Naturally Occurring Radioactive Concentrations and the Associated Cancer risks from Tailings by Kaolin Mining in Katsina, Nigeria

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Abstract

This research was carried out to assess the health hazards associated with environmental radioactivity around Kankara and Dutsinma Kaolin mining sites Katsina State; Nine (09) Kaolin samples were collected randomly from the mining sites for the study. The radioactivity analysis was done experimentally using NaI(Ti) gamma spectrometer at Center for energy research and training, Ahmadu Bello University Zaria. The health hazards were evaluated using numerous radiological, Statistical and USEPA models.

The hazard indices were found to be below the USEPA threshold limit for non-carcinogenic risks of 1.0 indicating that the exposed population ages are unlikely to experience any adverse non-carcinogenic risks. The radiological parameters obtained are 226Ra, 238U, 232Th and 40K of NORMs (Naturally Occurring Radioactive Materials) with activity concentration of 100.889, 0.129 and 224.040 (Bq-kg^{-1}) in the soil samples, at the studied area. Absorbed dose rate in air (D) measured in nGyh-1 at 1 meter above the ground, was calculated from the mean radioactivity concentrations and obtained as 77.575 while the total annual effective dose (AED) in mSv/y had average value of 0.099 in soil. The values obtained for external and internal radiological hazard indices Hext and Hint are 0.471 and 0.741 in soil; which were lower than the UNSCEAR recommended limit of unity. Finally, the overall excess lifetime cancer risk due to NORMs were 5.63E-06 and 4.15E-06 for adults and children respectively in whole population indicating that a maximum of 6 adults per 1 million may be affected while a maximum of 4 children per 1 million may be affected. These values were within the USEPA threshold of 1E-04.

Keywords: Norms, Gamma Spectrometer, Carcinogenic, Radiological, Hazard Indices

1. INTRODUCTION

Kaolin, is a mineral resource with significant deposit in Nigeria, it has been found very useful in several aspects of industrial, construction, agriculture and pharmaceutical activities. It has been known to be useful in plastic making, paper production, cosmetics and toothpastes. It is also a part of the constituents used in the production of ceramics, porcelain, cement and some medical items. In recent times, Kaolinite clay types have shown cost effectiveness when used as pozzolanic additives in cement and concrete [Aras et al., (2007; Yanguatin et al., (2017)]. In the agriculture sector, it has become reputable in the production of insect repellant and sunburn averting sprays for plant protections.

In Nigeria, mining of kaolin deposit is common without consideration for the heavy metal component and possible heightened background radiation present in them and the possible health risks posed to miners, residence of communities around mining sites, users of Kaolin products and the environment. Mining and processing of radionuclide bearing minerals can enhance the levels of radiation exposure to the workers and inhabitants in such locations (Adagunodo *et al*., 2019). While humans are exposed to radionuclides through ingestion and inhalation via internal exposure, irradiation by external gamma rays emitted from the radionuclide via external exposure is also a possibility. Furthermore, exposure to heavy metals, occurs as they enter into the human bodies via consumption of food, drinking water and air; subsequently bio-accumulating.

Mining has been identified as one of the potential sources of exposure to Naturally Occurring Radioactive Materials (NORMs) (UNSCEAR, 2000).

Elevated levels of radionuclides around mining sites, considered as Technological Enhanced Naturally Radioactive Materials (TENORM) have been reported by [Usikalu et al., (2018); Omeje et al., (2019) and Adagunodo et al., (2019)]. These studies have provided measurements of the naturally occurring radionuclides with the aim of determining the distribution of these radionuclides on the mining field and estimation of the gamma index risk exposure to the miners on the field (Kaniu *et al.*, 2018). There are two main contributors to natural radiation exposure, namely; high-energy cosmic ray particles incident on the earth's atmosphere and radioactive nuclides originating from the earth crust which are present everywhere in the environment, including the human body (UNSCEAR, 2000).

However, mining companies are often not regulated for heavy metals and for NORMs in most countries including Nigeria. In many cases, there are no clear implementable guidelines for their regulation by the Environmental and Radiation Protection agencies. With recent increase in

awareness of the potential exposure situations of NORMs. While the International Basic Safety Standards (BSS) for protection against ionizing radiation and the safety of radiation sources specify global basic requirements for the protection of health and the environment from ionizing radiation (IAEA, 1986), the BSS, based on the latest recommendations of the International Commission on Radiological Protection (ICRP, 2007) also provides a focus on regulation of practices and interventions and is applied to both natural and artificial sources of radiation in the environment and the consequences on living and non-living species.

In Nigeria, 800 million tons of quantified probable and proven deposits of the occurrence of kaolin, has been recorded in different parts of the country with specific abundant deposits in Anambra, Bauchi, Borno, Ekiti, Enugu, Kaduna, Katsina, Plateau, Ogun Ondo,Oyo and Sokoto states of up to (RMRDC, 2010). In the last two decades, interest in quantification of NORM in Kaolin samples has led to many studies by various researchers including [Atanda et al.,(2012); Talabi et al., (2012); Akinyemi et al., (2014) and Badmus & Olatinsu (2009)]. These studies adopted different methods and processes to assess the environmental and health implication of Kaolin mining and uses. Investigations are often carried out by different workers on separate sample for NORM in the same region while the spatial, particularly the depth, dimension are not considered in any of the study. In this present research effort, NORMs have been assessed from the same sample to determine the overall health and environmental impact of Kaolin mining from some mining location in Katsina. The study also includes a depth investigation to determine the relationship of the radionuclide with the depth of mining.

Mixture of Kaolin and sawdust in the ratio of 1:1 by volume has been often used in producing quality insulating fire bricks (Malu & Babson, 2007). Due to its optical suitability and such properties including whiteness, low viscosity, no abrasiveness, controlled particle sizes, flat hexagonal plates (Murray, 2010), chemical inertness and inexpensiveness compared to other minerals (Aderiye *et al.*, 2014), kaolin is largely used in the paper industry as a coating pigment and filler (Iyasara et al., 2014) for increasing brightness, opacity and giving the desired finish. It is also significantly used as a traditional raw material in the ceramic industry in the production of white ware, sanitary ware and household utensils; giving the ware a smooth surface finish, and electrical insulation (Benea & Gorea, 2004). As an extender pigment providing color or whiteness, kaolin is used in rubber and plastic industry; as protected for the temporary relief of real itching and disappear rash, it is used in the pharmaceutical industries; and as an insecticide against various arthropods that affect crops, it is used in agriculture. It is also a major component of many cosmetic product

including toothpastes. All these uses are characterized by very close human contact and interaction often without consideration for the radionuclide that may be present in the mined kaolin. These are apart from the fact that the mining process in itself may expose miners to heightened levels of radiation or the possibility of technological enhancement of NORM as a result of mining technology.

It is further noted that kaolin and other clay minerals contains quartz, long-term exposure to which may result in silicosis, lung cancer, chronic bronchitis and pulmonary emphysema in humans but found to be less toxic to aquatic organisms.

The radionuclide quantification of kaolin collected within Kankara and Dutsinma LGA of Katsina State will be carried out using NaI(Tl) Gamma Spectrometry respectively. The expected results from the study will provide the required activity concentration of the NORM present in Kaolin from the mining sites.

The study is sufficiently justified by the recent increased awareness and concern for the potential hazards associated NORMs in the environment which has resulted in implementation of new regulations dedicated to natural sources of radiation in national legislations (Faanu, 2011). In many developing countries including Nigeria, regulatory control has been based on radionuclide concentrations in raw materials residues, waste streams and public exposures are inadequate (Darko et al, 2005; Darko & Faanu, 2007). Consequently, there is the general lack of awareness and knowledge of the radiological hazards and exposure levels by legislators, regulators and operators. Additionally, the dust from the mining processes may consist of toxic heavy metals and natural radionuclides with varying concentrations that may further concentrate the natural existence of these elements and radionuclides in soil around the mine.

The study extends to cover all relevant three main primordial radionuclide ^{40}K, ^{232}Th and ^{238}U. The coverage also includes isotopic studies using NaI(Tl) Gamma Spectroscope whole NORMs has been considered. This study is limited to 9 samples each for isotopic studies taken from three mining sites across two local government areas of Katsina state.

2.0 MATERIALS AND METHOD

In this chapter, an overview of the materials, equipment and methods used for the research study is presented with emphasis placed on the technically required ingredients and processes to achieve the different objectives set for the study.

