

Natural Radionuclide Concentration in Kaolin Deposits and Dose Assessment Within Delta State, Nigeria

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ABSTRACT

The assessment of natural radionuclide concentration in kaolin deposits and dose rate within Delta state, Nigeria has been carried out. Four kaolin mining sites within four Local Government Areas in the state were studied. Background Ionizing Radiation levels of each of the mining site was measured using a Gamma Scout handheld radiation meter and a GPS meter used to obtain the actual positions where the BIR levels were taken around each kaolin site. A total of twenty-five (25) samples were collected for Gamma spectrometry analysis at National Institute of Radiation Protection and Research, University of Ibadan. The mean BIR exposure rates for Ukwuani, Aniocha South, Ughelli North and Ughelli South LGAs were 0.01mRh^{-1} , 0.013mRh^{-1} , 0.013mRh^{-1} and 0.014mRh^{-1} respectively and were within the world's recommended permissible limit of 0.013mRh^{-1} given by UNSCEAR. The mean activity concentrations of ^{238}U , ^{232}Th and ^{40}K in kaolin samples were estimated to be 137.028 ± 7.194 , 13.532 ± 1.568 and $6.724 \pm 0.396 \text{ Bqkg}^{-1}$ respectively for Ukwuani; 425.494 ± 22.16 , 18.63 ± 2.17 and $11.955 \pm 0.71 \text{ Bqkg}^{-1}$ respectively for Ughelli South; 596 ± 30.772 , 18.728 ± 2.124 and $8.912 \pm 0.526 \text{ Bqkg}^{-1}$ respectively for Ughelli North and finally, 120.026 ± 6.235 , 10.533 ± 1.182 and $15.024 \pm 0.879 \text{ Bqkg}^{-1}$ respectively for Aniocha South. These results compare quite well with the world's average value of activity concentrations for natural radionuclides reported by UNSCEAR, except for ^{40}K which is higher than the world's average at Ughelli North and South. The values of all the radiological hazard parameters for all the mining sites are less than the recommended limits. The calculated Excess Lifetime Cancer Risk also reveal that the values are all within the recommended limits, therefore, the risk of cancer inducement due to radiation exposure is within the acceptable limits for all the mining sites.

1. INTRODUCTION

Radiation can be found everywhere in the universe and in the surrounding environment of man. There are many types of radiation that range from ionizing radiation to non-ionizing radiation. Ionizing radiation possess enough energy to knock off electrons from materials in their path while non-ionizing radiation do not possess such energy. The sources of ionizing radiation in the environment include natural sources and artificial sources. The natural source includes cosmogenic

radiation as well as radionuclide deposits in the earth crust while artificial sources include manmade machines like the x-ray tube, cyclotron, accelerators, and nuclear reactors (Hatra, 2018). The radioactive materials can be divided into two categories with the first category being naturally occurring radioactive materials (NORM) which are radioactive materials that are found in their natural state in the environment, and the second category being technologically enhanced radioactive material (TENORM) which are radioactive materials synthesized from NORM in the laboratory or nuclear reactors. However, the greatest exposure to radiation comes from naturally occurring radionuclides. Natural sources of radioactivity do contribute to approximately 80% of the collective radiation exposure of the world's population (UNSCEAR, 1996). All of these natural sources of radioactivity can be lead to internal and external radiation exposure. External exposure occurs through the emission of penetrating gamma rays from radioactive sources. Therefore, it is important to measure the radioactivity levels in the built-up areas of the environment to assess the radiological consequences of the ionizing radiation eminent in such an environment and take commensurate measures to avoiding as well as minimizing such consequences.

Soil forms a major component of an ecosystem and it is the most endangered due to the influence of various human activities such as urban development, industrial and technological advancements, agricultural practices and indiscriminate waste disposal. Soil is considered contaminated when chemicals are present or other alterations have been made to its natural environment (Gowd et al., 2010).

Kaolin, which is also referred to as China clay or soft white clay, is a naturally occurring mineral that basically contains about 85 percent of kaolinite and it is formed through weathering or breaking down of the mother rock under favorable conditions. In addition, kaolin typically contains small amounts of mica, feldspar, illite and quartz. Kaolin is named after the hill in China (Kaoling) from which it was mined for centuries. Owing to its good physical, mineralogical and chemical characteristics, kaolin is extensively utilized as a raw material in the production of ceramics, cements, paints, refractory bricks, tiles, papers, drugs, toothpastes, fabrics, rubbers and plastics (Turhan, 2009). Kaolin is one of the various types of clay soil found in nature, with the chemical composition of $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$.

Natural occurring radioactive material (NORM) and their decay products in mining sites present severe environmental and radiological hazards to both miners and the general public. These NORMs contain primordial radionuclides like Uranium (^{238}U) and its series progenies, Thorium (^{232}Th) and its series progenies and ^{40}K and its series progenies. NORMs remain one of the major harmful radiation sources that cause radiological hazards (Akpan et al. 2020). Some primordial radionuclides have physical half-lives commensurable to age of the earth (Kolo, 2016) and studies have revealed that about 95% of the entire radiation exposure on the surface of this planet emanates from natural sources. On the other hand, artificial sources account for about 5 to 20% of the radiation exposure on the planet (Jaworowski, 1999). Therefore, the need to evaluate the radiological hazard levels and the distribution of natural occurring radioactive materials (NORM) - ^{238}U , ^{232}Th , and ^{40}K in these mining sites with suspected increase in radioactive concentration cannot be over emphasized. This will facilitate the general evaluation of all radiological hazard indices associated with these mining sites.

