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Activity Concentrations and Distribution of ^{40}K, ^{232}Th, and ^{238}U with Respect to Depth and Associated Radiation Risks in Three Kaolin Mining Sites in Umuahia, Nigeria.

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Abstract: Activity concentrations and distribution of ^{40}K, ^{232}Th, and ^{238}U with respect to depth and associated radiation hazards in three major kaolin mining sites in Umuahia, Nigeria was evaluated. Kaolin samples mined at different depth (5 Ft, 20 Ft, 40 Ft) from each of the three mining sites were analyzed. The average activity concentrations of ^{40}K, ^{232}Th, and ^{238}U at the mining sites are 164.96 Bq/Kg, 47.15 Bq/kg and 76.29 Bq/kg respectively. Estimated radiological hazard indices showed that the mean values of Radium Equivalent, External and Internal Hazards, Absorbed dose rate, Annual Effective Dose, Excess lifetime cancer risk and Gamma active index are 156.42 Bq/kg, 0.42, 0.63, 70.6 $nGyh^{-1}$, 0.433 $mSvy^{-1}$, 1.51 x 10^{-3} and 1.092 respectively. Comparing the activity concentration of the mean value and their radiation exposure levels with the recommended standards, the measured radiological hazards indices were within the recommended safety limit. Furthermore, the study shows that activity concentrations of Uranium and Thorium were marginally elevated at lower regions of Kaolin mining site in Umuahia.
Keywords: Radionuclides, Kaolin mining, Activity concentration, Radiation hazard, Umuahia.

1 Introduction

Natural occurring radioactive materials (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 [1]. Some primordial radionuclides have physical half-lives commensurable to age of the earth [2] 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 accounts for about 5% [3]. 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 aid the general evaluation of all radiological hazard indices associated with these mining sites.

Kaolin is a type of clay found in nature; it is majorly hydrous silicate of alumina with chemical composition of $Al_2Si_2O_5(OH)_4$ [4]. Kaolin deposits which are found few meters from the earth surface are usually excavated manually or mechanically. During excavation and processing, worker and individuals around the mining site are usually exposed to these harmful radiations which can cause mutation, leukemia, various types of cancer and other health challenges [5]. Industrially, kaolin is useful in the production of paints, ceramics, paper, glass and some other building materials [6]. They are also applied in the production of insecticides, drugs and pozzolanic additives in cement and concrete [7].

Radioactivity concentrations of natural occurring radionuclides materials and their radiological hazards from different mining sites have been reported by many researchers like, [8-11]. Most of these studies did not consider the variation of activity concentration with depth in those locations, a parameter which is very important for a holistic and realistic evaluation of radiological risks for miners. The results from these researches have shown some level of elevation in activity concentration of naturally occurring radioactive materials in most mining sites.

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Therefore, the major objective of this research is to determine the distribution of ^{40}K, ^{232}Th and ^{238}U with depth in the three major kaolin mining sites and to evaluate the radiation hazard in terms of Radium equivalent activity, annual effective dose, absorbed dose rate, External and internal hazard index, excess lifetime cancer risk and gamma active index.

2 Materials and Methods

2.1 Study Area

Umuahia the capital of Abia state Nigeria, has three major kaolin mining sites, overlaying an area of about 15.7 km^2. Kaolin is one of the major mineral resources available in commercial quantity in Abia state, majorly around Ohiya axis of Umuahia with a bulk reserve of about 74,360,000 metric tons [12]. In this research, the three major kaolin mining sites located at (5.5166^{0}N, 7.4539^{0}E), (5.5098^{0}N, 7.44250E), (5.5144^{0}N, 7.4541^{0}E) were selected for investigation. The site selection was based on high level of human and commercial activities on these sites.

Umuahia is underlain by the Benin Formation which is made up of shale/sand sediments which is part of the coastal plain sands of the Cenezoic Niger-Delta region of Nigeria that is majorly composed of assorted types of clay and alluvium deposits [13].