2.1 MATERIALS

Table 2.1 presents the material used for the different stages of the study and their function with respects to the different stages of the research effort.

Table 2.1: Stages of Research, Materials used and their Details and Functions

STAGE	MATERIAL	DETAILS/ FUNCTION
SAMPLE COLLECTION FROM MINES	Hoe	Digging Kaolin Sample from the earth crust
	Global Positioning System (GPS)	Locating point and taking coordinate at the sampling location
	Shovel	Collection of dug samples
	Polythene bag	Packaging collected sample at the collection point
	Plastic bucket	Packaging the whole sample at one point for easy movement
	Measuring Tape up-to 50ft (15m)	Measuring the depth of each sampling point
	Touch light	Providing illumination inside the dug mining well
SAMPLE PREPARATION FOR NORM	Electronic balance	For weighing the sample
	Crusher	For grinding into a powder
	Masking tape	Used for labeling the sample
	Rubber container	Cylindrical container of 7 by 7 diameters in dimension to suit detector geometry
	PVC Gum	Used for sealing the container to avoid escape of a Radon Gases
	Permanent Maker	Labelling the rubber containers containing the samples for identification
	Candle Wax	Used to supplement PVC Gum in sealing the container
	Vaseline	For filling and perfection sealing
NORM ANALYSIS	NaI(Tl) Detector	A detector used for quantifying radioactivity in samples
	MCA	Used to analyze the input signal from the detector

	Lead shield	Pre-World War Lead limiting detection of background radiation and noise
DATA ANALYIS	MS Office Excel	Used in statistical and theoretical analysis of acquired data

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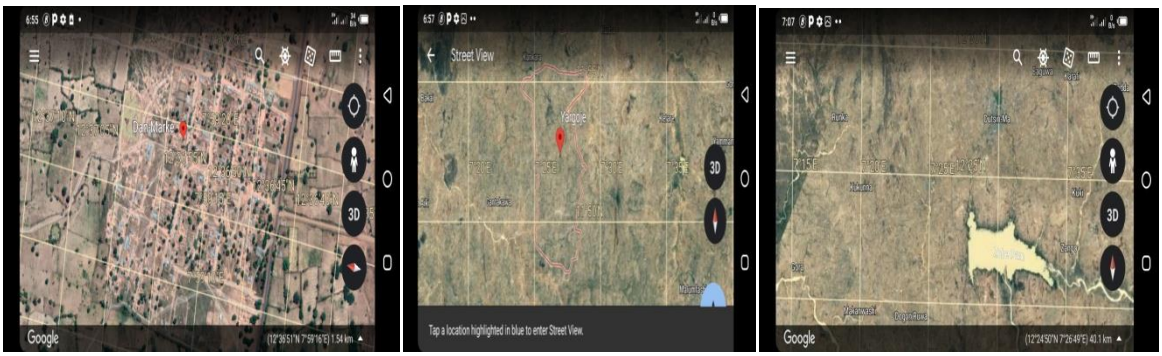
Other non-technical materials of no significant scientific consequence used in the research include; PC laptop computer, printer and paper for word processing and production of hard copies of the study reports.

2.2 METHOD

In this section the procedures for the study are capture to include procedure for sample collection, sample preparation for Gamma Spectroscopy, procedure for Na(TI) Gamma Spectrometry and the procedure for data analysis. The procedures are mostly captured as steps in chronological sequential order so as to present a clear idea of how the study were achieved in sequence with respect to achieving the overall aim of the study.

2.2.1. PROCEDURE FOR SAMPLE COLLECTION FROM MINE

The selection of the sampling locations was purposively carried out based on the accessibility to the public and proximity to the mine. The sampling strategy adopted for the samples collection was however random in line with ASTM (1983; 1986), IAEA (2004) and USEPA (1989). Nine (09) soil samples were collected from within the mining areas. The Nine Kaolin samples were collected from three different locations with three samples collected from each depth of 5, 15 and 25m in the mine well. The specific sample location is as indicated in Fig 1 from Sambisa-Danmarke and Dajin Gwamna-Yar'goje in Kankara Local Government Area and Farar Kasar Boto - Garfi/Haukan Zama in Dutsinma Local Government Area of Katsina State. All samples were labelled at the point of collection while their coordinates are read and recorded respectively using Global Positioning System (GPS). Fig. 2 presents also the map of Katsina state showing the two Local Government Areas where the samples were collected.



(a) (b) (c)

Fig.1(a), (b) and (c): GPS showing (a) First Sampling Point at Sambisa area of Danmarke Village; Kankara LGA (Elevated at an altitude of 168m (1978ft), Latitude $11^0$53’42’’N and Longitude $7^0$36’31’’E) (b) Second Sampling Point at DajinGwamna-Yar’goje Village of Kankara LGA (Elevated at an altitude of 1980ft (168m), Latitude $11^0$52’37’’N and Longitude $7^0$26’27’’E) (c) Third Sampling Point at FararKasar Boto-Garhi/Haukan Zama in Dutsinma LGA (Elevated at an Altitude of 51.2m (1102ft), latitude $12^0$24’15’’N and Longitude $7^0$26’46’’E).

Fig. 2: Map of Katsina, showing the Two Different Local Government of Dutsinma and Kankara, Katsina State in Relation to the Scope of the Study.

The nine samples were then re-bagged after removing foreign materials while drying and labeled, then set to the designated laboratory for NORMs Analysis using NaI (TI) detector. Each sample was

sealed in a polyethylene bag, firmly tied to avoid cross contamination before labelling. Nine samples were carried to the Radiation Biophysics laboratory at the Centre for Energy Research and Training, Ahmadu Bello University, Zaria for Gamma spectrometry.

It should be noted that for each sampling point, samples of about 1kg was collected and put in well labeled polythene bag consistent with procedure adopted in Kolo (2014) while the three samples per location were collected by stratified random sampling with the first sample at a depth of 5 m from the surface, the second sample at a depth of 15 m and lastly from the depth of 25 m considering the possibility of different weathering processes at different depth for minerals respectively.

2.2.2 SAMPLE PREPARATION FOR GAMMA SPECTROSCOPY

The samples were spread on cardboard sheets and all foreign materials were removed. They were then dried at Room temperature for about 24 hours and then grinded using a manual motor into fine powder and sieved using 2mm sieve. The homogenized samples were filled into 25g cylindrical plastic containers (7.0 cm diameter by 7.0 cm height) which were hermetically sealed with the aid of PVC tape, Vaseline and candle wax to prevent the escape of airborne ^{222}Rn and ^{220}Rn from the samples. The dimensions of the plastic containers were chosen in such a way that it suited the optimal soil mass of 350g for analysis of bulk samples. The samples were sealed and stored for 40 days to allow secular equilibrium to be reached between^{232}Th and ^{238}U and their progeny and radon and its daughters. The IAEA reference materials for gamma spectrometry (RGK-1, IAEA-448 and RGTh-1) were prepared in the same manner as the samples. At secular equilibrium, it is expected that the activity of each radionuclide in a given series is equal to the activity of the nuclide that heads the series, thus the parent nuclide.

2.2.3 PROCEDURE FOR NaI (Tl) GAMMA SPECTROMETRY

Firstly, the detector was calibrated for both energy and efficiency consideration with the prepared IAEA reference materials RGK-1, IAEA-448 and RGTh-1 for the quantitative determination of ^{40}K, ^{238}U and ^{232}Th respectively in the soil samples.

Each of the samples were then measured separately for more than eight (8.33) hours counting period before obtaining the result. The samples were analyzed using NaI (TI) detector situated at Radiation Biophysics Low Background Laboratory of the Center for Energy Research and Training, Ahmadu Bello University, Zaria. The detector has a 6cm thick lead shield, cadmium lined assembly with copper sheets for the detection of background radiation. The detector has pulse resolving time of about 0.25s, an incorporated preamplifier and a 1kV external source which permits its use for high

counting rates. The detector was coupled to a computer based multichannel analyzer ***(Maestro)*** program from ORTEC=-1656 for the acquisition and analysis of the gamma spectra.

Each of the prepared samples was counted for 30,000seconds (8.33 hrs) in the outlined detector geometry in order to mitigate the influence of background radiation from radioactive contaminants within the shielding materials of the detector assembly. The obtained data in counts per second were converted to conventional units of Bq/kg using calibration factors to determine the activity concentration of ^{226}Ra (^{238}U), ^{232}Th and ^{40}K.