The assessment of the background ionizing radiation and activity concentration in the environment and soil of numerous areas has gained much traction as the world radiation regulating bodies are poised to ensure that adverse impacts of ionizing radiation is reduced to a minimum (UNSCEAR, 2000). While some areas show results that are higher than the permissible standard or show results equal to or lower than it.

In Nigeria, the average activity concentration of radium, ^{238}U , and ^{232}Th equivalent obtained from some areas in Ogun State and Ekiti State is lower than 370 BqKg^{-1} which is the world's permissible limit (Usikalu et al. 2015; Usikalu et al. 2019). Also the ^{238}U , ^{232}Th and ^{40}K concentration from studies in parts of Ilorin South and Ilorin West in Kwara State, Ekiti State, Akwa-Ibom Coastal Line, and some mining sites in Abia State all showed lower results when compared to the permissible limit (Orosun et al. 2022; Akpan et al. 2020; Enyinna, 2015). Outside of Nigeria, results obtained by Kapanadze et al. (2019) from analysis of ^{40}K , ^{238}U , and ^{232}Th concentration in Khrami Late Variscan crystal massif occurring in Georgia are also found to within the permissible limit. On the other hand, ^{40}K concentration from a study by Usikalu et al. (2019) in mining and living areas in Ekiti State, Nigeria appears to be higher than the permissible limit. Same goes with the research in selected areas in Egypt where it was found that the concentration of radium in the kaolin of the area is higher than the world acceptable standard (Gad et al. 2019)

This study aims to measure the natural radionuclide concentration ^{40}K , ^{238}U , ^{232}Th in kaolin samples and dose assessment within some mining sites in Delta state, Nigeria. The first step to achieving this is by recording the background ionizing radiation (BIR) levels around the mining sites. The results obtained will be employed to estimate the radiological hazard indices as well as compare the results with the world safety limit and those obtained from researches in other places.

Study area

This research is conducted within Delta State. Delta State is an oil and agricultural producing State in Nigeria. It is situated in the region known as the South-South geo-political zone with a population of 4,112,445 (NBS, 2013). The capital city is Asaba, located at the northern end of the State, with an estimated area of 762 square kilometres (294 sq. mi), while Warri is the economic nerve centre of the State and also the most populated. It is located in the Southern end of the State. The State has a total land area of 16,842 square kilometres (6,503 sq. mi). There are various solid mineral deposits within the State – industrial clay, silica, lignite, kaolin, tar sand, decorative rocks, limestone etc. These are raw materials for industries such as brick making, ceramics, bottle manufacturing, glass manufacturing, chemical/insulators production, chalk manufacturing and sanitary wares, decorative stone cutting and quarrying. Large deposits of silica sand can be found throughout the State in various lithological formations and along the banks of rivers and streams. They are used in the production of various types of glass silica, which is the most important raw material in the production of glass. Finally, clay layers can be found in the Tertiary and Quaternary formations. These are particularly abundant in Ughelli, where stream clays are used for glass factory moulding. One mining area in four local governments areas were chosen in the State. They are Akokuo-Uno in Ukwuani, Otor-Edo in Ughelli-South, Eghighori in Ughelli-North, and Anioma in Aniocha-South.

