Assessment of Solid Mineral to Soil Radioactivity Contamination Index in Selected Mining Sites and their Radiological Risk Indices to the Public

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Assessment of Solid Mineral to Soil Radioactivity Contamination Index in selected Mining Sites and their Radiological Risk Indices to the Public

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Abstract

This study examined the radioactivity levels of soil samples within selected solid mining sites in Nigeria using high purity germanium (HpGe) detector. Sixty soil samples in all were collected from the ten solid mineral mining sites investigated and six samples were collected as control samples from non-mining environment for analyses. The results of the activity concentration values obtained for ^{40}K, ^{226}Ra and ^{232}Th are 100.22 Bq kg^{-1}, 33.15 Bq kg^{-1} and 77.31 Bq kg^{-1} respectively. The ^{226}Ra and ^{40}K activities were found to be within the United Nation Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) acceptable permissible limit, but the ^{232}Th mean value was above the permissible limit of 30 Bq kg^{-1} for the public. In comparison, ^{40}K, ^{226}Ra and ^{232}Th soil samples mean activity concentrations were higher than the control soil samples values by 48.6%, 43.7% and 62.3% respectively. The results of estimated radiation hazard indices indicate average values of 150.72 Bq kg^{-1}, 68.40ηGyh^{-1}, 83.65μSvy^{-1} and 454.70μSvy^{-1} for the Radium Equivalent (Ra_{eq}), Absorbed Dose Rate (D), Dose Equivalent (AEDE) and Annual Gonadal Equivalent Dose (AGED) respectively. The mean values for External Hazard Indices (Hex, Hin), Representative Gamma index (I_γ) and Excess Life Cancer Risk (ELCR) were 0.41, 0.50, 1.06 and 0.29 x10^{-3} respectively. The statistical analysis shows positive skewness.

Keywords: Radioactivity, Mineral, Soil, Percentage contribution, Mining area**.**

1.0 Introduction

Assessment of background radioactivity level plays a significant role in the protection of man from excessive radiation exposure *(Abodunrin et al., 2017).* Natural background radiation levels are likely to vary with human activities and natural processes, it may also change with locations due to different mineralogical, deformational and climatic factors responsible for the syngenetic processes for mineral formation. Although natural background radiation level is time dependent, it does not depend on any constant level, because it is terrestrial and cosmic induced *(Ahmed et al., 2020).* Radioactivity levels are evaluated as part of national and international survey at different areas and countries of the world, for radiation protection (*Mane et al. 2014; Ugbede, 2020).*

Naturally occurring radionuclide materials (Norms) are inherent in many geologic materials and consequently encountered during geologically related activities. Since radioactive materials are prevalent in many minerals and soil formation and in the water that meets them, extraction and processing of these mineral resources that emanate from these sources exposes and raises the concentration of naturally occurring radionuclide in the environment *(Avwiri, et al., 2012)*. Exposure to high radiation level causes a wide range of health problems such as cancer of the lung, bone and skin, kidney ailments and blood infections *(Kessaratikoon, et al., 2013; UNSCEAR, 2016)*. Other problems associated with high exposure to ionizing radiations to health include; alteration in the structure and functions of the cells and organs, deterministic effect, stochastic effects, irritations, sensitization, embryonic effects, etc. The knowledge of radionuclide distribution in the environment is therefore of immense benefits in assessing the effects of radiation exposure, thus, monitoring of radioactive materials are of primary importance to man and for the protection of the environmental *(Avwiri et al., 2012; Emelue et al., 2014: Dolchinkov and Nichev, 2017)*.

Most mineral deposits are associated with radionuclide like uranium, thorium and its' progenies. Primary uranium ore minerals when weathered, oxidized or decompose and form secondary uranium minerals which on interaction with groundwater drift and contaminate the soil, water and aquatic bodies, even some distance away from the original source *(Aliyu et al., 2015)*. They are also found in conglomerates, shale, limestone, sediment and hydrocarbon *(Xhixha et al., 2015)*. Because minerals are found in within a host rock, there is the tendency that the immediate soil when weathered these minerals are found content an appreciable amount of radionuclides found in these minerals due to radioactivity transfer. Soil radioactivity concentration is one of the main determinants of the natural occurring radiation *(Agbalagba et al., 2012; Ugbede, 2020)*. Measurement of the radioactivity level of some rock samples, potential sources rocks of hydrocarbons have been conducted in different parts of the world *(Silo et al., 2013; Guidotti et al., 2015)*. They reported that the radioactivity concentrations of the radionuclides from the eastern region were generally low compared to that of the other regions, but Th-232 was identified as the major contributor of the dose that can be received from the environment.

When minerals are disintegrated through either natural or anthropogenic processes, radionuclides are liberated into the soil by rain infiltration and percolation processes *(Taskin et al., 2009)*. It has been established from previous studies that some of these soil and minerals such as monazite, pyrochlore and xenotime, which are obtained as byproducts of tin mining

are radioactive, *(Eroglu and Kabadayi 2013, Ekeocha, 2016; Kritsananuwet et al., 2015; Omotehinse and Ako, 2019)*. Exposure to radiations emitted by some of these radioactive minerals is a major source of health hazards *(Charro et al., 2013; Todorovic 2015)*. However, some of these mining sites had persons do business and living in hunts around them, which overtime have developed into hamlets and villages where elevated level of radiation has been recorded. Literatures abound on research works that have been undertaken to precisely quantify the amount of radioactivity levels in different soil and solid minerals found in Nigeria in recent time *(Avwiri et al., 2010; Sadiq and Agba, 2011;Agbalagba et al., 2012; Ademola and Onyema, 2014; Azionu et al., 2019; Babatunde et al., 2019;)* and some countries of the world for radiation protection *(Ragheb, 2007; Belivermis, 2012; Charro et al., 2013; Kovacs et al., 2013; Santawamaitre et al., 2014; Guidotti et al., 2015; Milenkovic et al., 2015; Todorovic et al., 2015; Kavitha et al., 2016).* In the Northern and Western Nigeria, a sizeable number of research work have been conducted in this regard (*Ademola and Obed, 2012; Innocent et al., 2013; Ademola et al., 2014*), while little or nothing has been done in the eastern region of the country with rich solid mineral present. It is worth mentioning that investigation on the level of dosage and excess level of radiation in the risk of cancer in this area has been reported in previous studies *(Wahsha et al., 2016: Ugbede and Echeweozo 2017; Ugbede, 2020; Ugbede and Osahon, 2021)*. This has necessitated the focus of this research work on the Eastern region of Nigeria. Moreso, most of these studies focused on radioactivity concentration in solid mineral with little or no attention given to the measurement of radioactivity levels in the soil where the minerals are found and which the public make greater contact with for farming, building and other domestic uses.

The government of Nigeria in recent time are making deliberate efforts to revamp mining activities in these long abandoned mineral resources mining sites to boost the internally generated revenues (IGR) of these States and for regional developed and integration, though according to the Nigerian Mining Act (2007), all mineral resources regardless of where they are occurring it is under the control of the Federal Government. But the growing concern of the radiation safety and health status of those living and working within these mineral mining environments are always not put into consideration in the planning and implementation of mining companies and government. This lend credence to this research work, because results obtained will serve as baseline data in these study areas and data obtained shall be sources of reference for future radiological impact evaluation studies, serving as data base which may be incorporated to Nigeria Nuclear Regulatory Agency (NNRA) resources for National Planning.

