq Kg^{-1})

SID	^{40}K	^{226}Ra	^{232}Th
USB1	27±2	37±3	49±7
USB2	206±15	35±3	32±5
USB3	163±13	22±2	25±4
USB4	163±14	19±1	24±4
USB5	175±12	25±2	24±6
MAX	206±15	37±3	49±7
MIN	27±2	19±1	24±4
MEAN	147±11	28±2	31±5
(UNSCEAR, 2008)	412	32	45

Table 2: Comparison of Activity Concentration of Natural Occurring Radionuclides (NORs) of Soil of Present Study with other Peoples Work (Bqkg-1)

Country/Location	^{40}K	^{226}Ra	^{232}Th	Reference
Umuahia Nigeria	147±11	28±2	31±5	Present Work
Umuahia Nigeria	31±2	38±3	57±7	Onudibia *et al.*, 2023
Owerri, Imo Nigeria	88.4±1.5	20.7±3.6	25.0±0.7	Eke *et al.*, 2024
Ado-Odo/Ota Nigeria	134.3	-	94.4	Joel *et al.*, 2018
Ogun State Nigeria	195±20	-	135±8	Adewoyin *et al.*, 2018
Iraq	201.5±24.5		5.9±0.8	Tarbool *et al.*, 2022
Jeddah, Saudi Arabia	419.8	16.7	10.4	Al-Ghamdi, 2019
North of Malaysia	325.9 ± 9.8	102.1 ± 3.9	134.0 ± 3.0	Alzubaidi, 2016

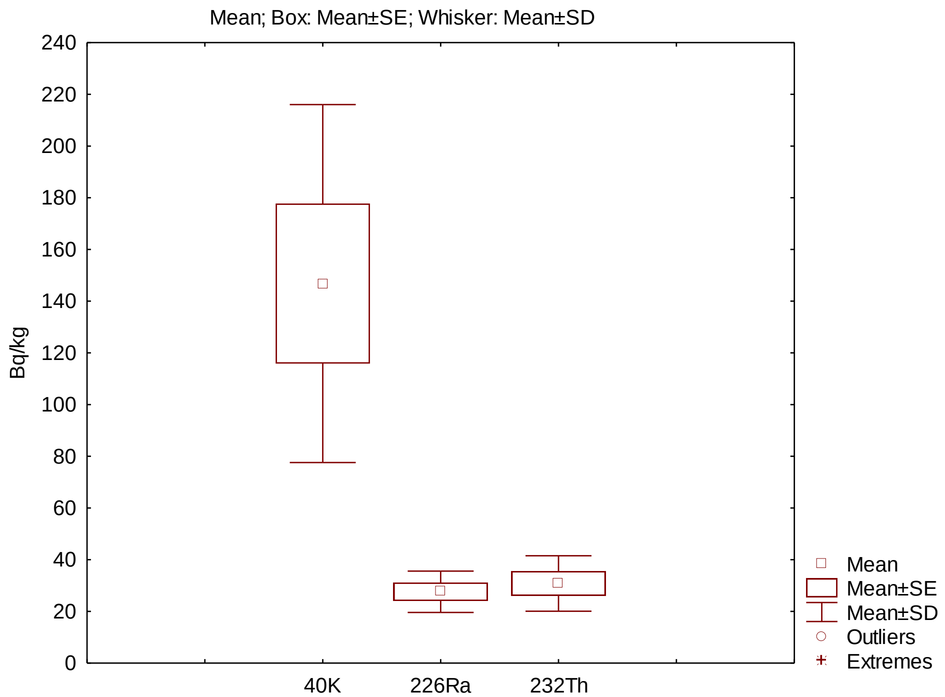

Figure 2: Bar Graph Mean Distribution Comparison of the Radionuclides ^{40}K, ^{226}Ra, and ^{232}Th with Present Study and UNSCEAR (2008)

Contour Map

Figures 3 to 5 represents the contour map of distribution for ^{40}K, ^{226}Ra and ^{232}Th concentrations in Umuahia South L.G.A. of Abia State. The iso-concentration lines show the regions of equal concentration of the radionuclides in the research domain. The gape existing in the lines defines the distance of each concentration level, and the color around every gape identifies the extent of concentration of the radionuclide at a particular area.