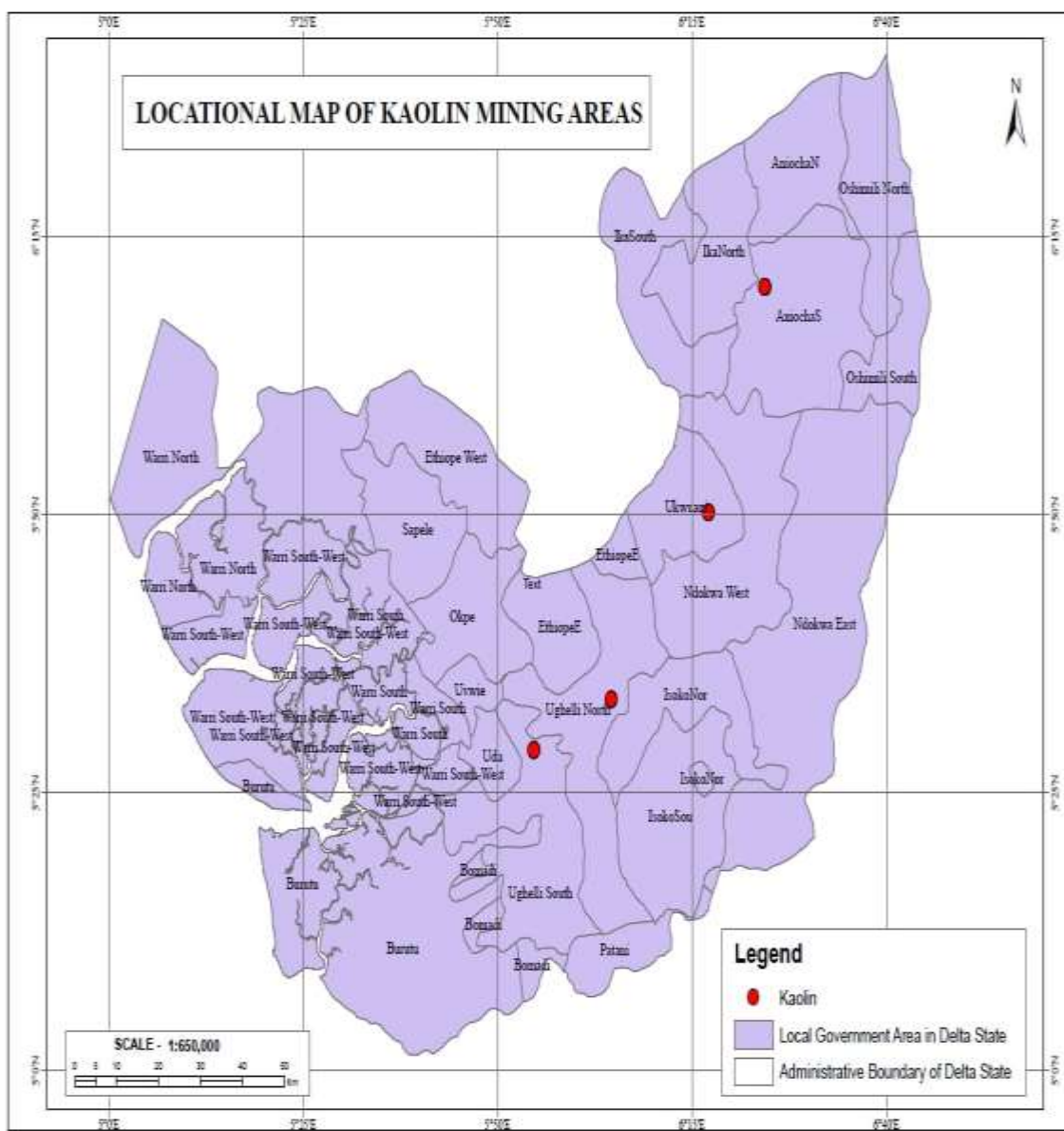


Figure 1: Map of Delta State showing Kaolin Mining Sites where Samples were obtained.

2. MATERIALS AND METHODS

2.1 Materials

A well calibrated Gamma Scout handheld radiation meter was used to measure the background ionizing radiation of the areas around the collection points and a GPS receiver was used to measure the coordinates as well as the elevation of the sites.

Gamma-ray spectroscopy is the quantitative study of the energy spectra of gamma-ray sources, both nuclear laboratories and astrophysical. Gamma-ray spectrometer also determines the energies

of the gamma-ray photons emitted by the source. It is an instrument of high proximity and accuracy in analyzing the emitted gamma rays in the speculated samples. NaI(Tl) detector is the standard scintillation material for gamma ray spectroscopy (Knoll and Kraner, 1989). It has an excellent light yield and a linear response to electrons and gamma rays.

2.2 Samples collection and preparation

Kaolin samples were collected from four mining sites in four different local government areas of Delta State in their raw form into black polythene bags from different spots around each mining site, a total of twenty-five (25) samples in all. Clean hand shovel was used in scooping the samples to forestall contamination. The kaolin samples were dried in an oven at low temperature, crushed into smaller bits and sieved using a mesh of 1 millimeter diameter. Samples were prepared into different portions of 700 grams per portion placed in Ziploc bags and transferred to the National Institute of Radiation Protection and Research at the University of Ibadan for γ -analysis.

2.3 Risk and health hazard analysis

The radiological health effects on humans from activity concentration of radionuclides (^{238}U , ^{232}Th , and ^{40}K) found in the collected samples were calculated using some radiation hazard indices. They include equivalent dose rate, absorbed dose rate, Annual Effective Dose Equivalent (AEDE), Excess Lifetime Cancer Risk, Gamma Dose Rate, Activity Concentration Index, External Hazard Indices and Radium Equivalent Activity.

3. RESULTS AND DISCUSSION

3.1 The Background Ionizing Radiation (BIR) and radiological health risks.

Table shows the background radiation and its associated health risk. The mean exposure rate range from $0.01 \pm 0.0017 \text{ mRh}^{-1}$ in Ukwuani LGA to $0.014 \pm 0.0009 \text{ mRh}^{-1}$ in Ughelli South LGA as shown in Table 1. The results obtained in Ughelli South Local Government Area was the same as that of Uzo-uwani (0.014 mRh^{-1}) as reported by Osimobi et al. (2015) but higher when compared with Enugu coal mining site (0.009 mRh^{-1}) as reported by Agbalagba (2016). The values are lower than that of the world's recommended limit (UNSCEAR, 2000) evident from Figure 2. The obtained values are lower due to the negligible quantities of radionuclide bearing sediments in the area.

The absorbed dose that is used to assess the potential for any biochemical changes in specific tissues as estimated from the background radiation ranges from 86.876 nGyh^{-1} in Ukwuani LGA to 119.99 nGyh^{-1} in Ughelli South LGA. The results are comparable to those of Agbalagba (2016) and Ilugo et al. (2021). According to Table 1, the results are higher than the standard of 84.0 nGyh^{-1} (UNSCEAR, 2000).

Estimated values for Annual effective dose equivalent (AEDE) in the four locations are higher than the world standard of $70 \text{ } \mu\text{Svy}^{-1}$ (ICRP, 2007).

The figures for excess lifetime cancer risk (ELCR)is which predicts the probability of an individual developing cancer over his/her lifetime due to low radiation dose exposure are higher than the world standard of 0.29×10^{-3} . The values which fall within those reported by Agbalagba (2016) and Ilugo (2021) raise some concern as they indicate the existence of possibilities that occupants of those surroundings could develop cancer in their lifetime.