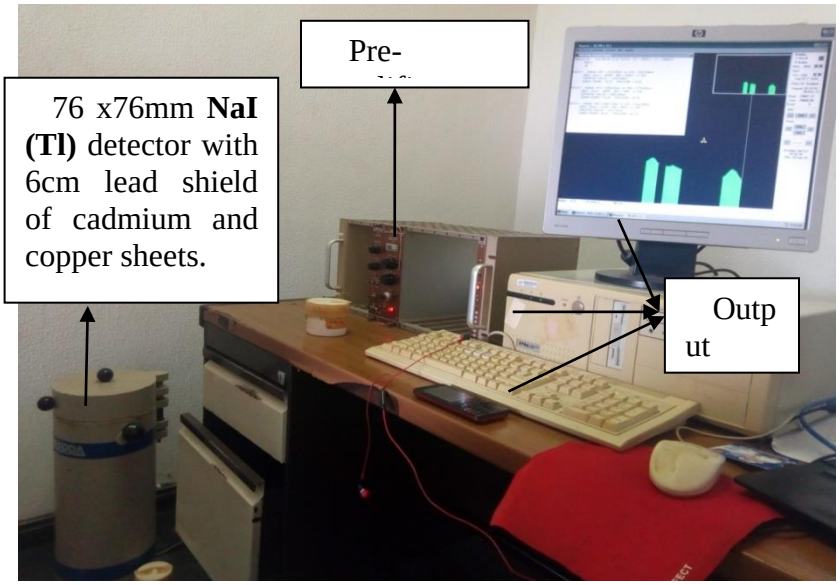


Fig 3: The NaI(Tl) Gamma Spectrometry Set Up at the Radiation Biophysics Laboratory, CERT, ABU Zaria

2.2.4 PROCEDURE FOR DATA ANALYSIS

MS Office Excel was used as the principal data analysis tool for the study based on its accessibility, simplicity and the availability of sufficient hands-on guide for the coding necessary for all the calculation and data analysis of interest in the quantification of NORM, determination of their hazard parameter and indices and in the comparative analysis.

MS Office Excel was also employed in the graphical representation of data and comparative analysis carried out in the study.

3.0 RESULTS AND DISCUSSION

The Activity Concentration of the NORMs determined using NAI(Tl) gamma spectroscopic counts and discussed in relevant health and hazard indices with depth dependence.

3.1 RESULT OF ACTIVITY CONCENTRATION OF NORM FROM NAI(Tl) GAMMA SPECTROMETRY

The activity concentration for ^{238}U, ^{232}Th and ^{40}K has been determined from gamma spectrometry measurement using NaI(Tl) set up. The measured Activity Concentration (NC_i), determined in Count Per Seconds (CPS) and converted to Activity Concentration in $BqKg^{-1}$ (At) was done using the semi empirical equation 4.1for ^{238}U, ^{232}Th and ^{40}K.

Sample Activity (A) in $Bqkg^{-1}$ = $\frac{NC_i}{\varepsilon \, X \, y_i \, X \, T \, X \, M} = \frac{NC_i}{M} \, x \, \frac{1}{CVF} \, x \, CF$ (3.1)

Here: NC_i = Net peak after subtraction of background of the gamma ray line at energy E.

$\boldsymbol{\varepsilon}$ = Detector efficiency of gamma ray line at photo peak energy E.

$\mathbf{Y_i}$ = emission probability of the gamma ray photons of energy, E under consideration.

T = Time of measurement in seconds.

M = Mass of sample in g.

CF = Calibration Factor = 10^{-4}

CVF = Conversion Factor uniquely determined for ^{238}U, ^{232}Th and ^{40}K and presented in Table 3.1 below.

Table 3.1: Calibration factor for NORM at CERT NaI(Tl) Facility

NORM TYPE	CALIBRATION FACTOR (CF)
$^{238}U/^{226}Ra$	0.00208600
^{232}Th	0.00211942
^{40}K	0.00155400

Also, the Mass in g for the 9 samples measured are reported in Table 3.2 where Wc (g) is the Weight of empty container in grams, Wc+s (g) is the Weight of container filled with sample in grams and Ws (g) is the actual weight of sample determined using Equation (3.2)

$$Ws = Wc+s - Wc \quad (3.2)$$

Table 3.2: Mass of Sample

SAMPLE ID	Wc (g)	Wc+s (g)	Ws (g)
Y01	26.26	281.11	254.85
Y02	31.57	229.73	198.16
Y03	28.21	282.22	254.01
S01	22.06	291.46	269.40
S02	44.59	272.47	227.88

S03	36.19	287.14	250.95
G01	33.62	300.61	266.99
G02	26.71	258.80	232.09
G03	32.53	265.60	233.07

The Activity Concentration A, for the nine Kaolin Samples, as seen in the Appendix, ranged from $133.31 \pm 1.88 \geq A_{235U} \geq 81.50 \pm 2.72$ Bq/Kg for ^{235}U while for ^{232}Th the values ranged as $83.87 \pm 2.01 \geq A_{232Th} \geq 23.79 \pm 3.07$Bq/Kg and for ^{40}K it ranged as$1.53 \pm 0.02 \geq A_{40K} \geq 1.16 \pm 0.02$ Bq/Kg.

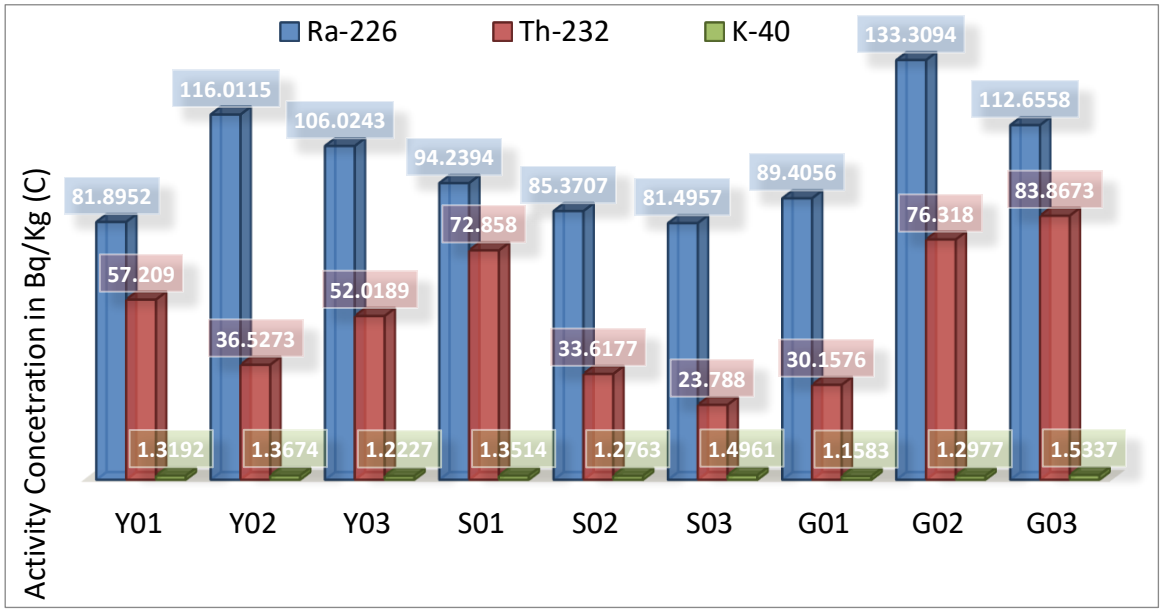


Figure 4: Activity Concentration of Samples of Mined Kaolin

Figure 4: presents the calculated Activity Concentration (C) for ^{238}U, ^{232}Th and ^{40}K for the 9 samples. Equation (3.1) with Table 3.1 and Table 3.2 have been used for the determination of data for the figure 4.