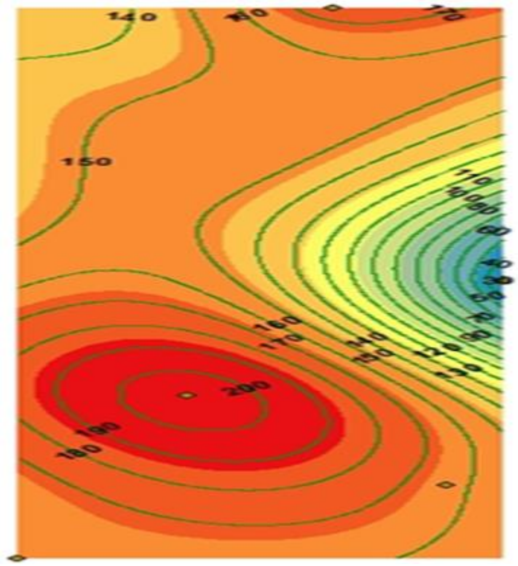

Figure 3: Contour Map Distribution of ^{40}K

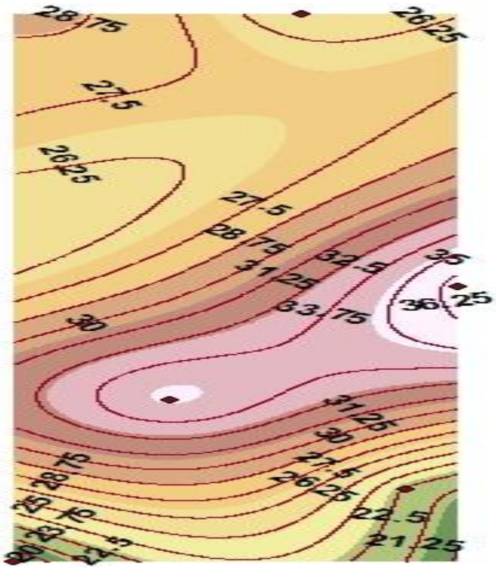

Figure 4: Contour Map Distribution of ^{226}Ra

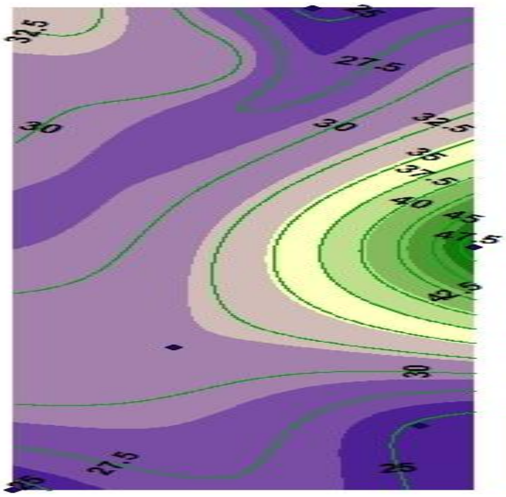

Figure 5: Contour Map Distribution of ^{232}Th

Figure 3 shows the distribution of ^{40}K in the study area. The region marked with light blue shows the region of low concentration of ^{40}K, while regions marked with dark red show the regions of higher concentration of ^{40}K. Similarly, Figure 4 shows the arrangement of ^{226}Ra in the study area, in which marked areas in white shows region of high concentration of ^{226}Ra while the area marked with light green shows the region of low concentration of ^{226}Ra. Also Figure 5 represents the distribution of ^{232}Th in the study area in which areas marked with green show the region of high concentration of ^{232}Th, while the area marked with purple shows the region of low concentration of ^{232}Th.

Comparing the three contour maps, it was observed that both ^{40}K and ^{226}Ra have high concentrations towards the southern part of the study area and both also show low concentration towards the eastern part of the study area. Meanwhile, ^{232}Th was concentrated more at the eastern part of the study area and less concentrated towards the southern part of the study area unlike the both ^{40}K and ^{226}Ra. Looking at the northern part of the study area, both ^{226}Ra and ^{232}Th were less concentrated in that region while ^{40}K is more concentrated.