Table 1: Average values of BIR and the calculated hazard indices of all the study areas.

LOCATION	GPS Coordinates	BIR (mRh ⁻¹)	EDR (mSvy ⁻¹)	ADR(nGyh ⁻¹)	AEDE (μSvy ⁻¹)	ELCR (×10 ⁻³)
UKWUANI	N05° 50.207' E006°17.127'	0.01 ± 0.0017	0.841 ± 0.14	86.876± 14.57	106.55 ± 17.87	0.373
ANIOCHA SOUTH	N06°10.484' E006°24.382'	0.013±0.003	1.071±0.243	110.8 ± 25.166	135.89±30.864	0.476
UGHELLI SOUTH	N05°28.812' E005°54.690'	0.014±0.0009	1.162±0.079	119.99±8.226	147.16±10.09	0.515
UGHELLI NORTH	N05°33.392' E006°04.654'	0.013±0.003	1.096±0.316	109.12±29.007	133.90±35.55	0.469

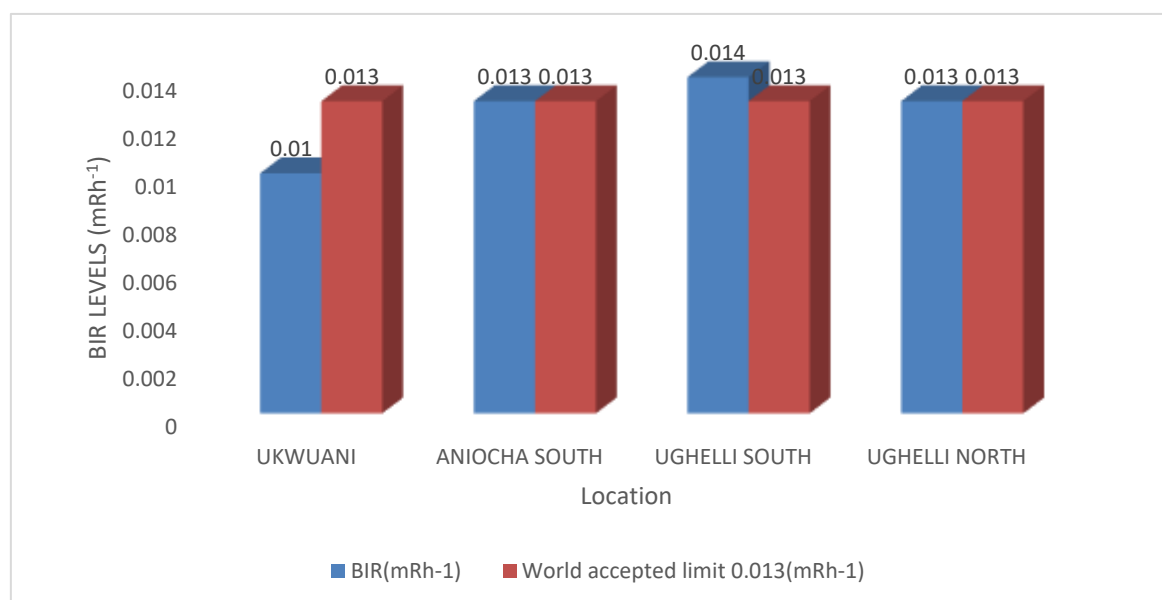


Figure 2: Comparison of the BIR values of the four locations with the world acceptable limit (UNSCEAR, 2000).

3.2 Discussion of Activity Concentrations of ²³⁸U, ²³²Th and ⁴⁰K in Kaolin and hazard indices.

The mean values for the activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in Kaolin samples are shown in Table 2. The values of activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th vary from 26.96, 3.1 and 4.7 Bqkg⁻¹ to 212.38, 25.82 and 8.22 Bqkg⁻¹ in Ukwuani, with mean values of 137.028, 13.532 and 6.724 Bqkg⁻¹ respectively. In Ughelli South, the activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th range from 338.42, 9.37 and 9.62 Bqkg⁻¹ to 490, 35.73 and 13.84 Bqkg⁻¹ with mean values of 425.494, 18.63 and 11.955 Bqkg⁻¹ respectively. Ughelli North has activity concentrations for ⁴⁰K, ²³⁸U and ²³²Th ranging from 373.63, 10.95 and 6.11 Bqkg⁻¹ to 765.97, 31.33 and 11.75 Bqkg⁻¹ with mean values 596, 18.728 and 8.912 Bqkg⁻¹ respectively, while for Aniocha South, the activity concentrations of ⁴⁰K, ²³⁸U and ²³²Th range from 19.05, 0.61 and 9.68 Bqkg⁻¹ to 252.35, 30.03 and

22.46 Bqkg⁻¹ with mean values of 120.026, 10.533 and 15.024 Bqkg⁻¹ respectively. The activity concentration of ⁴⁰K, ²³⁸U and ²³²Th in Kaolin samples from the studied areas were compared with similar works investigated in other parts of Nigeria and other countries and the summary results presented in Table 3. All four mining sites under study had values that were lower than the world's permissible limits (UNSCEAR, 2000) except for Ughelli North and Ughelli South that had higher values of ⁴⁰K as indicated in Table 3.