From Figure 4: it can be clearly observed that the Activity Concentration or Specific Activity A for ^{235}U is significantly the most important to be considered for the Kaolin samples with a value of A_{Mean} [^{235}U] ± A_{SD} of 100.05 ± 16.97 Bq/Kg. The maximum value of the Activity Concentration measured, $A_{Max,}$[^{235}U] as can be seen in Figure 4 is 133.31 Bq/Kg from the Garfi mine at depth of 15.0 meters while the minimum A_{Min} [^{235}U] is 81.45 Bq/Kg from the Sambisa mine at depth of 25.0 Meters. The distribution of the measurements around the mean is determined using the Standard Error (SE) and found to be SE [^{235}U] = 1.89, a value that indicate moderate uniform distribution of ^{235}U across the nine different mines.

Furthermore, it is observed that *A* for ^{232}Th follows ^{235}U in significance in its importance as NORM in the Kaolin samples with a value of A_{Mean} [^{232}Th] ± A_{SD} of 51.82 ± 20.87 Bq/Kg. The maximum value of the Activity Concentration measured, A_{Max} [^{232}Th], as can be seen in Figure 4 is 83.87Bq/Kg from the Garfi mine at depth of 25.0 Meters while the minimum A_{Min} [^{232}Th] is 23.79Bq/Kg from the Sambisa mine at depth of 25.0 Meters. The distribution of the measurements around the mean is determined using the Standard Error (SE) and found to be SE [^{233}Th] = 2.32 a value that indicates a limited uniform distribution of ^{232}Th across the nine different mines.

Also, A for ^{40}K provides the least significance in its importance as NORM in the Kaolin samples with a value of A_{Mean} [^{40}K] ± A_{SD} of 1.34 ± 0.11Bq/Kg. A_{Max} [^{40}K], as can be seen in Figure 4 is 1.54Bq/Kg from the Garfi mine at depth of 25.0 Meters, while A_{Min} [^{40}K] is 1.16Bq/Kg from the Garfi mine at depth of 5.0 Meters. The distribution of the measurements around the mean is determined using the Standard Error (SE) and found to be SE [^{40}K] = 0.01 a value that indicates a very high uniformity of the distribution of ^{40}K across the nine different mines studied.

The analysis of the activity concentration contribution of norms with respect to locations and depth can be further highlighted with Figure 5 which presents the contribution of NORM as a function of the combination of both location and depth.

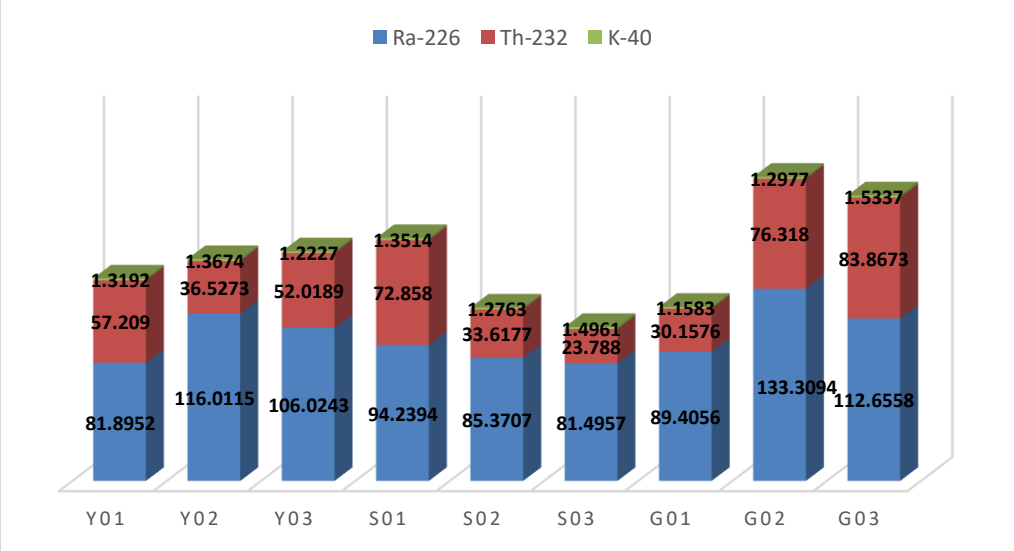


Figure 5: Contribution of NORM With Respect to Location and Depth

From the bar chart, the five most significant contributions to the measured activity concentration are noted as:

A. ^{238}U from Garfi mine at a depth of 15m, from Yargoje mine at a depth of 15m, from Garfi mine at a depth of 25m, from Yargoje mine at a depth of 25m and from Sambisa mine at a depth of 5m.

B. ^{232}Th from Garfi mine at a depth of 25meters, from Garfi mine at a depth of 15m, form Sambisa mine at depth of 15m, from Yargoje at a depth of 5m and from Yargoje at a depth of 5m.

C. ^{40}K from Garfi mine at a depth of 25m, Sambisa mine at a depth of 25m, Yargoje mine at depth of 15m, Sambisa mine at a depth of 5 m and Yargoje mine at a depth of 5m.

The distribution of five most significant NORM Activity Concentrations indicates distinction in the radionuclidic features of the different locations. This distribution may be indicative of geological or meteorological factors. Anthropological factors as causes for the distinction in distribution are ruled out because of the similarity of the human activities taking face in all the 3 locations which started about the same time. Wide variation in Activity Concentrations has been noted to be a measure of the spatial distribution due to anthropogenic activities, soil types and other natural factors of the radionuclides in different study samples and also indicative of the influence of physical and geo-chemical processes on the accumulation of radionuclides (Isinkaye & Emelue, 2015). Also, the high Activity Concentration of ^{238}Uin most of the samples in the present study maybe due to the solubility and mobility of Uranyl from $^{226}Ra/^{238}U$ chain while that of ^{232}Thmay be as a result of its extremely slow decay rate due to its very long half-life of 14 Billion years (UNSCEAR, 2000). For ^{40}K, the very low Activity Concentration may be due to low content of monazite in Kaolin mining area (Orgun et al., 2007). It should be noted that, monazite, a primarily reddish-brown phosphate mineral containing rare-earth elements, is indicative of radioactive elements such as ^{40}K and of higher natural radioactivity (Singh, Shanker, Neelakandan & Singh 2007).

Meanwhile, Garfi mine appears to provide the most significant indicator for both ^{238}U and ^{232}Th followed by Yargoje mine and least indicator provided by the Sambisa mine. Meanwhile, the most significant indicator for ^{40}K appears to be provided by Sambisa mine followed closely by Yargoje mine with the least indicator from Garfi mine despite recording the highest value of Activity Concentration at 25m depth.

This has implication for planning mining site specific radiation protection arrangement with respect to ensuring As Low as Reasonably Achievable (ALARA) exposure to radiation. While the limit of exposure of worker in the different site will be influenced by standard limits, site specific

arrangement that may reduce actual exposure with respect to time and distance of operation may be required.

Furthermore, these distinctions in distribution may also imply the need for unique approaches for each site with respect to the domestic, cosmetic and medical uses of the mined Kaolin.

In Figure 6, the graphical plot of the depth dependence of the Activity Concentration from the different mines is presented from 5m to 25m. The study presumes a uniform depth distribution of the samples within a 10m range such that measurement at 5m represent 0 – 10m depth, at 15m represent 10 – 20m depth and 25m represent 20 – 30m range it should be noted that the depth of the noted depth of the 3 mine locations are within the range of 30 ± 2m.

From Figure 6, it can be observed that there is no uniform depth distribution of Activity Concentration form $^{226}Ra/^{238}U$, ^{232}Th and ^{40}K. This implies that the depth of mining does not determine the quantity of NORMs present in mined Kaolin sample. From this study, it can be inferred that the radiation safety and processing requirement for domestic, medical, cosmetic and construction uses of mined Kaolin may not be a function of the depth of mining. As such, specific sample from different depths will have to be treated uniquely even when they are form the same mining site.