Radiological Parameters Indices (RPI)

Table 3 presents the calculated RPIs D (nGyh^{-1}), $AEDE_{out}$ (mSvy^{-1}), $AEDE_{in}$(mSvy^{-1}), AGED (μSvy^{-1}), Ra$_{eq}$ (BqKg^{-1}) and I (mSvy^{-1}) of the study area and their range (maximum to minimum) and mean value is as shown in Figures 6 and 7. The compared mean value of RPIs of the present study with the global average or safe limit of 59 (nGyh^{-1}), 0.08 $AEDE_{out}$ (mSvy^{-1}), 0.42 $AEDE_{in}$ (mSvy^{-1}), 300 AGED (μSvy^{-1}), 370 Ra$_{eq}$ (BqKg^{-1}) and 1 (mSvy^{-1}) were lower than the reported by ICRP (1991) and UNSCEAR (2008) as shown in Figure 8. Table 4 shows the comparison of the RPIs with other studies. Comparing this study with related Nigerian and international studies, reports from North of Al-Najaf Governorates Iraq, AlHusseinea Iraq, and Saudi Arabia were lower than this present study while in the Umuahia south Nigeria, about 3km away from a clay mining site, Ado-Odo/Ota, Nigeria, Ogun State Nigeria, and North of Malaysia were higher than this present work.

Table 3: Radiological Parametric Indices (RPIs) for Soil, Umuokwom Ohia in Umuahia

SID	D (nGyh^{-1})	$AEDE_{out}$ (mSvy^{-1})	$AEDE_{in}$ (mSvy^{-1})	AGED (μSvy^{-1})	Ra$_{eq}$ (BqKg^{-1})	I (mSvy^{-1})
USB1	48	0.06	0.23	327	109	0.38
USB2	44	0.05	0.21	304	96	0.35
USB3	32	0.04	0.16	223	70	0.25
USB4	30	0.04	0.15	210	66	0.24
USB5	34	0.04	0.17	236	74	0.26
MIN	32	0.04	0.15	210	70	0.24
MAX	48	0.06	0.23	327	109	0.38
MEAN	37	0.05	0.18	260	83	0.29
GLOBL MEAN	59	0.080	0.42	300	370	1

Table 4: Comparison of Radiological Parameters Indices (RPIs) for Soil of Umuokwom Ohia in Umuahia

Location	**D ($nGyh^{-1}$)**	**$AEDE_{out}$ ($mSvy^1$)**	**$AEDE_{in}$ ($mSvy^1$)**	**Ra_{eq} ($BqKg^1$)**	**AGED (μSvy^1)**	**I ($mSvy^{-1}$)**	**Reference**
Umuahia Nigeria	37	0.05	0.18	82.91	260.03	0.29	Present Work
FUTO, Nigeria	28.75±10.39	35.28±12.74		59.79± 16.98	186.00± 50.65	0.16± 0.05	Eke *et al.*, 2024
Umuahia, Nigeria	75±12	0.09±0.01	0.37±0.06	137±28	530±83	-	Onudibia *et al.*, 2023
Iraq	1 6.673±1.7	0.020±0.02	0.082±0.08	34.143±3.54		0.092±0	Tarbool, *et al.*, 2022
AlHusseinea Iraq	29.7±1.2	0.03±0.001	-	61.11±2.5	212.1±9.0	0.165±0.01	Ibrahim *et al.*, 2021
Jeddah, Saudi Arabia	31.68	0.039	-	63.93	-	0.17	Al-Ghamdi, 2019
Ado-Odo/Ota Nigeria	81.32	-	0.86	185.82	--	0.50	Joel *et al.*, 2018
Ogun State Nigeria	110.15	0.13	-	252.33	-	0.68	Adewoyin *et al.*, 2018
North of Malaysia	141.62	0.169	-	458.785	-	0.859	Alzubaidi, 2016
GLOBAL MEAN	59	0.080	0.42	370	300	1	---