The Gamma dose rate in kaolin samples ranged from 16.0271 nGyh⁻¹ in Ukwuani to 38.8884 nGyh⁻¹ in Ughelli North with a mean value of 26.8581 nGyh⁻¹. When compared with Akpan et al. (2020) value of 39 nGyh⁻¹ and Echeweozo et al. (2021) value of 70.6 nGyh⁻¹, it can be noted that values in this present study are fairly below world average of 84 nGyh⁻¹ given by UNSCEAR (2000) as shown in Figure 3, and may not pose any threat to the health of the miners and the residents living in the environs.

The Annual Effective Dose Equivalent (AEDE) values ranged from 19.6556 μSvy⁻¹ for Ukwuani mining site to 47.6927 μSvy⁻¹ for Ughelli North, with a mean value of 32.9230 μSvy⁻¹. Figure 4 shows that these values are lower than the world standard of 70 μSvy⁻¹ and compare quite well with other works (Echeweozo and Okeke, 2021).

The Excess Lifetime Cancer Risk (ELCR) in kaolin samples ranges from 0.068 for Ukwuani LGA to 0.167 for Ughelli North LGA, with mean value of 0.115. It is clearly shown in Figure 5 that these values were below the limit as presented by UNSCEAR (2000) which indicates that the miners and residents living around the mining sites are safe and may not likely develop cancerous growth due to kaolin mining.

The Radium Equivalent activity (Ra_{eq}) ranged from 33.6985 in Ukwuani LGA to 77.3642 in Ughelli North LGA with a mean value of 55.2027 Bqkg⁻¹. These values compared quite well with Adagunodo et al. (2018) and, from Figure 8, are well below the permissible limit of 370 Bqkg⁻¹ given by (UNSCEAR, 2000).

Figures 6 and 7 shows the comparison of the gamma representative Index (I_γ) and hazard indices with the world permissible standard respectively. Gamma Representative Index (I_γ) ranged from 0.2488 in Ukwuani LGA to 0.6113 in Ughelli North LGA with a mean value of 0.422. This compared well with Adagunodo et al. (2019) value of 0.6 and lower than the recommended limit of unity set by (UNSCEAR, 2000). The external hazard indices ranged from 0.091 in Ukwuani LGA to 0.2089 in Ughelli North LGA with mean value of 0.149. These values compared well with other works done in Nigeria and are also below the world standard of unity.

Table 2: Mean values of activity concentrations in Kaolin Samples gotten from Gamma Spectroscopy Analysis in Delta State.

S/N	SAMPLE LOCATION	⁴⁰ K (Bq/kg)	²³⁸ U (Bq/kg)	²³² Th (Bq/kg)
1.	UKWANI	137.028 ± 7.194	13.532 ± 1.568	6.724 ± 0.396
2.	UGHELLI SOUTH	425.494 ± 22.166	18.63 ± 2.17	11.955 ± 0.71

3.	UGHELLI NORTH	596 ± 30.772	18.728 ± 2.124	8.912 ± 0.526
4.	ANIOCHA SOUTH	120.026 ± 6.235	10.533 ± 1.182	15.024 ± 0.879
5.	WORLD AVERAGE (UNSCEAR, 2000)	420	33	45

Table 3: Comparison of mean activity concentration with other studies.

LOCATION	SAMPLES	²³⁸ U(Bq/Kg)	²³² Th(Bq/Kg)	⁴⁰ K(Bq/Kg)	REFERENCES
Umuahia, Abia	Kaolin	76.29	47.15	164.96	Echeweozo et al. (2021)
Ilorin-south, Kwara	Kaolin	42.73	27.06	92.34	Orosun et al. (2021)
Ilorin-west, Kwara	Kaolin	23.8	32.4	189.28	Orosun et al. (2021)
Ekiti state	Clay	33.6	20.1	207.2	Isinkaye et al. (2009)
Ifoyintedo	Kaolin	38.2	65.1	93.9	Adagunado et al. (2018)
Southern Iraqi Governorates	Soil	3.95	2.53	260.36	Mohammed et al. (2018)
River Uzo-uwani, Enugu state	Kaolin	-	67.3	81.6	Osimobi et al. (2015)
Rivers state	Soil	29.16	32.21	222.15	Avwiri et al. (2014)
Ukwuani, Delta state	Kaolin	13.532	6.724	137.028	Present study
Ughelli South, Delta state	Kaolin	18.63	11.955	425.494	Present study
Ughelli North, Delta state	Kaolin	18.728	8.912	596	Present study
Aniocha South, Delta state	Kaolin	10.533	15.024	120.026	Present study
World Average (UNSCEAR, 2000)		33	45	420	

Table 4: Calculated radiometric parameters such as Absorbed Dose rate (ADR), AEDE, ELCR, I_r , H_{ex} , and R_{eq} .