2.0 Materials and Methods

2.1 Location of Study Area

Mineral resources abound in different part of Nigeria, proper harnessing of the resources based on the host rocks as presented in the Figure 1, hence giving rise to the different types of mineral deposits as shown in Figure 2. To conform to international best practices is the greatest challenge of the industry (see Figure 2). This study was conducted in five Eastern Geopolitical Zone (Made up of Abia, Anambra, Ebonyi, Enugu and Imo States). The region lies between longitudes 7° 6" E and 7° 54" E and latitudes 5° 56" N and 6° 52" N. It encompasses an area of about 7161km^2 with elevation ranging from 32.0m to 590.2m above mean sea level *(Osimobi et al., 2018).* The region has two main landforms viz; a high relief central zone with undulating hills and ridges and lowland area. The high relief zone is geologically associated with the syncline composed of Ajali Sandstone and Nsukka Formation, while the eastern lowland zone is associated with rocks of Asu River group, Eze Aku Shale group, Awgu/Ndeabor Shale group, Asata/Nkporo Shale group and parts of Mamu Formation *(Osimobi et al., 2018).*

2.2 Sample Collection and Preparation Techniques

Soil samples were collected from coal and silica mining sites in Ewe in arochukwu in Abia State, glass-sand mine in Mbara-Ozu sand sites in Ihiala in Anambra State, limestone and iron-stone mine in Akpuoach and Ishiagu sites in Ebonyi State. Soil samples were also collected from bitumen, coal and gypsum mine Ezeagu, Udi and Aninri sites in Enugu State, and clay and kaolin mine Isu and Okigwe sites in Imo State. A total of forty-eight soil samples were collected in all, and one control sample each from a non-mineral mining location from the five states. The soil samples were collected at depths of 0 to 10 cm (which represents the soil permeability to particle settlement depth variation), within the different mineral mine sites in black paper bags (to prevent interaction with sunlight to avoid breaking down of the radionuclides present). The soil samples collected were spread on stainless still sheets at ambient temperature for seven days to dry in a controlled environment to prevent local dust contamination. Samples were further dried in an oven at regulated temperature of 60°C to attain a constant weight.

The dried samples were then grounded using mortar and pestle to pulverize form and then filtered using 100-mesh sieve. At each interval of pulverization, the pestle and mortar were clean using methylated spirited to avoid crossed contamination. The dried homogeneously pulverized samples with dry-weight of 250 g were filled in air tight cylindrical plastic container

(Marinelli beaker) that is of the detector geometry, and stored for a period of 28 days before counting to allow for secular equilibrium to be attained between ^{226}Ra and its short lived ^{222}Rn progeny *(Zarie and Al Mugren, 2010;Avwiri et al., 2012; Ononugb et al., 2017; Wang, et al., 2017; Ugbede, 2020).*

2.3 Radioactivity Analysis of Samples

The soil samples analysis for the natural radionuclide concentration were carried out using a computerized γ-ray spectrometry system with high purity germanium (HpGe) detector. The relative efficiency of the detector system was 39% and resolution of 1.8 kev at 1.33 MeV of Co-60. The spectrometer was attached to conventional electronics connected to a multichannel analyzer (MCA) card installed in a laptop computer. MAESTRO-32 software program was deployed to accumulate and analyze the data of the natural radionuclides present in the samples. The detector is located inside a cylindrical lead shield of 5 cm x 24 cm x 60 cm geometry. The metal (lead) shield was lined with different coatings of copper, cadmium and Plexiglas, of thickness 3 mm each. A counting time of 10 hours was adopted from the system calibration result for the acquire samples spectral data.

The high resolution of the HpGe detector made it possible to identify many γ-rays of the analyzed samples. The radioactivity levels of the uranium series were obtained using γ-ray emissions of ^{214}Pb at 351.9 keV (35.9%) and ^{214}Bi at 609.3 keV (44.9%), for the ^{232}Th-series, the emissions of ^{228}Ac at 911 keV (26.6%), ^{212}Pb at 238.6 keV (43.2%) and ^{208}Tl at 583 keV (30.2%) and were used as the radionuclide emission probabilities of γ_p. The ^{40}K activity levels was acquired straight from its emission line of 1460.8 keV (10.7%). The background spectra measured were used to correct the computed sample activities concentration in accordance with standard procedures *(Zarie and Al Mugren, 2010, Avwiri et al., 2012 and Ononugbo et al., 2017).*

The radioactivity content (A_c) in Bq kg^{-1} of the radionuclides were computed after decay correction was made using the expression *(Adamu et al., 2013).*

$$A_c = \frac{N_p}{\varepsilon_{ff}\, x\, M_s\, x\, T_c x\, \gamma_p} \quad Bqkg^{-1} \qquad (1)$$

Where; $A_c$is the sample activity concentration, N_p is the net peak area of a peak at energy, ε_f is the efficiency of the detector for a γ-energy of interest, M_s is the sample mass, $T_c\, is\, the$ total counting time and γ_p is the emission probability of radionuclide of interest.

2.4. Radiological Hazard Indices

2.4.1 Radium Equivalent Activity (Ra_{eq})

The radium equivalent (Ra_{eq}) activity is the measured number of activities of the natural radionuclides (Radium, Thorium, Potassium) and is established on the proven fact that 1 Bq kg^{-1} of ^{226}Ra, 0.7 Bq kg^{-1} of ^{232}Th, and 13 Bq kg^{-1} of ^{40}K generate equal radiation dose rates *(Osimobi et al., 2018)*. Radium equivalent (Ra_{eq}) equates the specific activity levels of the sample contained in the three natural radioactivity (^{40}K, ^{226}Ra and ^{232}Th) by a sole amount and account for the radiological risk *(Agbalagba et al., 2012)*. The index is very useful in regulating safe allowable standards and is estimated using the expression (*Kavitha et al., 2016)*:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_k \quad (2)$$

where Ra_{eq} (Bq kg^{-1}) is the radium equivalent, C_{Ra}, C_{Th} and C_k are the activity levels (Bq kg^{-1}) of ^{226}Ra, ^{232}Th, and ^{40}K respectively. Every material or environment whose Ra_{eq} values exceed 370 Bq kg^{-1} is strongly advised to be avoided *(Wang et al., 2017)*.

2.4.2 Absorbed Dose Rate (D_R)

The outdoor (D_R) is the gamma emission in air which represents an even dispersal of ^{40}K ,^{226}Ra and ^{232}Th. The outdoor (D_R) value is computed using the guidelines given by *UNSCEAR* and is expressed as *(Ashraf et al., 2010; UNSCEAR 2010)*:

$$D_R = 0.462C_{Ra} + 0.621C_{Th} + 0.0417C_K \quad (3)$$

where D_R ($\eta Gy\ h^{-1}$) is the outdoor dose rate, C_k, C_{Ra}, C_{Th}, are the activity content levels in (Bq kg^{-1}) for ^{40}K, ^{226}Ra and ^{232}Th, respectively.

UNSCEAR (2010) reported that the global permissible limit value of absorbed dose for the public should be 59 nGy h^{-1}.