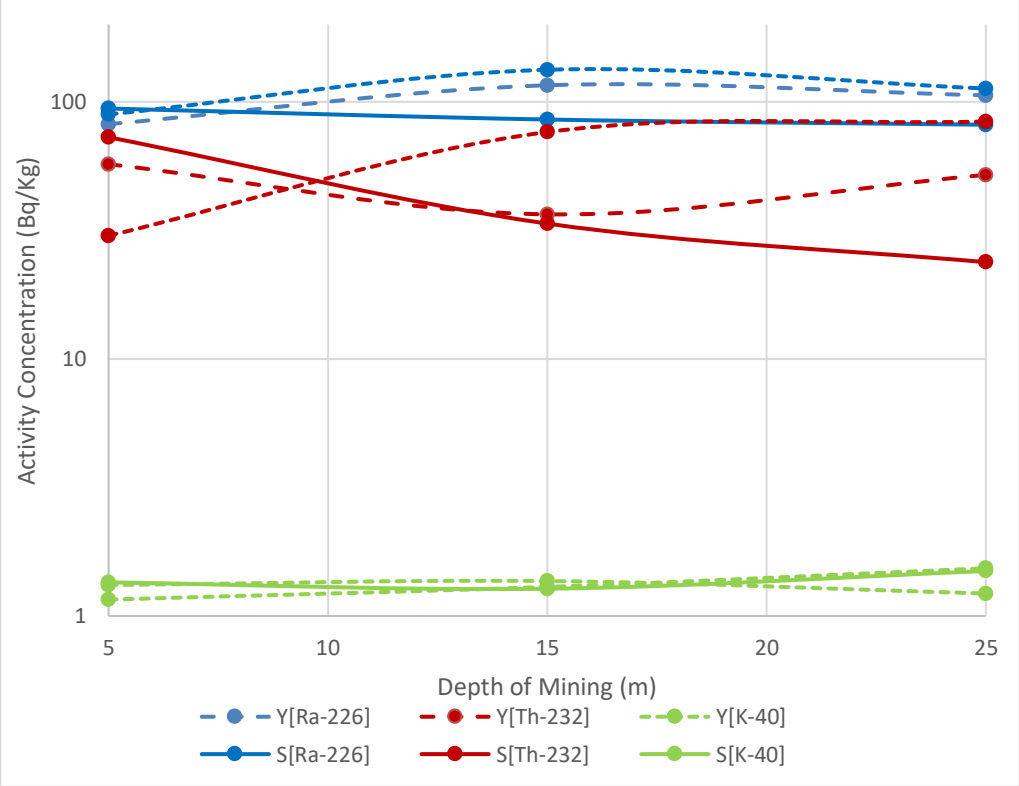


Figure 6: Activity Concentration as a Function of Depth of Mining

While studies have shown downward migration rates of fallout radionuclide in soil layers as estimated from depth profiles and historical fallout patterns of the radionuclides using a least-square fitting method. Migration rates of some radionuclides were found to be 0.1-0.6 $cmyr^{-1}$ for each layer up to 13cm deep Iyogi et al., (2004); the present study provides no data from which such inference can be made and reveal no indication a constant direct or inverse dependence of the Activity Concentration of radionuclide with depth of mining.

3.2 DETERMINATION OF HEALTH HAZARD PARAMETERS FROM NORM

Health hazard parameters of consequences resulting from the measurement of NORM from mined Kaolin have been determined for each of the mining location and depths. The parameters determined includes the Absorbed Dose Rate (D_R), Radium Equivalent (Ra_{eq}) External Hazard Index (H_{ext}), Internal Hazard Index (H_{int}), Outdoor Annual Effective Dose Equivalent($AEDE_O$), Indoor Annual Effective Dose Equivalent ($AEDE_I$),Total Annual Effective Dose Equivalnent, Annual Gonadal Dose Equivalent (AGDE), Gamma Level Index (Iγ), Activity Utilization Index (AUI), Exposure Rate (ER), the Excess Lifetime Cancer Risk (ELCR).

Furthermore, analysis of the probable health impact of the various parameters is presented based on the global limit references considering safety standards.

A. ABSORBED DOSE RATE

Figure 7 present the absorbed dose rate determined for the 9 mine Kaolin samples plotted with the recommended limit of 84.00 $nGyhr^{-1}$ as standard above which the absorbed dose rate value is considered excessive. As it has been reported for sediment, the contribution of natural radionuclides to the absorbed dose rates depends on the concentrations of various radionuclides in the mined sample (Erenturk et al, 2014).

The range of the results of the calculated absorbed dose rate in mined Kaolin samples is given as 52.09 ± 3.85 – 107.74±74 $nGyhr^{-1}$ with a mean value of 77.57 ± 18.18 $nGyhr^{-1}$. The average absorbed dose rate in this study is higher than the worldwide average value of 84.00 $nGyhr^{-1}$ reported by UNSCEAR (2000).

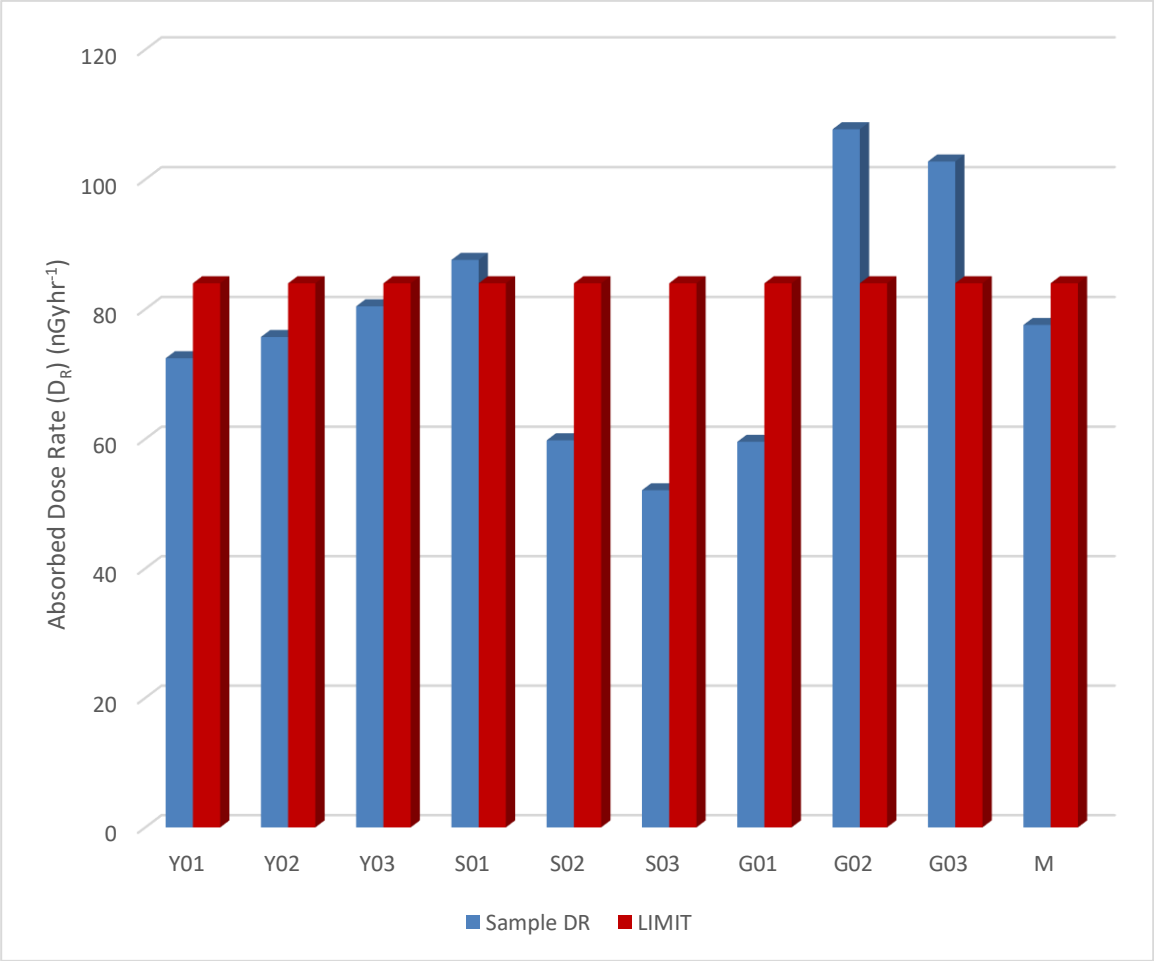


Figure 7: Absorbed Dose Rate for the Different Samples Compared to Standard Limits

From Figure 7, Absorbed Dose Rate (D_R) for three namely Garfi mine at a depth of 15m, Garfi mine at a depth of 25m and Sambisa mine at a depth of 5m are observed to exceed the recommended UNSCEAR limit while the average of the study is 0.92 times less than the recommended limit.

It can be concluded that, while, generally, the energy deposited locally in any absorbing media such as the human body per hour by Kaolin from the selected mining site is below the recommended limit, specific concern should be exercised for Garfi and Sambisa mines especially at depth below 10 m. This could be includes the use of enhanced radiation protection gear and reduction of the extent of time spent at depth below 10m in the mines.