S/N	LOCATION	ADR (nGyh ⁻¹)	AEDE (μ Svy ⁻¹)	ELCR ($\times 10^{-3}$)	I_r	H_{ex}	R_{eq} . (Bqkg ⁻¹)
1.	UKWANI	16.0271	19.6556	0.068	0.2488	0.0910	33.6985
2.	UGHELLI SOUTH	33.5710	41.1715	0.1441	0.5274	0.1850	68.4887
3.	UGHELLI NORTH	38.8884	47.6927	0.1669	0.6113	0.2089	77.3642
4.	ANIOCHA SOUTH	18.9458	23.2351	0.0813	0.3005	0.1114	41.2593
5.	WORLD AVERAGE	84	70	0.29	1	1	370

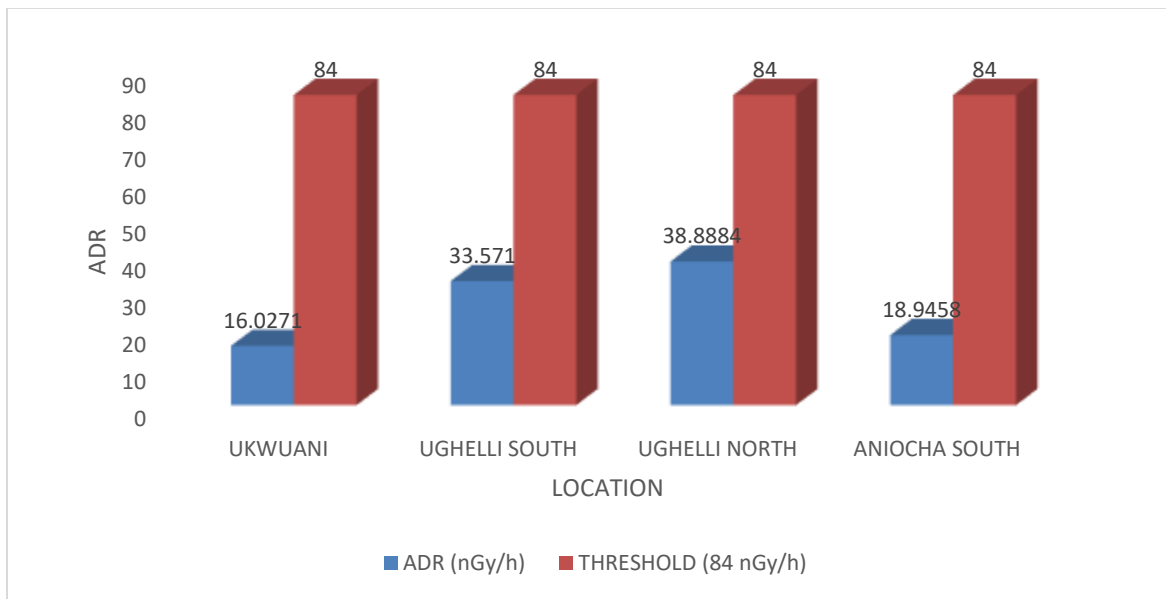


Figure 3: Comparison of the Dose Rate in the various mining sites with the world's recommended limit (UNSCEAR, 2000).

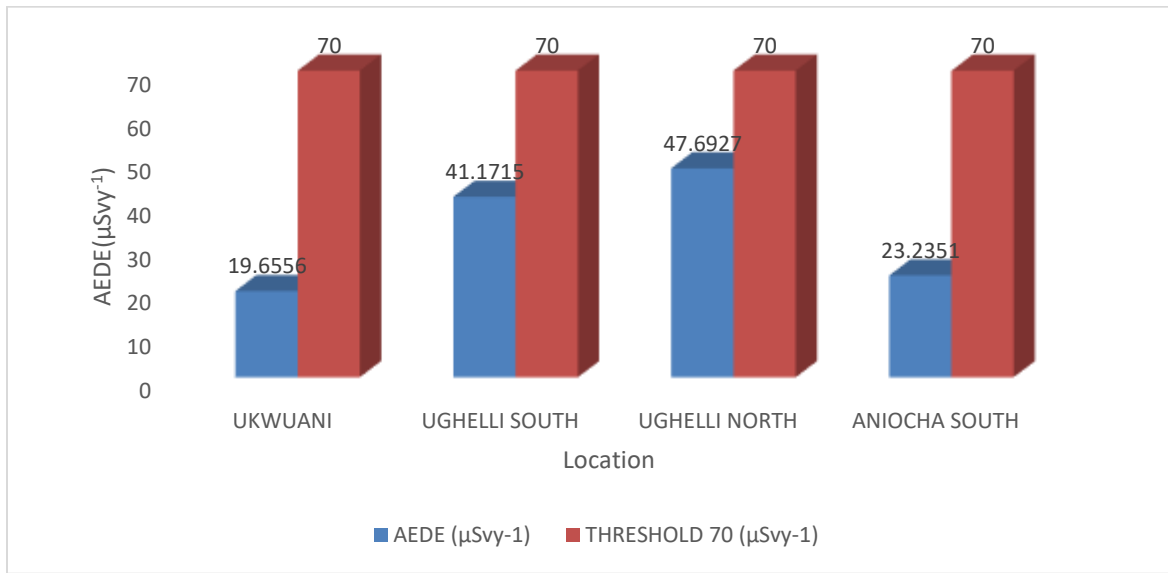


Figure 4: Comparing AEDE of the different locations with world standard (UNSCEAR, 2000).