2.4.3 Annual Gonadal Equivalent Dose (AGED)

Protecting the vital organs outer layers is of key importance to the radiation community *(UNSCEAR 2000; 2010)*. The AGED is estimated using equation 4:

$$\boldsymbol{AGED = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_k} \quad (\mathbf{4})$$

where AGED is the Annual Gonadal Equivalent Dose (mSv y^{-1}), and C_{Ra}, C_{Th}, and C_k (Bq kg^{-1}) are the radioactivity levels of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

2.4.3 *Hazard Index (External $\boldsymbol{H_{ex}}$)*

The hazard index ($\boldsymbol{H_{ex}}$) was a derivative of the Ra_{eq} calculation with the assumption that the maximum permissible value agrees with the 370 Bq kg^{-1} upper limit of Ra_{eq} value, with its equivalent radiation dose value limited to 1.0 mSv y^{-1}. The ($\boldsymbol{H_{ex}}$) index is computed applying the expression *(Wang et al., 2016)*:

$$H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_k/4810 \quad (5)$$

where H_{ex} is the external hazard index (Bq kg^{-1}), and C_k, C_{Ra} and C_{Th} are the radioactivity levels in (Bq kg^{-1}) for ^{40}K, ^{226}R and ^{232}Th respectively.

2.4.5 *Internal Hazard Index* ($\boldsymbol{H_{in}}$)

The internal index (H_{in}) is estimated as *(Kavitha et al., 2016)*:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_k/4810 \quad (6)$$

where H_{in} is the internal hazard index (Bq kg^{-1}), and C_k, C_{Ra}, C_{Th} are radioactivity levels in (Bq kg^{-1}) for ^{40}K, ^{226}R and ^{232}Th, respectively. $H_{in} \leq 1$ implies negligible radiation risk. Internal exposure to radon is very hazardous and can result to lung diseases like asthma and lung cancer.

2.4.6 *Representative Gamma (I_{yr})*

The representative gamma was formulated to estimate the γ-radiation risk linked to a specific natural radionuclide samples being investigated. It is an analytical tool for categorizing samples that might cause radiological implications if deployed for construction *(Agbalagba et al 2012)*. Values of $I_\gamma \leq 1$ correspond to 1.0 mSv, while $I_\gamma \leq 0.5$ is within an annual effective dose of 0.3 mSv *(Wang et al 2017)*.

The I_γ is expressed as *(Ashraf et al., 2010)*:

$$I_\gamma = C_{Ra}/150 + C_{Th}/100 + C_K/1500 \quad (7)$$

where I_γ is the representative gamma index (Bq kg^{-1}), and C_k, C_{Ra}, C_{Th} are the radioactivity content values (Bq kg^{-1}) for ^{226}R, ^{232}Th and ^{40}K respectively.

2.4.7 *Annual Effective Dose Equivalent (AEDE) Outdoor*

The *AEDE* keeps control on the effects of radiation on reproductive organs. This hazard index received outdoor by an individual is given as *(Avwiri et al., 2012; Ononugbo et al., 2017)*:

$$AEDE\ (Outdoor) = DR \times 8760 \times 0.7 \times 0.2 \times 10^{-3} \quad (8)$$

Where;

$AEDE\ (Outdoor)$ is given in $\mu Sv\ y^{-1}, DR\ in\ nGy\ h^{-1}$, 0.7 is the dose conversion factor given in $SvG\ y^{-1}$, 8760 h is the in a year and $0.2\ x\ 10^{-3}$ is the occupancy factor for outdoor.

2.4.8 Excess Lifetime Cancer Risk (ELCR)

Excess lifetime cancer risk estimates the likelihood of contracting cancer over a lifetime at specific exposure rate. It is the estimated number of extra cancers probable in each population of persons on exposure to a radiation at a specific dose.

The ELCR is computed using the expression *(Taskin et al., 2009)*:

$$ELCR = AEDE \times DL \times RF \quad (9)$$

where $ELCR\ has\ no\ units,$ AEDE is as defined in equation 8, the average Duration of Life (70 years) is the DL, while RF is known to as the Risk Factor, i.e., lethal cancer risk per Sievert (Sv^{-1}). ICRP recommend RF as 0.05 for stochastic effects for the public *(Taskin et al., 2009).*

In order to further understand our results, statistical analyses were performed using the SPSS software tool for mathematical/statistical data analysis. These include; Skewness, Kurtosis, mean, median, mode, standard deviation, minimum and maximum values.

2.5 Total Effective Dose

The total effective dose parameters depicting the occupational risk to oil and gas workers and the public' were estimated employing relevant conversion coefficients available in the literature (Table 3) using the equations *(Kola et al., 2016)*:

External exposure (D_{ext}) to gamma radiation from mine site and the exposed tailings, is calculated using the equation:

$$D_{ext} = \sum A_i\ C_{ext}\ T_e \quad (10)$$

Internal exposure (D_{inh}) from inhalation of solid mineral dust and contaminated air, is estimated using the expression

$$D_{inh} = \sum A_i\ C_{inh}\ \eta_{inh}\ D_f\ T_e \quad (11)$$

Internal exposure (D_{ing}) from any accidental ingestion of solid minerals, is estimated using the equation:

$$D_{ing} \sum A_i\ C_{ing}\ \eta_{ing}\ T_e \quad (12)$$

where A_i is the specific activity of nuclide i in Bq kg^{-1}, C_{ext}, is the effective dose coefficient for the nuclide in the contaminated surface measured in Sv h^{-1}/Bq g^{-1}, C_{inh}, is the dose coefficient for inhalation of the nuclide measured in Sv Bq^{-1}, η_{inh} is the breathing rate measured in $m^3\ h^{-1}$, and D_f is the dust loading factor, C_{ing}, is the dose coefficient for ingestion of the

nuclide measured in Sv Bq^{-1}; η_{ing} is the ingestion rate for adults, measured in kgh^{-1} and T_e is the exposure duration in years *(ICRP, 1991, 1996).*

3.0. Results and Discussion

3.1 Results of Radioactivity Analysis

The results of the soil γ-ray spectroscopy analysis in the ten solid mineral mine sites of the five eastern states of Nigeria are presented in Tables 1. Table 2 presents the summary of the analyzed radionuclides and the radiation risk indices while Table 3 present the computed occupational risk estimation to workers in the solid mineral mine sites

3.2. Discussion of Results

3.2.1 Specific Activity Concentration

The specific radioactivity levels obtained for the three natural radionuclides ^{40}K, ^{226}Ra and ^{232}Th in the investigated soil samples collected within solid mineral mine sites are shown in Table 1. The analyzed data obtained for the Iron-stone mine site soil activity concentration in Ebonyi State, shows activity value range of 32.45- 80.58 Bq kg^{-1}, 7.29- 30.66 Bq kg^{-1} and 25.88-67.61 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. Their mean values are higher than the control values by 47%, 5% and 46% respectively, while the mean ^{232}Th activity concentration of 77.28 Bq kg^{-1} obtained is above the *ICRP, IAEA* and *UNSEAR*, recommended permissible limit of 30 Bq kg^{-1} for the public. This may be attributed to the parent rock material from which iron- stone was formed *(UNSEAR 2010; IAEA, 2011)*. The activity concentration range for the kaolin mine sites soil samples in Imo State are 18.19-40.72 Bq kg^{-1} , 54.33-91.64 Bq kg^{-1} and 37.46-142.42 Bq kg^{-1}, for ^{226}Ra, ^{232}Th and ^{40}K, respectively, while their mean percentage elevation over the control values are 45%, 37% and 62% respectively. This elevation over the values obtained from the control sample can be attributed to the presence of these solid minerals within and around these sampled soils. At the silica mine site in Abia State, the range of activity concentration of the soil samples obtained are 127.08-289.79Bq kg^{-1}, 36.61-71.01 Bq kg^{-1} and 72.04-112.45 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively and the degree of their mean values elevation over the control value are 54%, 48% and 77% respectively, with the mean activity concentration values of ^{226}Ra (52.64 Bq kg^{-1}) and ^{232}Th (97.68 Bq kg^{-1}) exceeding their *ICRP* maximum permissible limits for the public (*ICRP*, *1996)*. These high values may be attributed to the influence of these radionuclides presents in solid minerals that are within the sampled soil environment. The percentage elevation of ^{40}K, ^{226}Ra