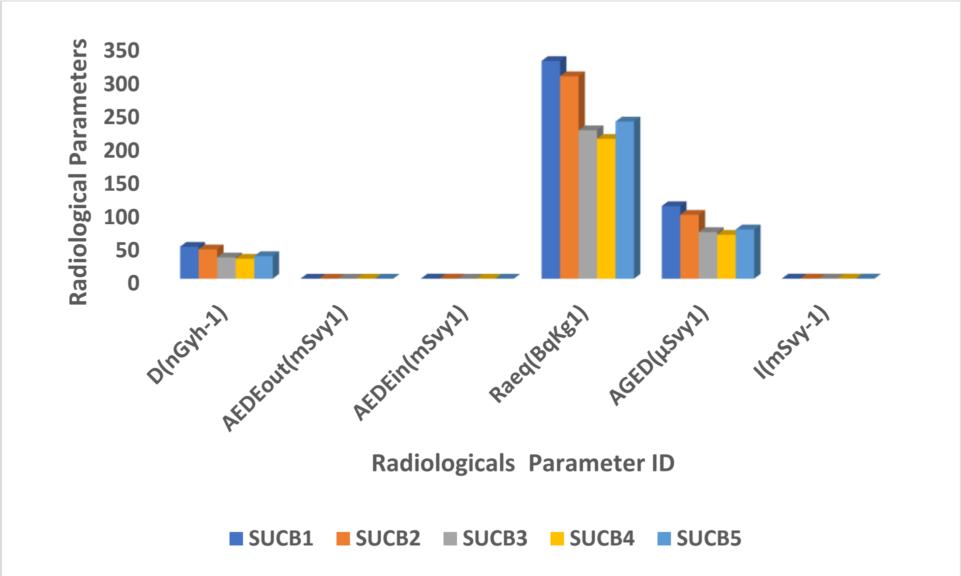

Figure 6: Bar Graph of Distribution Comparison of the Radiological Parameters Indices (RPIs)

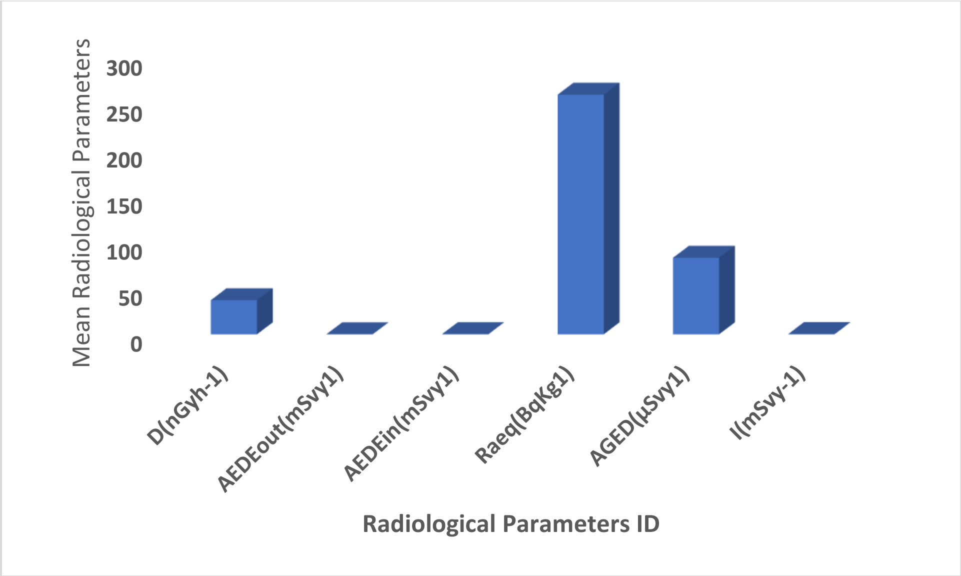

Figure 7: Bar Graph of Mean Distribution of the Radiological Parameters Indices (RPIs)