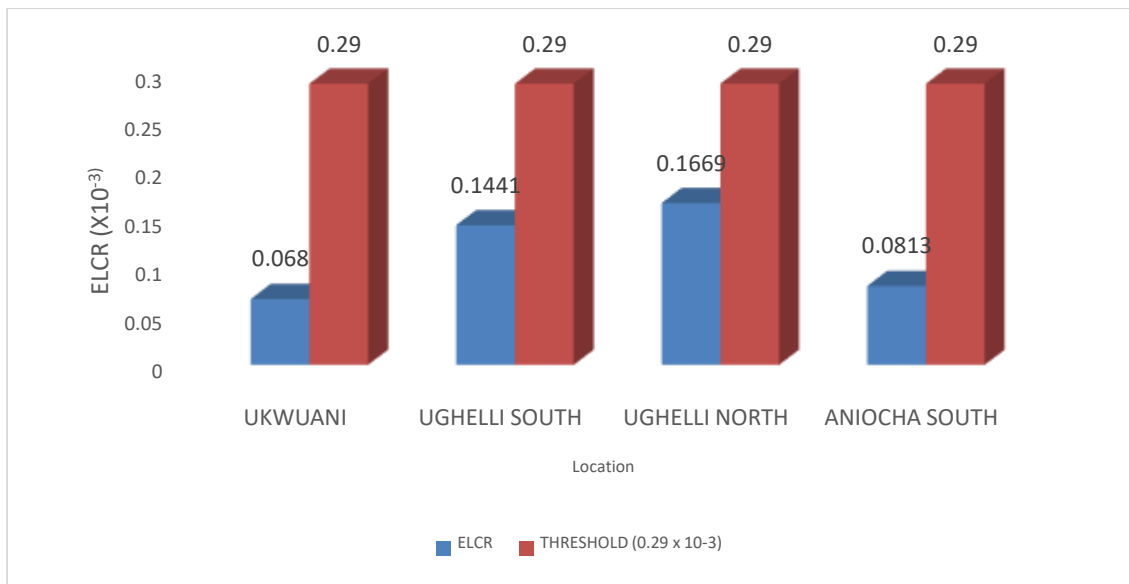


Figure 5: Comparing ELCR of Kaolin samples the different locations with world standard (UNSCEAR, 2000).

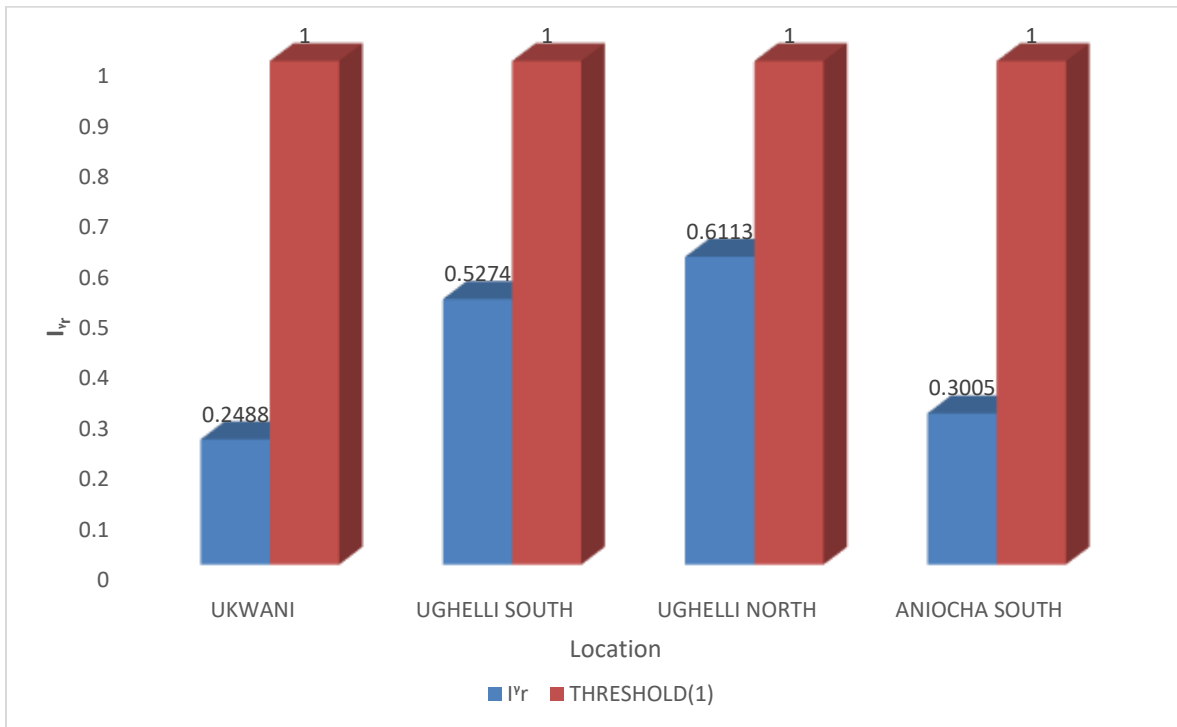


Figure 6: Comparing I_r of the different locations with world standard (UNSCEAR, 2000).