B. RADUIM EQUIVALENT ACTIVITY AND EXTERNAL AND INTERNAL HAZARD INDICES

Radium Equivalent Activity (Ra_{eq}) is determined within the context the activity concentration of a radionuclide equivalent to 370Bq kg^{-1} of ^{226}Ra, which gives outdoors an external effective dose rate

of 1.5mGy (1mSv) per year. It is often used as a relative measure of the gamma-ray exposure rates and therefore external exposure risk associated with ^{226}Ra vs. ^{232}Th vs. ^{40}K.

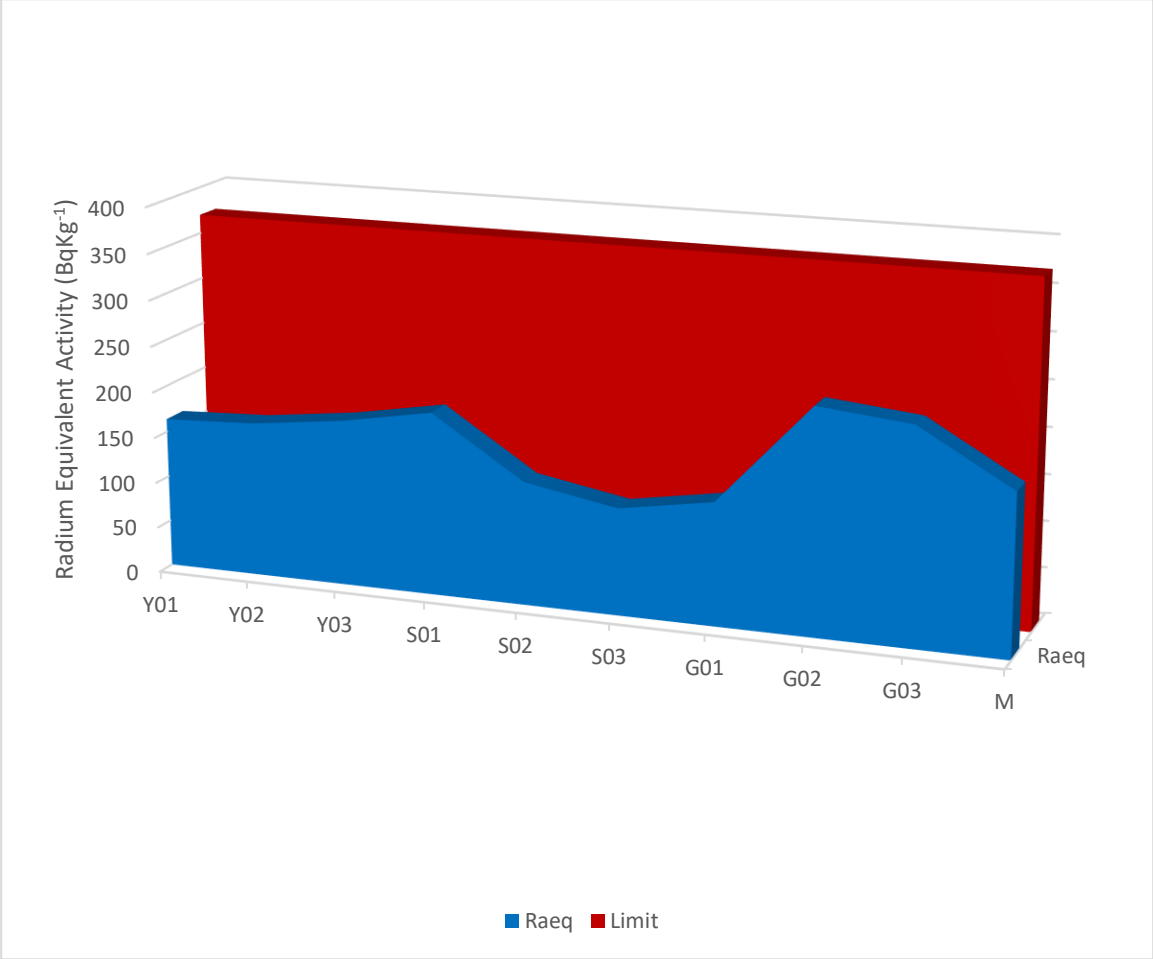


Figure 8: Radium Equivalent Activity for the Different Samples Compared to Standard Limits

In Figure 8, Ra_{eq} in $BqKg^{-1}$is presented for the nine Kaolin samples studied from the three mining sites. The Range of the determined Ra_{eq} value for the study is given as 115±8.70 – 242.54±5.90$BqKg^{-1}$ with a mean of 174.25 ± 41.34$BqKg^{-1}$.

These values reported are all below the recommended limits of 370$BqKg^{-1}$ and it is noted that Ra_{eq} for the nine samples are within the acceptable limit of less than 370$BqKg^{-1}$ as recommended by UNSCAER (2010). The mean value is observed to be 0.47 times less than the recommended limit.

It can thus be concluded that; there is no significant risk of hazard when the samples are used for construction purposes or found in building materials with the attendant possibility of respiratory tract doses to the inhabitants of the structure due to the external gamma dose and the internal alpha dose from Radon and its progeny through the decay of Radium. This is based on the assumption of an

annual exposure limit of 1mSvy^{-1} for which 370 BqKg^{-1} of ^{226}Ra, 259BqKg^{-1} of ^{232}Th, and 4810BqKg^{-1} of ^{40}K all produce the same gamma-ray dose rate (Beretka and Matthew 1985).

Furthermore, the related concept of Internal and External Hazard Indices, H_{int} and H_{ext}, have been calculated to approximate the relative risk of Radium vs. Thorium vs. Potassium from inhalation of alpha particles emitted from the short-lived radionuclides of Radon as it applies to the safe use of Kaolin as construction and building materials for dwellings (Righi and Bruzzi 2006).