2.2 Sample Collection and Preparation

About one kilogram of kaolin samples was taken from each point of the selected mining sites at depth of 5 ft, 20 ft and 40 ft. These sample collection points were evenly selected for each depth. Each sample was amassed separately in neat black polythene bags, well labeled, and carefully conveyed to environmental radiation laboratory of Ladoke Akintola University of Technology, Oyo State, Nigeria. Each sample was sun dried for five (5) consecutive days and grinded using mortar and pestle, sieved through a mesh sieve 2 mm to achieve homogeneity. Each homogenized sample was then oven-dried at 120 ºC for 10 hours until the it attained constant weight. To prevent radon–222 from escaping, 600 g of each sample was separately put in properly labeled marinelli containers. hermetically sealed. Information of each sample like name of sample, date of acquisition and net weight was neatly written on each of the container. The samples were left for 28 days for parent and daughter radionuclides and their gaseous decay progenies to attain secular equilibrium.

2.3 Measurements of Radionuclide Concentration in all Samples

The measurement of radioactivity concentrations of samples was performed deploying 3" x 3" NaI(TI) detector with PMT base (made by Canberra Inc. model-802). The gamma-ray detector was surrounded by lead with 10 cm thickness to ensure reduction in background and scattered radiations.

The detector was coupled to a Canberra series 10 plus multi-channel analyzer (MCA) connected to a preamplifier base. The photopeak regions of ^{40}K (1.46 MeV); ^{214}Bi (1.76 MeV) and ^{208}Tl (2.165) respectively were used for the analysis ^{40}K, ^{238}U and ^{232}Th in the samples. A region of interest was created around the 0.662 MeV to detect and measure any trace of ^{137}Cs as an index of artificial radionuclides. All samples were counted at a constant geometry and for a constant time of 10 hours. The counting technique employed in this study is described in [14].

Ambient background counts measurement for empty container with the same container geometry was done. Considering the low level of photons emission and long decay rates of ^{238}U and ^{232}Th, their activity concentrations were difficult to measure directly. Therefore, under the condition of radioactive secular equilibrium, photopeaks of short-lived decay products were applied to determine activity concentration of ^{238}U and ^{232}Th [15,16].

The activity concentrations of radionuclides in each sample were obtained from total counts at peak emissions by subtracting background counts and divided by efficiency of the photopeak, gamma intensity of the radionuclide, mass of the sample and counting time.

The activity concentrations (C_i) of ^{40}K, ^{232}Th and ^{238}U in Bq/kg were estimated using Equation 1 [1].

$$C_i\left(\frac{Bq}{kg}\right) = \frac{NC_i}{Y_d x(E_r)_{MxT}} \qquad 1$$

NC_i = the net peak counts of the ith radionuclide, Y_d = absolute gamma decay intensity for the specific energy photopeak of the ith radionuclide, E_γ the absolute efficiency of the detector at energy, M = mass of the sample in kg, T = counting time in sec.

2.4 Associated Radiation Hazard Evaluation Indices

In general, radiological hazard evaluation considers hazard identification, risk characterization, dose-response and exposure assessment [18].

In this research, radiological hazard evaluation was achieved by measuring associated radiological hazard indices / parameters. Results of these evaluations are presented in Table 2. These associated indices are as follows;

2.4.1 Radium Equivalent Activity

Radium equivalent activity (Ra_{eq}) is the sum of activity concentrations of ^{40}K, ^{232}Th and ^{238}U [17]. It introduces uniformity in the measurement of radiation exposures from natural occurring radionuclides materials [18].

$Ra_{eq} = 0.007C_k + 1.43_{Th} + C_U$ 2 where C_k, C_{Th} and C_U are the respective activity concentrations of ^{40}K, ^{232}Th and ^{238}U. Ra_{eq} must be below 370Bq/kg so as to consider radiation hazard to be negligible. The estimated radium equivalent activities from the mining sites are

presented in Table 1.

2.4.2 Assessment of External and Internal Hazard Index

These complementing parameters measure the external and internal exposure of the radiation emanating from each sample. The external hazard index (H_{ex}) and the internal hazard index (H_{in}) were evaluated with Equations 3 and 4 respectively [19].

$$H_{ex} = \frac{C_K}{4810} + \frac{C_{Th}}{259} + \frac{C_U}{370} \quad 3$$

$$H_{in} = \frac{C_K}{4810} + \frac{C_{Th}}{259} + \frac{C_U}{185} \quad 4$$

where C_k, C_{Th} and C_U represent activity concentrations of ^{40}K, ^{232}Th and ^{238}U respectively. The specific values of H_{ex} and H_{in} should be less than unity (≤ 1) for negligible level radiation hazard [20,21].