and ^{232}Th activity concentrations in the soil samples of the bitumen mine site in Enugu State over the control sample are 29 %, 41% and 62% respectively. The obtained activity concentrations in the soil samples from these mining sites compared favourably and agreed with the value reported from river sand sediment from across Enugu east in Enugu state *(Ugbede, 2020).* The ^{232}Th (68.79 Bq kg^{-1}) mean value was found to be well above the ICRP recommended permissible limits. Similarly, the percentage increase of ^{40}K, ^{226}Ra and ^{232}Th radioactivity levels in the sampled soil for the coal mine site at Enugu state over the control sample are 43%, 47% and 66% respectively, with ^{232}Th activity concentration grossly exceeded the global permissible limit for the public. This high value of ^{232}Th in the soil samples can be attributed to the high content of ^{232}Th in coal mineral *(Faanu et al., 2011; Innocent et al., 2013; Wang et al., 2017).*

The radioactivity content range in the sampled soil at the clay mine sites in Imo State are 34.84-275.58 Bq kg^{-1}, 17.55-46.40 Bq kg^{-1} and 36.25-84.87 Bq kg^{-1} in ^{40}K, ^{226}Ra and ^{232}Th respectively. These range of values agrees with the reported natural activity concentration value in soil samples from Slovenia (*Kovács et al., 2013).* Their mean activity concentration percentage elevation over the control values is 51%, 50% and 47% respectively with ^{232}Th (58.91 Bq kg^{-1}) mean activity concentration still exceeding the global permissible limit for the public. The percentage elevation of ^{40}K, ^{226}Ra and ^{232}Th activity levels in the sampled soil of the gypsum mine site at Enugu State over the control sample are 61%, 63% and 63% respectively. The ^{226}Ra (49.46 Bq kg^{-1}), and ^{232}Th (67.25 Bq kg^{-1}) mean activity levels are above the global permissible limit 35 Bq kg^{-1} for ^{226}Ra and 30 Bq kg^{-1} for ^{232}Th for the public. At the coal mine site in Abia State, the activity concentration range of the sampled soil obtained are 25.67-174.22 Bq kg^{-1}, 15.38-27.12 Bq kg^{-1}, and 37.99-64.36 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. The percentage mean radioactivity levels elevation over the control value are 55%, 28% and 54% respectively, with mean ^{232}Th (55.31 Bq kg^{-1}) value observed to be above UNSCEAR recommended permissible limits for the general public *(UNSCEAR, 2000).* The activity concentration range of sampled soil at the limestone mine site in Ebonyi State as obtained are 23.73-313.74 Bq kg^{-1}, 18.73-48.26 Bq kg^{-1} and 29.70-70.41 Bq kg^{-1}, for ^{40}K, ^{226}Ra and ^{232}Th respectively. Their mean percentage elevation over the control values is 62%, 60% and 64% respectively with ^{232}Th (48.78 Bq kg^{-1}) mean activity concentration agreeing with the value reported at Ezillo paddy rice field in Ebonyi State, but also exceeding the global permissible limit for the general public *(Ugbene and Osahon, 2021).* Similarly, the activity concentrations range for sampled soil at the glass stone mine site in Anambra State as measured

are 21.00-70.10 Bq kg^{-1}, 14.19-28.90 Bq kg^{-1} and 74.18-588.93 Bq kg^{-1} for ^{40}K, ^{226}Ra and ^{232}Th respectively. Their mean percentage rise of ^{40}K, ^{226}Ra and ^{232}Th activity concentrations in the soil samples over the control sample are 39%, 58% and 82% respectively, with ^{232}Th (189.11 Bq kg^{-1}) activity concentration grossly exceeded the public permissible limit.

The soil specific activity levels obtained in the entire surroundings of the mining sites for the different solid mineral clearly indicate that ^{232}Th radioactivity concentration is higher and exceed the three naturally occurring radionuclides examined permissible limits for the public. This is an indication that most of the solid minerals are laced with radioactivity with thorium activity most prominent, which may be attributed to the geological formation of the subsurface rocks of the studied area and the weathering processes that takes place. It was observed that the mean activity of ^{226}Ra and ^{40}K reported in this study-areas are above many reported values in literatures in similar environment within Nigeria, West Africa and other parts of the globe *(Faanu et al., 2011; González-Fernández et al., 2012; Innocent et al., 2013; Wang et al., 2017).* Moreover, the ^{226}Ra value obtained in this study is well within reported values in literatures in similar solid mineral mining environments in Nigeria and in other parts of the world *(Amrani and Tahtat, 2001; El Afifi et al., 2006; Kam and Bozkurt, 2007; Al-Hamarneh and Awadallah, 2009; Ademola and Obed, 2012; Avwiri et al., 2013; Kovács et al., 2013; Ademola et al., 2014; Hannan et al., 2015).*

3.2.2 Radiological Hazard Parameter Statistical Analysis

Table 2 shows the summary of the result of the statistically analyzed specific activity levels and radiation hazard/ risk indices. The eight radiation risk parameters were computed using reported standard and internationally established equations in literatures *(Ashraf et al., 2010; UNSCEAR, 2010; Avwiri, et al., 2012; Gang, et al., 2012; Sivakumar, et al., 2014; Wang, et al., 2016).* From the result, the estimated Radium Equivalent (Ra_{eq}) varied from 87.51 Bq kg^{-1} to 300.13 Bq kg^{-1} with a mean and mode values of 150.72 Bq kg^{-1}, and 87.51 Bq kg^{-1} respectively with a standard deviation of 61.25 Bq kg^{-1}. This Ra_{eq} result obtained is above reported value obtained in s solid mineral mine site in south-western Nigeria and sampled soil valued obtained in some cities and towns in Nigeria (*Agbalagba, et al., 2012; Avwiri, et al., 2012; Innocent et al., 2013; Ademola et al., 2014; Aliyu et al., 2015; Ononugbo et al., 2017; Ugbede, 2020; Ugbede and Osahon, 2021).* The Absorbed dose rate (D) has a minimum value of 38.96 ηGyh^{-1} and a maximum measured value of 133.00 ηGyh^{-1} with 68.40 ηGyh^{-1} be the mean. The mean absorbed dose value recorded exceeded the *UNSCEAR, (2010)*

recommended worldwide ambient value of 59 ηGyh^{-1}. The outdoor Annual Effective Dose Equivalent (AEDE) has its values varied from 48.26 μSvy^{-1} to 160.78 μSvy^{-1} with the mean and mode values of 83.65 μSvy^{-1} and 48.26 μSvy^{-1} respectively across the study-area and a standard deviation value of 32.66 μSvy^{-1}. The estimated average value of the outdoor Annual Effective Dose Equivalent of 83.65 μSvy^{-1} correspond to the reported values obtained in measured soil samples in Bethlehem Province of Palestine and soil from open landfills site in Rivers State Nigeria, but it is higher the worldwide annual effective dose equivalent value of 70 $\mu Svy^{-1}$for outdoor *(Agbalagba et al., 2012; Mohammad et al., 2014; Ononugbo et al., 2017; Vukasinovic et al., 2017; Ugbede, 2020).*

The estimated value of the Annual gonadal equivalent dose (AGED) varied from 263.62 μSvy^{-1} to 879.51 μSvy^{-1} with a mean value of 454.70 μSvy^{-1}. The estimated value recorded is above the ambient level recommended world permissible value of 300 μSvy^{-1} and values obtained in reported research of wasteland soil in Namibia, but they are below the values reported in southern dump site sampled soil and Northern soil samples from solid mineral mining environment Nigeria as reported in literatures *(UNSCEAR, 2010; Aliyu et al., 2015; Ononugbo et al., 2017; Onjefu et al., 2021)*. The accumulative dose rate at the present exposure rate over a twenty years' time may impair the reproductive organs (ovaries and testis) of those working and living around these mine sites if not properly shielded.