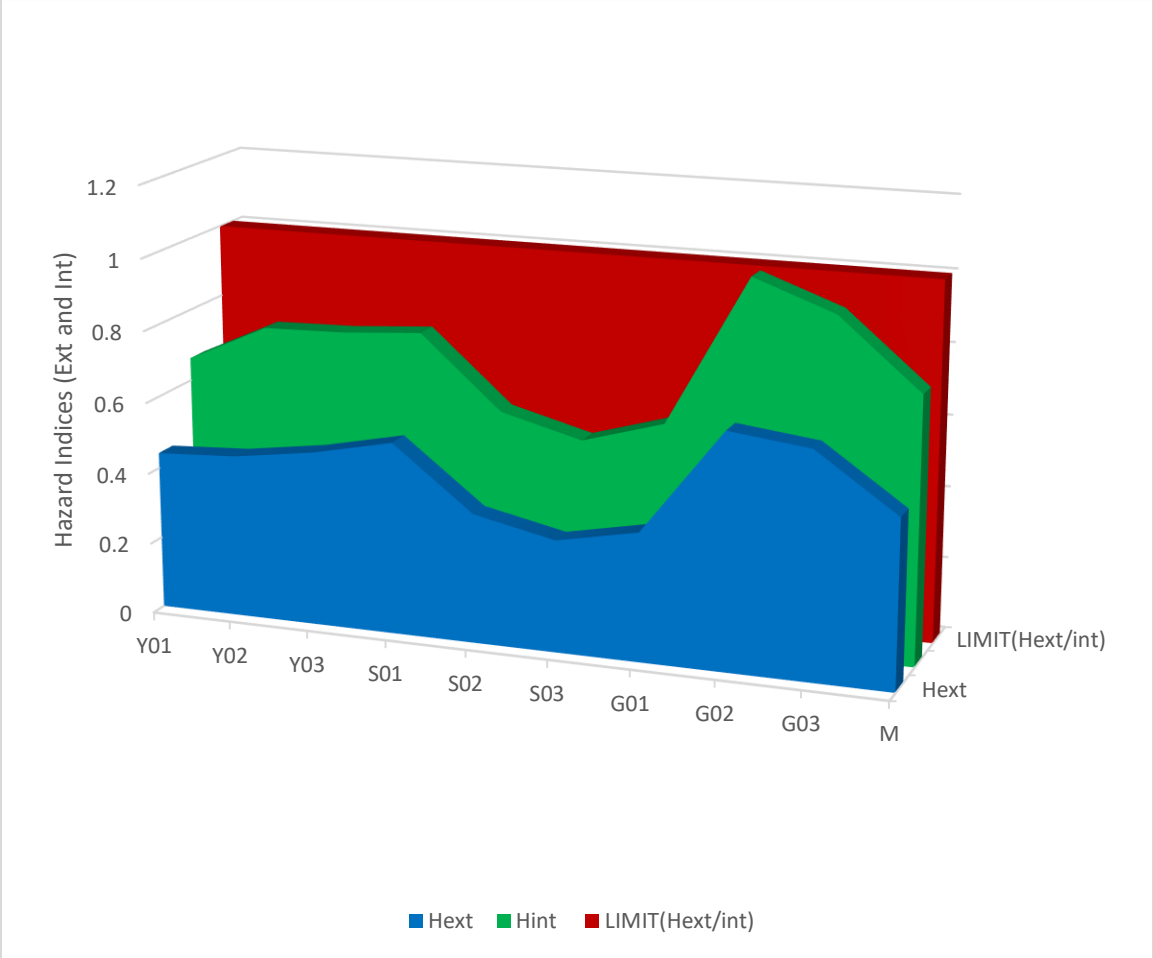


Figure 9: External and Internal Hazard Indices for the Different Samples Compared to Standard Limits

Generally, however, since the mean for the H_{ext}t is 0.47 times less and that of H_{int} is 0.53 time less than recommended limits, Kaolin cannot be classified as significantly contribution to both the external and internal exposure to carcinogenic radon and its short-lived progeny.

C. ANNUAL DOSE EQUIVALENTS

The Annual Dose Equivalents from Kaolin from Kanakara and Dutsima LGA of Katsina State has been assessed by determining the Outdoor Annual Effective Dose Equivalent ($AEDE_o$), Indoor Annual Effective Dose Equivalent ($AEDE_i$), Total Annual Effective Dose Equivalent ($AEDE_t$), and the Annual Gonadal Dose Equivalent (AGDE).

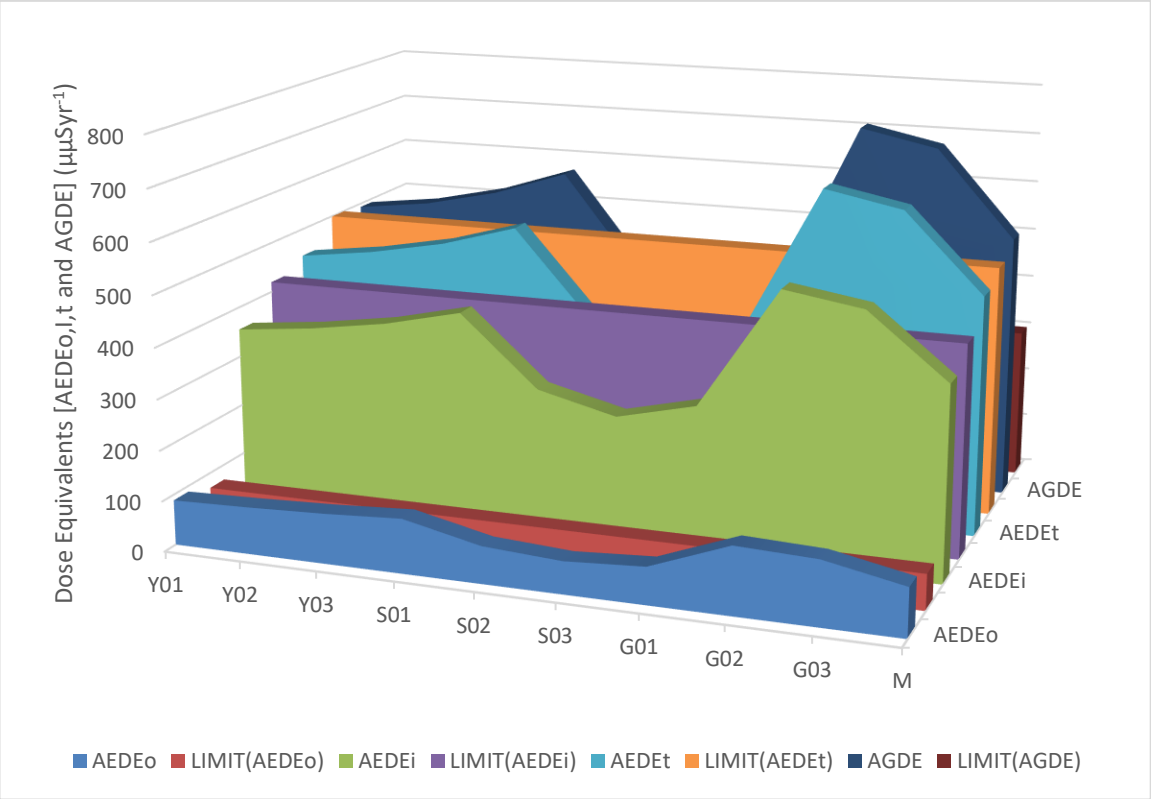


Figure 10: Annual Dose Equivalents for the Different Samples Compared to Standard Limits

The $AEDE_o$ and $AEDE_i$ are indices which measure the risk of stochastic and deterministic effects on individuals exposed to radiation as a result of outdoor and indoor irradiation sources respectively. The $AEDE_t$ is the sum of the contribution of both the outdoor and indoor measures and gives an overall assessment of the risk on individuals exposed indoor or outdoor. Also, the AGDE provides a measure of the risk to the gonads, the active bone marrow and the bone surface cells which are considered organs which experiences significant consequences when exposed to radiation beyond acceptable limits.

From the study, the range of value determined for the $AEDE_o$ is $63.92 \pm 4.72 - 132.22 \mu Svyr^{-1}$ with mean of $95.20 \pm 22.31 \mu Svyr^{-1}$ which is 1.38 times higher when compared to the global average of

70.00μSvyr^{-1}. The range of $AEDE_i$ is 255.66 ± 18.88 – 528.89 ± 12.65μSvyr^{-1}with mean of 380.81 ± 89.22μSvyr^{-1} which is multiply by1.08 less when compared with the world average of 410.00μSvyr^{-1}. Also, $AEDE_t$ which is the sum of $AEDE_o$ and $AEDE_i$ is within the range 319.58 ± 23.60 – 661.11 ± 15.82μSvyr^{-1}with mean of 476.14 ± 111.53μSvyr^{-1} which is 1.09 times less when compared to the global average of 520.00μSvyr^{-1}and 2.10 times less than the criterion limit of 1000μSvyr^{-1}.

Furthermore, for the AGDE, the range of value determined is 351.73 ± 26.10 – 132.22μSvyr^{-1}with mean of 521.16 ± 139.70μSvyr^{-1} which is 1.74 times higher when compared to the global average of 300.00μSvyr^{-1}.

Also, Gamma Utilization Index, Activity Utilization Index, Exposure rate and the Excess Lifetime Cancer Risk have been evaluated and presented in Figure 4.10, 4.11 and 4.12 all indicating need for enhanced radiation protection from Kaolin