2.4.3 Estimation of Absorbed Dose Rate (ADR)

The ADR is usually associated to human absorbed gamma dose. It is an important index in radiological hazard evaluation. For kaolin samples containing concentrations of natural radionuclides, the estimated absorbed dose rate ADR in nGyh^{-1} in air 1meter above the ground was calculated with the Equation 5;

$$ADR = aC_K + bC_{Th} + cC_U \quad 5$$

Where a, b and c are the dose rate per unit ^{40}K activity concentration (0.0417 Gyh^{-1}), dose rate per unit ^{232}Th activity concentration (0.604Gyh^{-1}), dose rate per unit ^{238}U activity concentration (0.462 Gyh^{-1}) respectively. Furthermore, C_U is the concentration of ^{238}U in the sample in (BqKg^{-1}), C_k is the concentration of ^{40}K in the sample in (BqKg^{-1}), C_{Th} is the concentration of ^{232}Th in the sample in (BqKg^{-1}).

2.4.4 Annual Effective Dose (AED)

The indoor and outdoor annual effective dose (AED_{in} and AED_{out}) in mSvy^{-1} were deduced using the Equations 6 and 7 respectively.

$$AED_{in}(mSvy^{-1}) = ADR(nGyh^{-1})x\ 0.7(SvGy^{-1})x\ 0.8\ x\ 8760(hy^{-1})x10^{-6} \quad 6$$

$$AED_{out}(mSvy^{-1}) = ADR(nGyh^{-1})x\ 0.7(SvGy^{-1})x\ 0.2\ x\ 8760(hy^{-1})x10^{-6} \quad 7$$

where ADR is referred to as absorbed dose rate in air, 8760 hours the total hours in a year; 0.7 is known as dose conversion factor from absorbed dose in air to the effective dose and occupancy factor for indoor and outdoor exposures respectively [18].

Therefore, the total Annual effective dose in (mSvy^{-1}) is the total sum of indoor and outdoor annual effective doses. That is;

$$AED_{TOTAL} = AED_{in} + AED_{out} \quad 8$$

2.4.5 Excess Lifetime Cancer Risk (ELCR)

The excess lifetime cancer risk is the possibility of an individual to develop cancer in a lifetime due to exposure to low level radiations [19-22]. It was estimated using Equation 9.

$$ELCR = AED_{TOTAL} x\ DL\ x\ RF \quad 9$$

where AED_{Total} = total annual effective dose, DL = average duration of life (70years) of an average individual [22-24] and RF = fatal cancer risk factor (0.05 Sv^{-1}) measured in per Sievert, [23-24].

2.4.5 Gamma Active Index (I_γ)

The gamma active index is a parameter for the evaluation of Υ-radiation risk linked with the natural radionuclide in measured samples at a given location. It was calculated using Equation 10

$$I_\gamma = \frac{C_K}{1500} + \frac{C_{Th}}{100} + \frac{C_U}{150} \quad 10$$

The gamma active index value ≤1, is equivalent to annual effective dose of ≤1 mSv [17,25].

3 Results and Discussion

The activity concentrations of radionuclides with respect to depth for major kaolin mining sites in Umuahia, Abia state were presented in Tables 1. From Table 1, it can be seen that the average activity concentrations of ^{40}K for different depth for each of the mining site (UM001, UM002 and UM003) were 149.25 Bqkg^{-1}, 179.82 Bqkg^{-1} and 165.8 50 Bqkg^{-1} respectively. These were far below the world average 400 Bqkg^{-1}. A closer look at the variation of activity concentrations of ^{40}K shows non-systematically decrease of activity concentration with depth. For ^{232}Th, the average activity concentration for different depth for each of the mining site (UM001, UM002 and UM003) were 38.44 Bqkg^{-1}, 54.76 Bqkg^{-1} and 48.25 Bqkg^{-1} respectively. These were slightly above the world average 30 Bqkg^{-1}. The variation of activity concentrations of ^{232}Th shows marginal non-systematically increase in activity concentration with depth. ^{238}U average activity concentration for different depth for each of the mining site (UM001, UM002 and UM003) were 68.53 Bqkg^{-1}, 86.30 Bqkg^{-1} and 74.04 Bqkg^{-1} respectively. These were above the world average 30 Bqkg^{-1}. The variation of activity concentrations of ^{238}U shows marginal non-systematically increase of activity concentration with depth. It is necessary to note that ^{238}U activities concentrations at the 40ft depth in all the selected sites were higher than the world average. Generally, mean values of the activity concentrations for all the sites were in agreement with findings made by [8] and [9] and some other mining sites in Nigeria.