The mean results of the estimated health hazard indices indicates that External hazard index (H_{ex}) has a mean value of 0.41, while the estimated Internal hazard index (H_{in}) value is 0.50 and the Representative gamma index (I_{γ}) had a value of 1.06. The results are comparable favourably with the values reported in Wasteland soil of Okakarara in Namibia, the shore sediment of North dune beach in Namibia and the values reported in soil and sediment of Al-Nigella in Egypt *(Onjefu et al., 2017; Ahmed et al., 2020; Onjefu et al., 2021)*. The mean hazard indices values obtained for H_{ex} and H_{in} are less than the 1.0 (critical value), the general public recommended permissible limit, however, Representative gamma index (I_{γ}) value of 1.06 obtained was slightly above the UNSEAR recommended value for the general public *(UNSCCEAR, 2010)*. These obtained estimated values of the Excess Life Cancer Risk (ELCR) result ranged from 165.45 x 10^{-6} to 559.31 x10^{-6} with a regular value of 289.14x10^{-6}. This mean ELCR value of $289.14\text{x}10^{-6}$ obtained is approximately the recommended ambient public permissible limit of $0.29\text{x}10^{-3}$ *(UNSCEAR, 2000)*. This implies that the likelihood of radiation induced health risks among residence and workers in the environs of these mine sites is probable, especially for prolong and continuous radiation exposures from these studied

minerals sites. The statistical analysis of the results of radionuclides and the radiological risk indices are indicated in figure 3, while figure 4 shows the sequential chart distribution of the three natural radionuclides investigated with the radiological risk parameters examined. It was observed from figure 3 that the histogram showing the spatial dispersion of specific activities of the three natural radionuclides (^{40}K, ^{226}Ra and ^{232}Th) in the analyzed soil samples from the investigated sites, were asymmetrical distribution with the skewness of 0.19, 0.61, 2.53 respectively. The median values for ^{40}K, ^{226}Ra and ^{232}Th were 99.80, 32.23 and 68.34 Bq kg^{-1} respectively, thus data obtained in this study were accepted as reliable for use in the determination of the radiological risk *(Kessaratikoon et at., 2019).* From figure 4, the two rigs in the sequence chart are an indicative of the areas of high aggregation of the activity concentrations and areas of strong correlation of activities concentration with radiological risk parameters.

3.2.3 Occupational Risk Estimate

This model account for the occupational hazard associated with work environment, which in this study is the solid mineral mining sites. As a result of constant working at the sites and in most cases not wearing the appropriate Personnel Protective Equipment (PPE), workers are often exposed to radionuclide. The three major paths for this exposure are according to *Kolo et al. (2016)*:

The total of these three different paths of exposure gives the total effective dose which is the concerned parameter. To be within the safe ICRP limit, the Total Effective Dose from these three pathways must not be more than 1.0m Sy^{-1} for the public *(ICRP, 1991)*.

The result presented in Table 3 of the Occupational Risk Estimate indicates that the external exposure to gamma radiation (D_{ext}) has the highest Occupational risk ranging from 0.51mSv y^{-1} to 1.3 mSv y^{-1}, followed by Internal exposure from inhalation of radiation from solid mineral dust and contaminated air (D_{inh}) ranging from 0.01 mSv y^{-1} to 0.99 mSv y^{-1} and the least is Internal exposure from any accidental ingestion of radiation from solid minerals (D_{ing}) ranging from 0.08 mSv y^{-1} to 0.24 mSv y^{-1}. The reason for this result distribution is obvious as one in a mining site would be exposed externally on the skin and inhalation before even having to experience accidental ingestion. This result indicates that the external organs like the eye and skin of the people working at these mining sites may be at risk of eye and skin radiation related infections. However, the overall results suggests that the effect put together is within control limit as the whole organs of the body fights together to wear the would-be effect from one pathway.

Conclusion

The evaluation of natural radioactivity levels of sampled soil from some selected solid minerals mining sites and soil from non-mineral mining areas (control) in the Eastern region of Nigeria has been investigated using gamma spectroscopy analysis. The measured soil activities of ^{40}K, ^{226}Ra and ^{232}Th were deploy to compute the percentage gamma radiation elevation over the control sample and the risk parameters. Radioactivity analysis of the sampled soil shows that some radionuclides values measured exceeded their standard limits. The overall average percentage rise in ^{40}K, ^{226}Ra and ^{232}Th value in the mine sites soil samples over the control soil samples are 48.6%, 43.7% and 62.3% respectively, with thorium having the highest percentage rise. This affirmed previous research report of ^{232}Th be the major contributor of the dose that can be received from the terrestrial environment. The occupational risk estimation results indicate that the external organs of the people working and living around these mining sites are at risk which may lead to eye and skin radiation related infections.The exceeding of global recommended permissible and ambient limits of certain radiation hazard indices estimated compared to previously reported values from similar mineral mining environment is an indication of a radiologically contaminated environment, which is attributable to the solid minerals mining and processing in the studied areas. The researcher therefore recommends that proper kitting of workers and discouragement of people residing around these mining sites to reduce the radiation impact on people and the environment.

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Ethical Approval

This paper abides by all ethical standard of the journal and contain no ethical issues whatsoever.

Consent to Participate

All authors consent was sorted and agreed before the research work commenced. All authors also consent to participate in the writing of this article and the submission of the final paper to this journal.

Consent to Publish

This is to certify that all authors consents were sorted and approval gotten to publish this paper in this Journal. We the authors declare that there is no conflict of interest among us in this research work.

Authors' contributions

The research work was conceived by Dr. Agbalagba E.O and redesign for wider readability by Prof. Stephen O. Egarievwe, while Dr. Mohammed S. Chaanda did the geological identification and characterization of sites and solid minerals in each site. All the three authors contributed mutually from the development and data analysis in this research work. Dr. Agbalagba E.O. did the writing which other authors vetted and agreed before sending for consideration in this Journal.