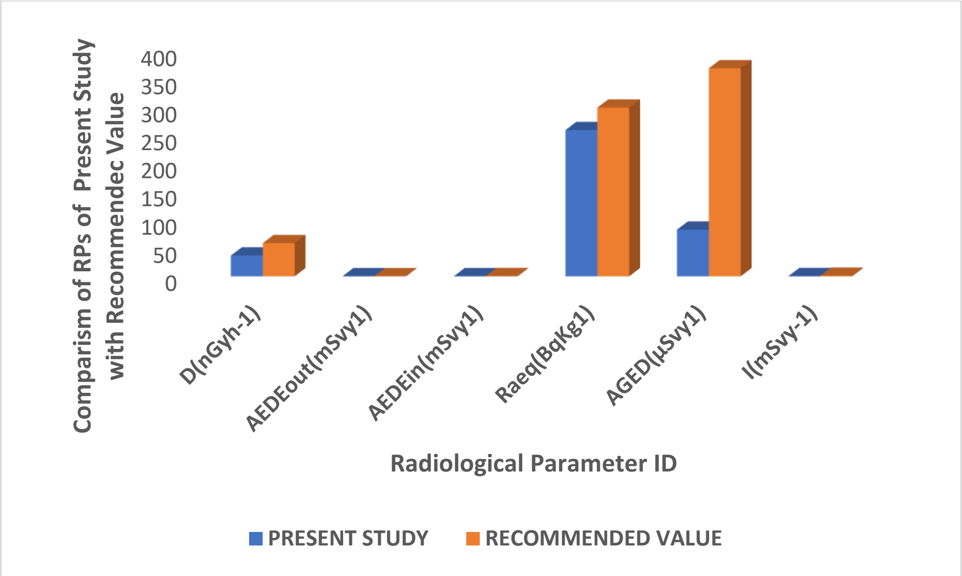

Figure 8: Bar Graph of Mean Distribution Comparison of the Radiological Parameters Indices (RPIs) of the Present Study with Recommended Value

Correlation Analysis

Pearson's Correlation Coefficient (PCC) was used to examine the relationship existing between the activity concentration of the NORs (^{40}K, ^{226}Ra and ^{232}Th) and RPIs (D, $AEDE_{out}$, $AEDE_{in}$, AGED, Ra_{eq} and I.

PCC is categorized into the following direct relation that exist between all the terms with the confidence ranges of the coefficients which are: very strong (0.8–1.00), strong (0.4–0.79), weak (0.00–0.19) and moderate (0.2–0.39) (Ion *et al.,* 2022). The obtained PCC are shown in Table 5.

Potassium-40 indicated a negative correlation with all RPIs. The radionuclides ^{226}Ra and ^{232}Th indicated a very strong relationship with all RPIs. This indicate that ^{226}Ra and ^{232}Th are the responsible for the observed doses and radiological parameters values obtained and that 40K have a different source of the U and Th decay products.

Table 5: Correlation-Analysis between the Activity Concentration of Radionuclides and Radiological Parameters Indices (RPI)

	^{40}*K*	^{226}*Ra*	^{232}*Th*	*D*	$AEDE_{out}$	$AEDE_{in}$	*AGDE*	Ra_{eq}	*I*
^{40}K	1								
^{226}Ra	-0.4481	1							
^{232}Th	-0.8446	0.84342	1						
D	-0.5387	0.99213	0.90019	1					
$AEDE_{out}$	-0.7282	0.91682	0.98130	0.95937	1				
$AEDE_{in}$	-0.5577	0.99133	0.90651	0.9992	0.96014	1			
AGDE	-0.5200	0.99367	0.89110	0.99974	0.95440	0.99821	1		
Ra_{eq}	-0.5988	0.9810	0.92991	0.99724	0.97672	0.99770	0.99544	1	
I	-0.5474	0.98701	0.90776	0.99890	0.96620	0.99695	0.99867	0.99741	1

CONCLUSION

The activity concentration values for NORs ^{40}K, ^{226}Ra, ^{232}Thfound in this study were lower than average value according to UNSCEAR with a relatively uniform distribution in the studied area. As expected, ^{40}K had the higher values, but is the one with the lower contribution for the dose exposure. The mean value of the RPIs were also lowers than global average or permissible limit reported in literature. The finding of this work has shown that the radionuclides concentration available in the soil is low and do not represent a threat for human health, hence the soil is safe for indoor and outdoor activities such as Agriculture, building and construction purposes.

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