From radiation hazard evaluation indices displayed in Table 2, the Ra_{eq} was between 134.99 to 178.46 Bq/kg and it is lower than the permissible limit of 370 Bq/kg [18]. The H_{ex} values ranged from 0.36 – 0.48 while H_{in} was from 0.55-0.72 fortunately, these values are less than one, which is the standard limit. The ADR was between 61.10 nGyh^{-1} and

70.26 nGyh^{-1}which is higher than the estimated world average of 57 nGy/h [18], thereby suggesting that there is elevated concentration of radionuclides in the sites. The total AED ranged from 0.374 - 0.493 mSvy^{-1} these values are less than the permissible limits of 1.00 mSvy^{-1} for individual in public place and 20.00 mSvy^{-1} for occupational workers within a year as recommended by [24] and [18].

Excess life cancer risk was between 1.309 x10^{-3} to 1.726 x10^{-3}, these values are higher than estimated world average of 0.29×10^{-3} [18]. This implies that there is slight probability of miners developing cancer over a life time. The gamma active index is within the range of 0.946-1.242. The high gamma active index above one, signify high level of gamma radiation in deeper regions of the kaolin mines. Radiologically, this calls for caution with respect to the health of miners.

The distribution of ^{40}K, ^{232}Th and ^{238}U shows great variation with depth from site to site. This means that activity concentrations of considered radionuclides are not uniform but varies from site to site and changes with depth. Most of the radiological hazard indices of all the mining sites were within acceptable safe limits [18]. This means these sites are radiological safe for mining activities. However, the marginal elevation in gamma active index signifying higher gamma radiations and possibly elevated radon exhalations signify higher presence of ^{232}Th and ^{238}U at the lower depth of the mines [26]. Which could also be confirmed with the elevated activity concentration of ^{232}Th and ^{238}U at lower depth shown in Table 1.

Activity concentration of ^{40}K, ^{232}Th and ^{238}U in kaolin samples collected at different depth from various kaolin mining sites located in Umuahia, Nigeria is as shown in Table 1.

Table 1: Activity Concentration variation with depth for kaolin samples collected from major Kaolin mining sites in Umuahia,

SITE CODE	UM001		
LOCATION	5.5166^{0}N, 7.4539^{0}E		
	Average Radionuclide Activity (Bqkg^{-1})		
DEPTH	^{40}K	^{232}Th	^{238}U
5ft (upper depth)	200.32±4.31	30.36±2.31	20.94±0.91
20ft (middle depth)	162.81±3.61	34.36±2.62	60.30±2.11
40ft (lower depth)	84.61±3.04	50.60±2.42	124.36±3.30
Mean value	149.25±3.65	38.44±2.45	68.53±2.11
***World Average**	400Bqkg^{-1}	30Bqkg^{-1}	30Bqkg^{-1}
SITE CODE	UM002		
LOCATION	5.5098^{0}N, 7.44250E		
	Average Radionuclide Activity (Bqkg^{-1})		
DEPTH	^{40}K	^{232}Th	^{238}U
5ft (upper depth)	254.20±5.32	48.65±1.54	40.34±1.32
20ft (middle depth)	183.94±3.11	54.76±2.11	75.43±2.43
40ft (lower depth)	101.32±2.30	60.87±2.74	143.23±3.42
Mean value	179.82±3.58	54.76±2.13	86.30±2.39
***World Average**	400Bqkg^{-1}	30Bqkg^{-1}	30Bqkg^{-1}
SITE CODE	UM003		
LOCATION	5.5144^{0}N, 7.4541^{0}E		
	Average Radionuclide Activity (Bqkg^{-1})		
DEPTH	^{40}K	^{232}Th	^{238}U
5ft (upper depth)	238.64±5.43	34.67±1.76	34.35±1.55
20ft (middle depth)	189.22±4.67	46.87±1.87	48.23±1.54
40ft (lower depth)	69.56±4.86	63.21±2.08	139.54±3.32
Mean value	165.81±4.98	48.25±1.90	74.04±2.14
***World Average**	400Bqkg^{-1}	30Bqkg^{-1}	30Bqkg^{-1}

* [18]

Table 2: Associated Radiation Hazard Evaluation Indices base on mean values from major Kaolin mining sites in Umuahia.