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Availability of Data and Material

The datasets generated and/or analyzed during the current study are not publicly available, but will be made available from the corresponding author on reasonable request

Competing Interests

Not applicable

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List of Tables

Table 1: Activities (Bq kg^{-1}) of ^{40}K, ^{226}Ra, ^{232}Th from solid mineral mining soils in South-Eastern Nigeria

S/N	geographical location	Mining State	Mineral found	^{40}K (Bq kg^{-1}) Mean (Range)	^{226}Ra (Bq kg^{-1}) Mean (Range)	^{232}Th (Bq kg^{-1}) Mean (Range)
1	N06°22′45" E007°27′52"	Ebonyi	Ironstone	54.26 (32.45-80.58)	20.45 (7.29-30.66)	48.18 (25.88-67.61)
2	N06°45′44" E007°16′17"	Imo	Kaoline	81.65 (37.46-142.42)	31.83 (18.19-40.72)	71.08 (54.33-91.64)
3	N06°28′08" E007°27′03"	Abia	Silica	179.15 (127.08-289.79)	52.64 (36.61-71.01)	97.68 (72.04-112.45)
4	N06°05′27" E007°26′34"	Enugu	Bitumen	34.80 (21.93-60.46)	32.66 (21.25-40.41)	68.79 (48.74-91.00)
5	N06°25′47" E007°28′00"	Enugu	Coal 1	117.03 (64.11-238.10)	36.37 (26.09-43.73)	69.94 (60.87-80.04)
6	N06°09′56" E007°29′07"	Imo	Clay	117.34 (34.84-275.58)	35.23 (17.55-46.40)	58.91 (36.25-84.87)
7	N06°31′05" E007°26′34"	Enugu	Gypsum	136.0 (87.40-161.82)	49.46 (41.24-53.65)	67.25 (42.08-90.54)
8	N06°23′53" E007°27′15"	Abia	Coal 2	82.58 (25.67-174.22)	22.47 (15.38-27.12)	55.31 (37.99-64.36)

9	N06°24′34" E007°28′52"	Ebonyi	Limestone	155.68 (23.73-313.74)	30.66 (18.73-48.26)	48.78 (29.70-70.41)
10	N06°24′54" E007°27′45"	Anambra	Glass-sand	48.02 (21.00-70.10)	19.85 (14.19-28.90)	189.11 (74.18-588.93)
	Overall Mean Value			**100.22±8.20**	**33.15±3.31**	**77.31 ±6.10**
	Control (Mean)			28.60	19.63	25.92
	Global Recommended Value (UNSEAR 2010)			**400**	**35**	**30**

1

2 **Table 2. Risk Estimate for workers in solid mineral mining site**

Variables	**K-40 (Bq kg^{-1})**	**Ra-226 (Bq kg^{-1})**	**Th-232 (Bqkg^{-1})**	**Raeq (Bq kg^{-1})**	**D (ηGyh^{-1})**	**AEDE (μSvy^{-1})**	**AGED (μSvy^{-1})**	**Hex**	**Hin**	I_γ	**ELCR x10^{-6}**
Mean	100.22	33.15	77.31	150.72	68.40	83.65	454.70	0.41	0.50	1.06	289.14
Median	99.80	32.23	68.34	136.45	60.61	74.33	409.40	0.37	0.46	0.95	260.17
Mode	34.80	19.88	44.30	87.51	38.96	48.26	263.62	0.24	0.29	0.61	167.24
Std. Deviation	48.71	11.18	43.34	61.25	26.63	32.66	179.23	0.17	0.17	0.43	114.30
Skewsoilness	0.19	0.61	2.53	1.81	1.73	1.73	1.71	1.81	1.18	1.80	1.73
Kurtosis	-1.14	-0.33	6.94	3.67	3.34	3.34	3.26	3.67	1.28	3.64	3.34
Minimum	34.80	19.88	44.30	87.51	38.96	48.26	263.62	0.24	0.29	0.61	165.45
Maximum	179.15	52.65	193.11	300.13	133.00	161.80	879.51	0.81	0.86	2.10	559.31

Table 3: Occupational Risk Estimate for workers in solid mineral mining sites

Code Name	Mineral Mined	Ra-226 (Bq kg^{-1})	Th-232 (Bq kg^{-1})	K-40 (Bq kg^{-1})	D_{ext} (mSv y^{-1})	D_{inh} (mSv y^{-1})	Ding (Sv y^{-1})	Total Eff. Dose (μSv y^{-1})
LS	Limestone	30.63	48.87	155.68	0.97	0.50	0.10	51.30
CL1	Coal	36.37	69.96	117.03	0.99	0.71	0.13	72.40
CY	Clay	35.23	59.10	117.34	0.97	0.01	0.12	61.70
BN	Bitumen	32.66	69.44	34.80	0.73	0.71	0.12	71.42
IS	Ironstone	20.45	47.98	54.26	0.53	0.48	0.08	49.26
SC	Silica	52.64	97.72	179.15	1.47	0.99	0.18	10.11
KL	Kaolin	31.80	70.04	81.65	0.82	0.71	0.12	72.02
GM	Gypsum	49.49	67.25	136.04	1.30	0.69	0.15	71.14
GS	Glass-sand	19.88	193.11	48.02	0.51	0.02	0.24	19.27
CL2	Coal	22.48	53.31	82.58	0.64	0.54	0.09	54.73

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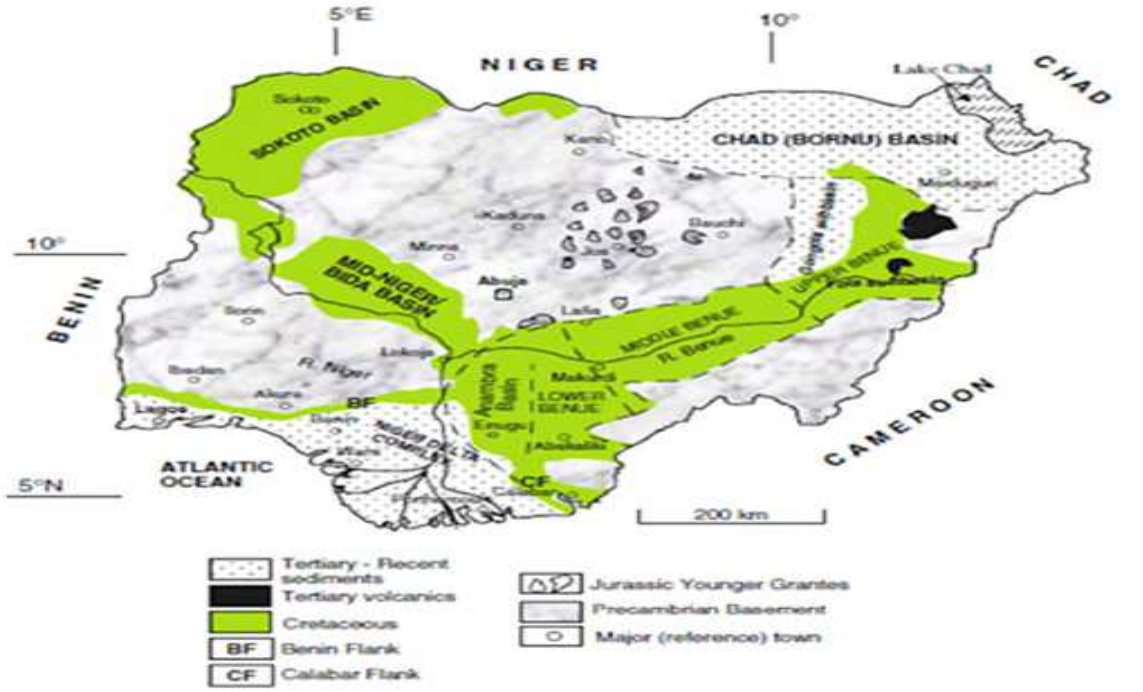

Figure 1: Geological Map of Nigeria showing the location of the area studied *[59]*

Figure 2: Solid Minerals and Locations Found in Commercial Quantity in Nigeria *(Source: Report of the Vision 2020 National Technical Group on Minerals and Metals Development)[60]*