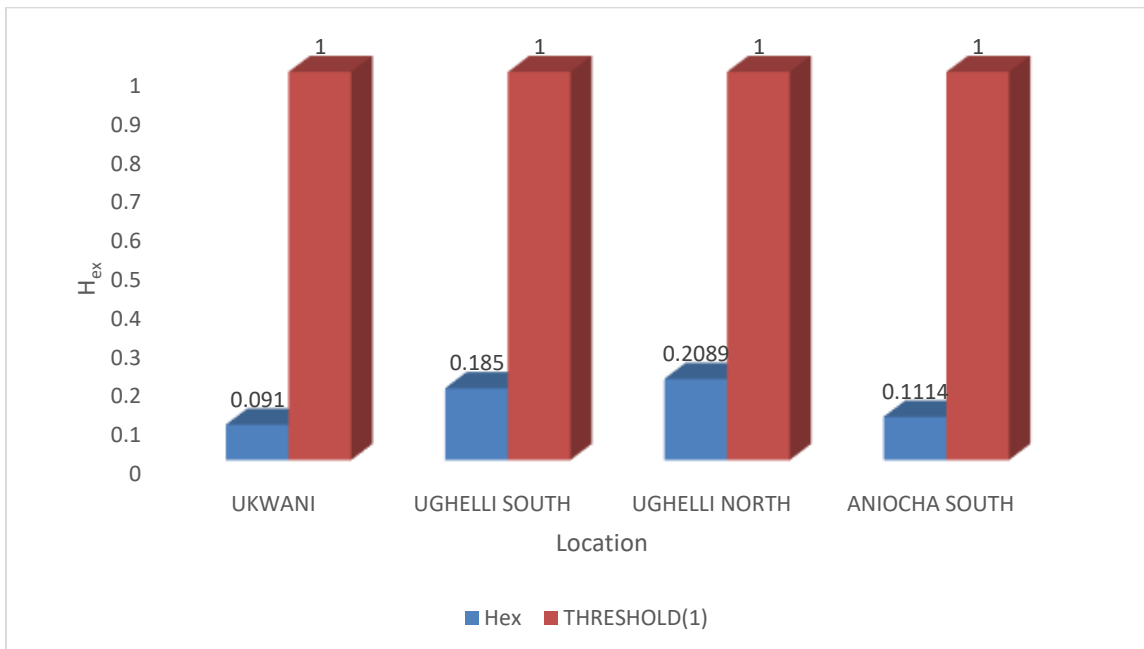


Figure 7: Comparing H_{ex} of the different locations with world standard (UNSCEAR, 2000).

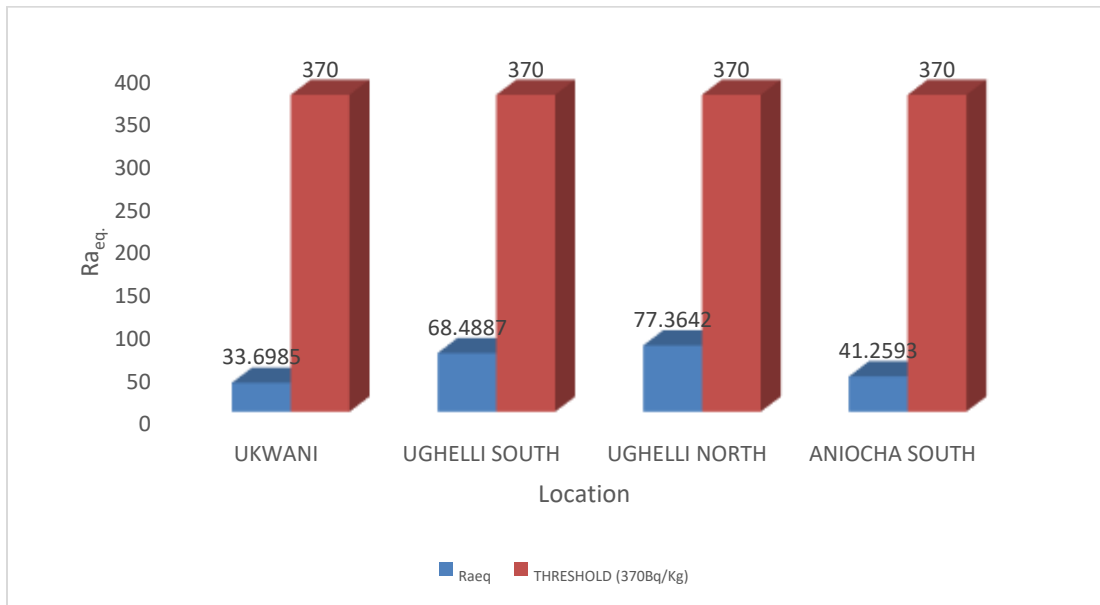


Figure 8: Comparing Ra_{eq} of the different locations with world standard (UNSCEAR, 2000).

4 Conclusion

This study tries to obtain the activity concentrations of natural radionuclides (^{238}U , ^{232}Th and ^{40}K) found in Kaolin samples from selected mining sites in of Delta state, Nigeria. The data obtained was used to estimate the radiological health risk factors that the entire population in the area could be exposed to. Some of the radiological risks indices include the Absorbed Dose, Annual Effective Dose Equivalent, Excess Lifetime Cancer Risk, External Hazard Indices, Gamma Representative Index, Radium Equivalent. The estimated mean values (lab results) of ^{238}U and ^{40}K for the kaolin mining fields at Ughelli North and Ughelli South are higher than those of Ukwani and Aniocha South. However, the activity concentration of ^{232}Th at Ughelli South and Aniocha South were higher compared with those of Ukwani and Ughelli North. The mean estimated values for all the radiological parameters for Ughelli North and Ughelli South were comparably higher than those of Ukwani and Aniocha South. This shows that the Ughelli North and Ughelli South kaolin mining fields pose more significant source of radiation hazard. The values of all the hazard parameters for all the mining sites are less than the recommended limits. The calculated Excess Lifetime Cancer Risk also reveal that the values are all within the recommended limits, therefore, the risk of cancer inducement due to radiation exposure is within the acceptable limits for all the mining sites. The study also recommends that assessment of radiological concentrations in plants grown in the study areas should be carried out to forestall any possibility of consumptions of food that has high radionuclide contents.

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