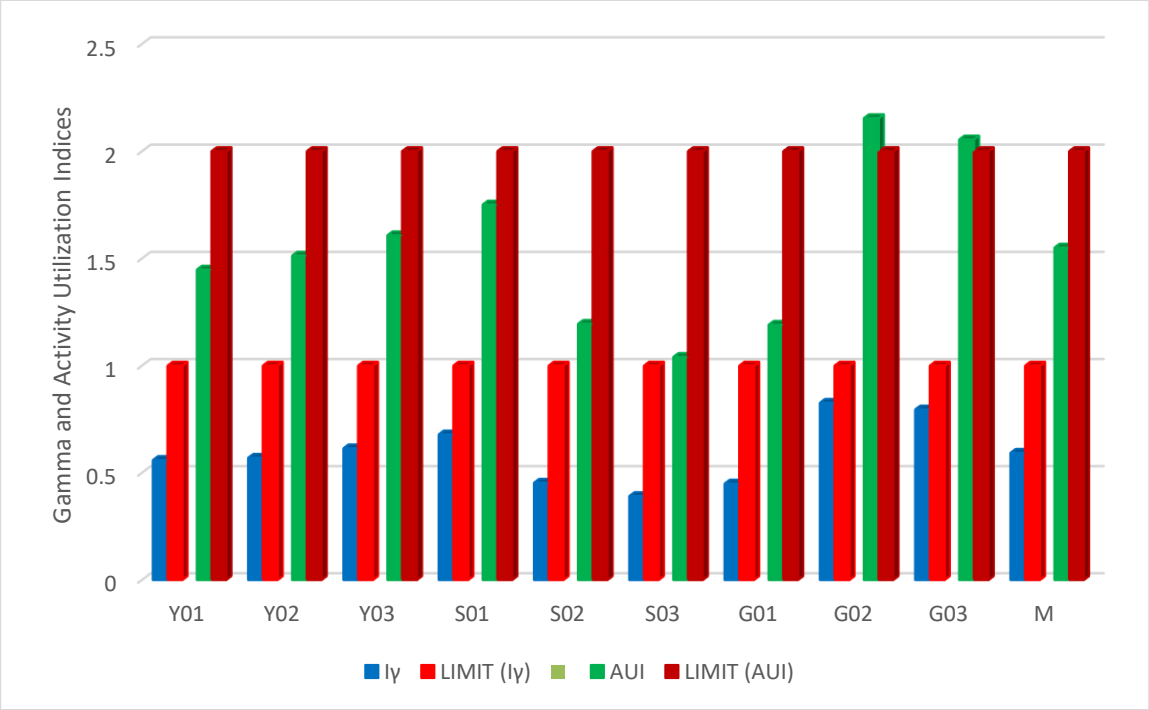


Figure 11: Gamma and Activity Utilization Indices for the Different Samples Compared to Standard Limits

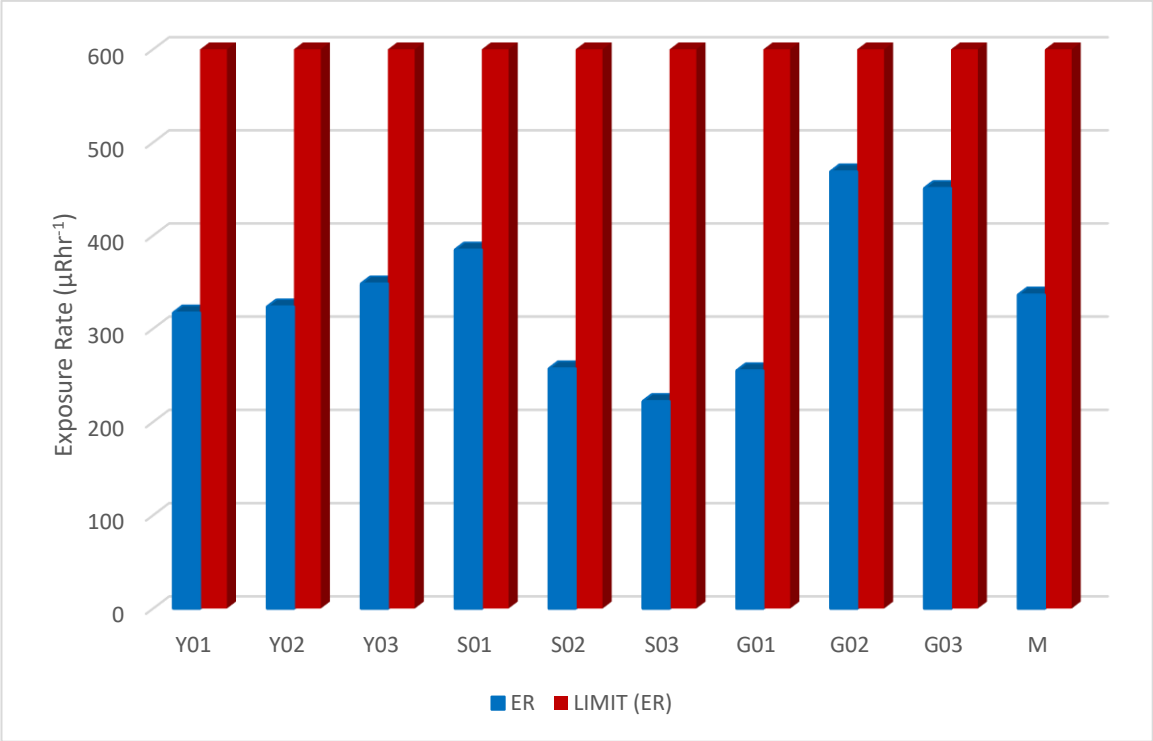


Figure 12: Exposure Rate of the Different Samples Compared to Standard Limits

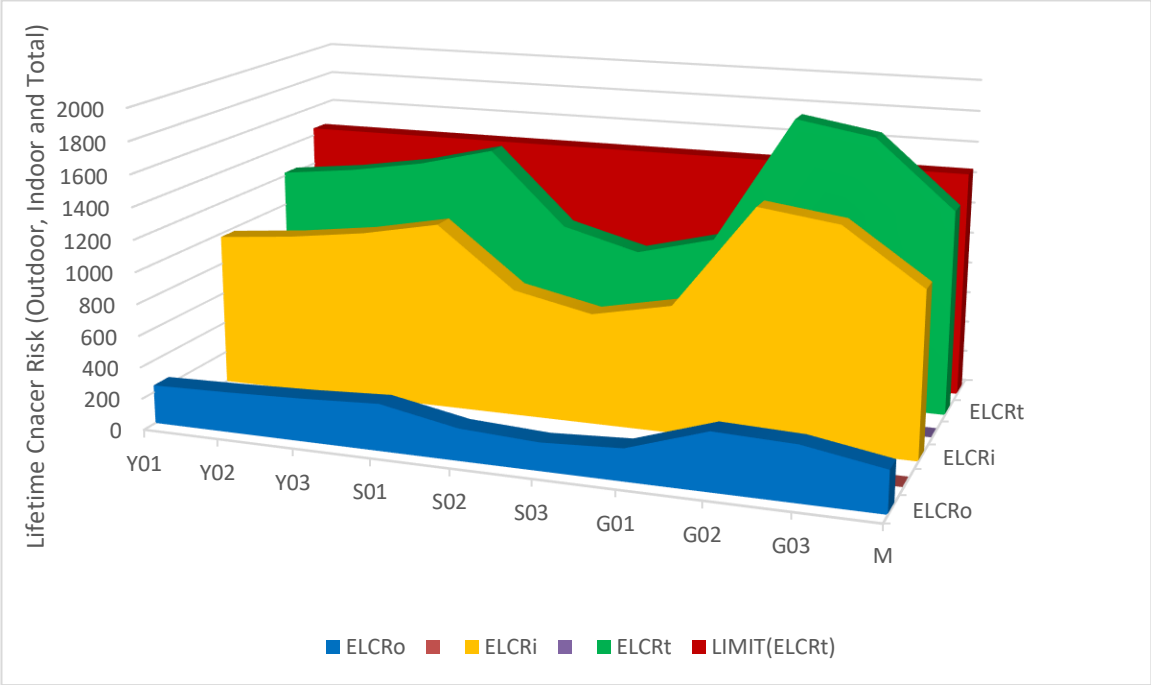


Figure 13: Excess Lifetime Cancer Risk for the Different Samples Compared to Standard Limits

4.0 SUMMARY

This research summarized the major findings and presents the results obtained. It also focused on the measurement of the NORM in mined Kaolin for two LGA in Katsina State named Kankara and Dutsinma. Nine samples were collected and analyzed using NaI(Tl) Gamma Spectrometry methods.

5.0 CONCLUSION

The main objectives of the research is to determine the activity concentration of relevant NORMs in Kaolin extracted from Kankara and Dutsinma Mining sites in order to quantify the actual Concentration of ^{238}U via the ^{226}Ra, ^{232}Th and ^{40}K.

The Internal and External Hazard indices obtained is within the range of 0.31 ± 0.02 to 0.66 ± 0.02 for the external Hazard indices (H_{ext}) while for Internal Hazard Indices ranges from (H_{int}) 0.53 ± 0.04 to 1.02 ± 0.02 with the average mean of 0.47 ± 0.02 and 0.74 ± 0.03. The average value obtained for both are less than unity, but a single value from H_{int} is above the range as it is greater than Unity value at Garfi mining site which is exactly at the depth of 15 m. It shows that, a serious measure is needed on the workers as well as the dwellers within since the limited exposure is above the recommended value of the sites.

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