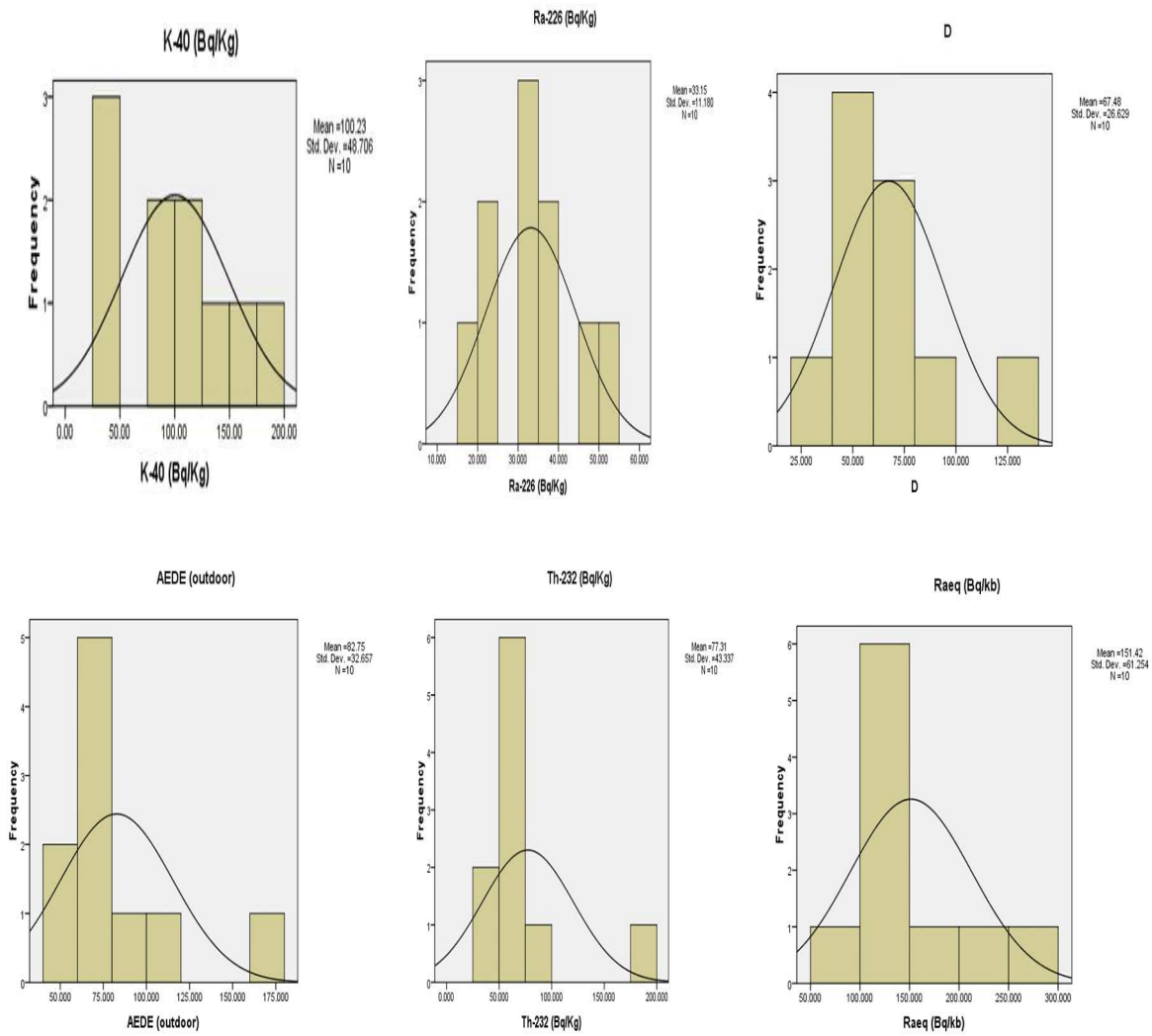

Figure 3: Histogram and Skewness plot of some Radiation parameters.

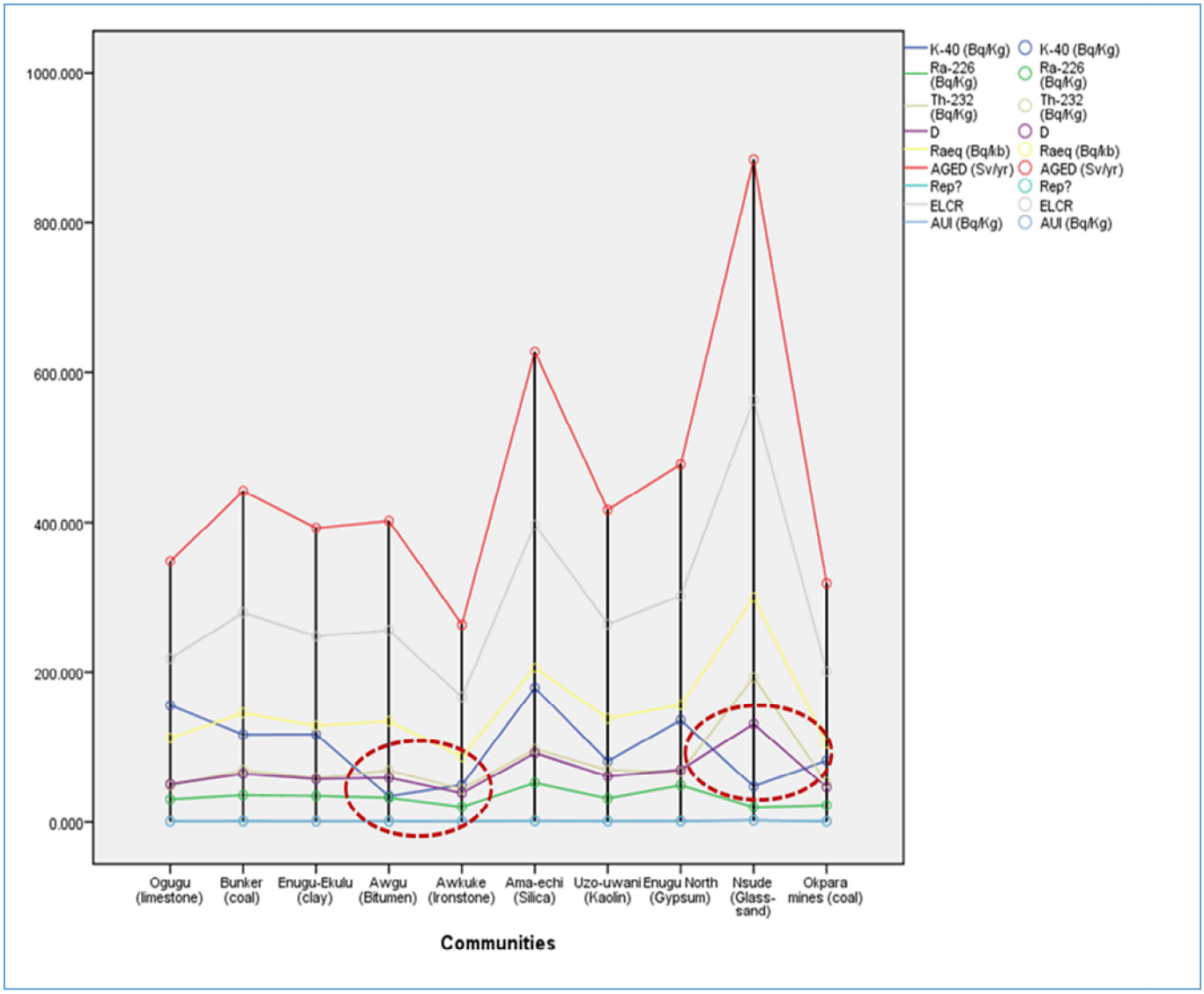

Figure 4: Sequence Chart of the Statistical Analysis.