SITE CODE	Ra_{eq} (Bq/kg)	H_{ex}	H_{in}	ADR ($nGyh^{-1}$)	AED_{in} ($mSvy^{-1}$)	AED_{out} ($mSvy^{1}$)	AED_{total} ($mSvy^{-1}$)	ELCR X 10^{-3}	I_γ
UM001 (Average)	134.99	0.36	0.55	61.10	0.299	0.075	0.374	1.309	0.946
UM002 (Average)	178.46	0.48	0.72	80.44	0.394	0.099	0.493	1.726	1.242
UM003 (Average)	155.81	0.42	0.62	70.26	0.345	0.086	0.431	1.509	1.087
(Main Average)	156.42	0.42	0.63	70.60	0.346	0.086	0.433	1.515	1.092

Table 3: Gaussian distribution data for activity concentration from major Kaolin mining sites in Umuahia.

	UM001			UM002		
Statistical Variables	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U
Max	200.32	50.60	124.36	254.20	60.87	143.23
Min	84.61	30.36	20.94	101.32	48.65	40.34
Mean	149.25	38.44	68.53	179.82	54.76	86.33
SD	59.04	10.72	52.20	76.52	6.11	52.30
SEM	34.09	6.19	30.14	44.18	3.53	30.19
CV%	39.56	27.89	76.17	42.56	11.16	60.58
Skewness	-0.33	0.49	0.23	-0.80	0.0	0.30

	UM003			General Distribution Statistics		
Statistical Variables	^{40}K	^{232}Th	^{238}U	^{40}K	^{232}Th	^{238}U
Max	238.64	63.21	139	179.82	54.76	86.30
Min	69.56	34.67	34.35	149.25	38.44	68.53
Mean	165.81	48.25	74.04	164.96	47.15	76.29
SD	86.94	14.32	57.15	15.30	8.22	9.11
SEM	50.19	8.27	32.99	8.83	4.75	5.26
CV%	52.43	29.68	77.18	9.28	17.42	11.94
Skewness	-0.37	0.18	0.66	-0.10	-0.24	0.43

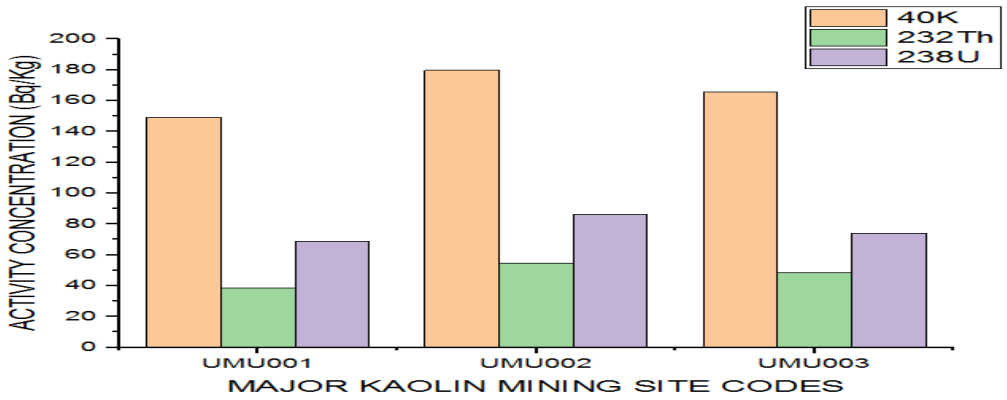

Fig.1: Variation of Activity Concentration and Distribution of ^{40}K, ^{232}Th, ^{238}U in three major kaolin mining sites in Umuahia, Abia state, Nigeria.

4 Conclusion

The average activity concentrations of ^{40}K, ^{232}Th, and ^{238}U at these mining sites are 164.96 Bq/Kg, 47.15 Bq/kg and 76.29 Bq/kg respectively. The estimated radiological hazard indices showed that the Radium Equivalent, External and Internal Hazards, Absorbed dose rate, Annual Effective Dose, Excess lifetime cancer risk and Gamma index were 156.42 Bq/kg, 0.42 0.63, 70.6 $nGyh^{-1}$, 0.433 $mSvy^{-1}$, 1.51 x 10^{-3},1.092 and 0.382 respectively. The study concluded that all measured radiological hazard indices were within the recommended safety limit values and do not pose major radiation hazard except at very lower depth (40ft) where elevated activity concentrations of Uranium and Thorium were observed.

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Conflicts of interest
There are no conflicts of interest.

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