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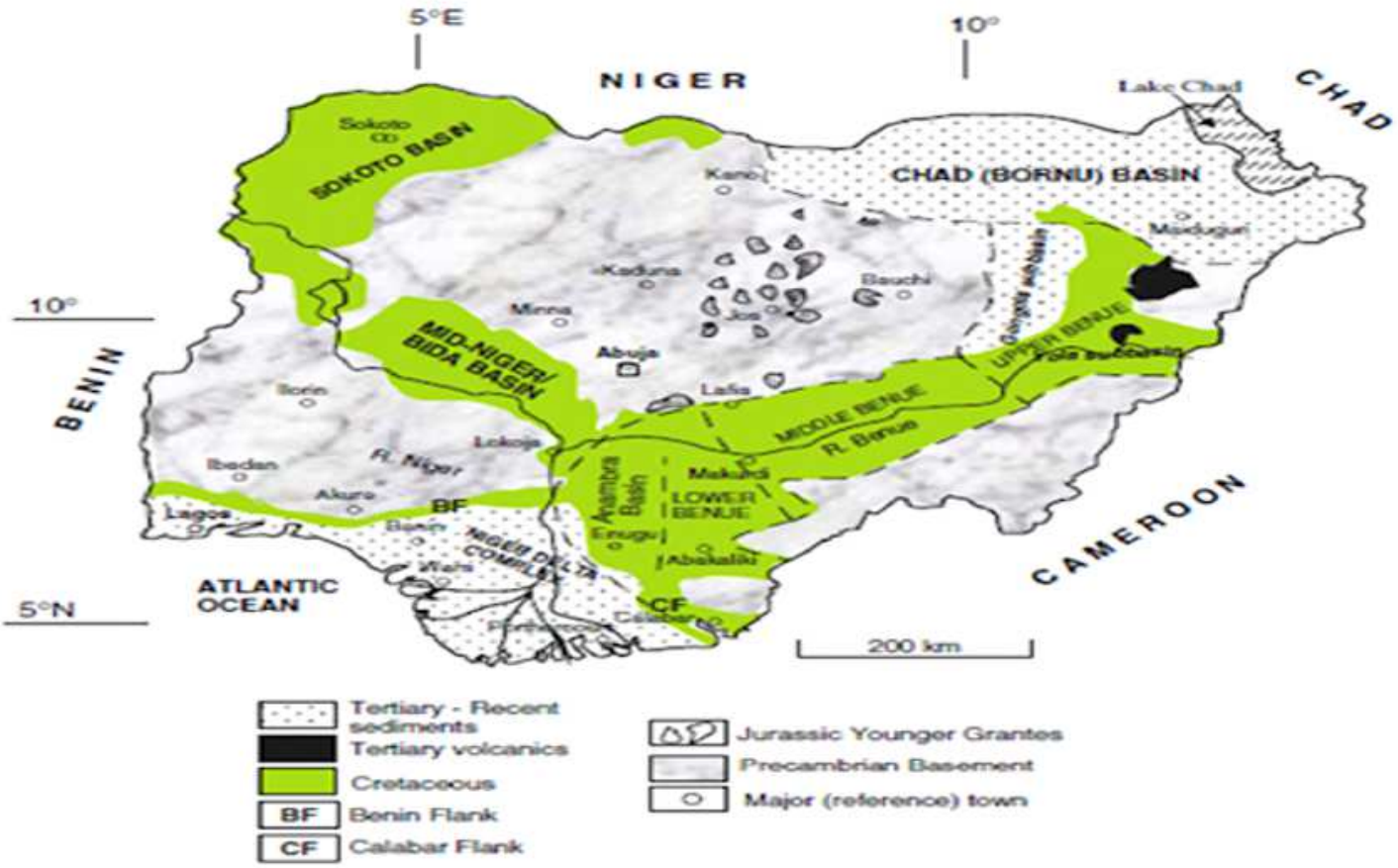

Figure 1

Geological Map of Nigeria showing the location of the area studied [59] Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

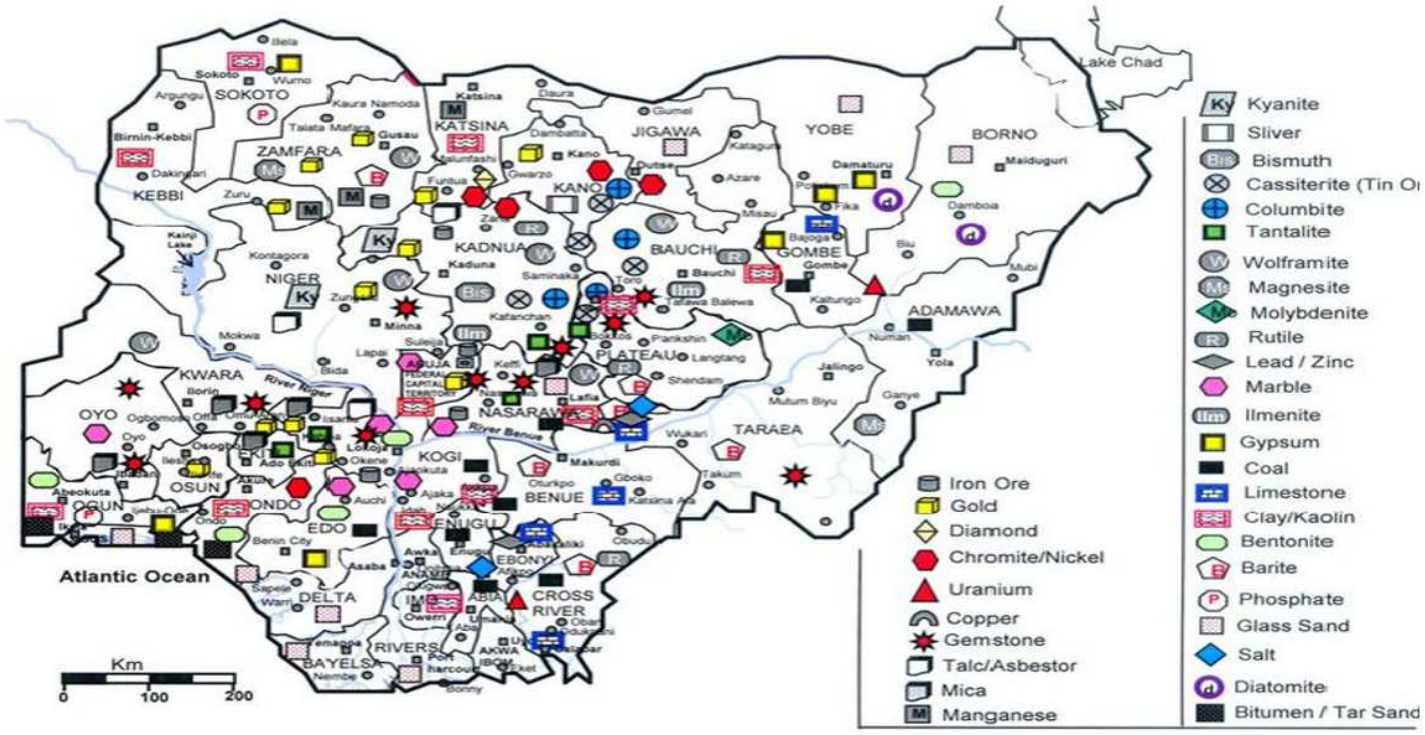

Figure 2

Solid Minerals and Locations Found in Commercial Quantity in Nigeria (Source: Report of the Vision 2020 National Technical Group on Minerals and Metals Development)[60] Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

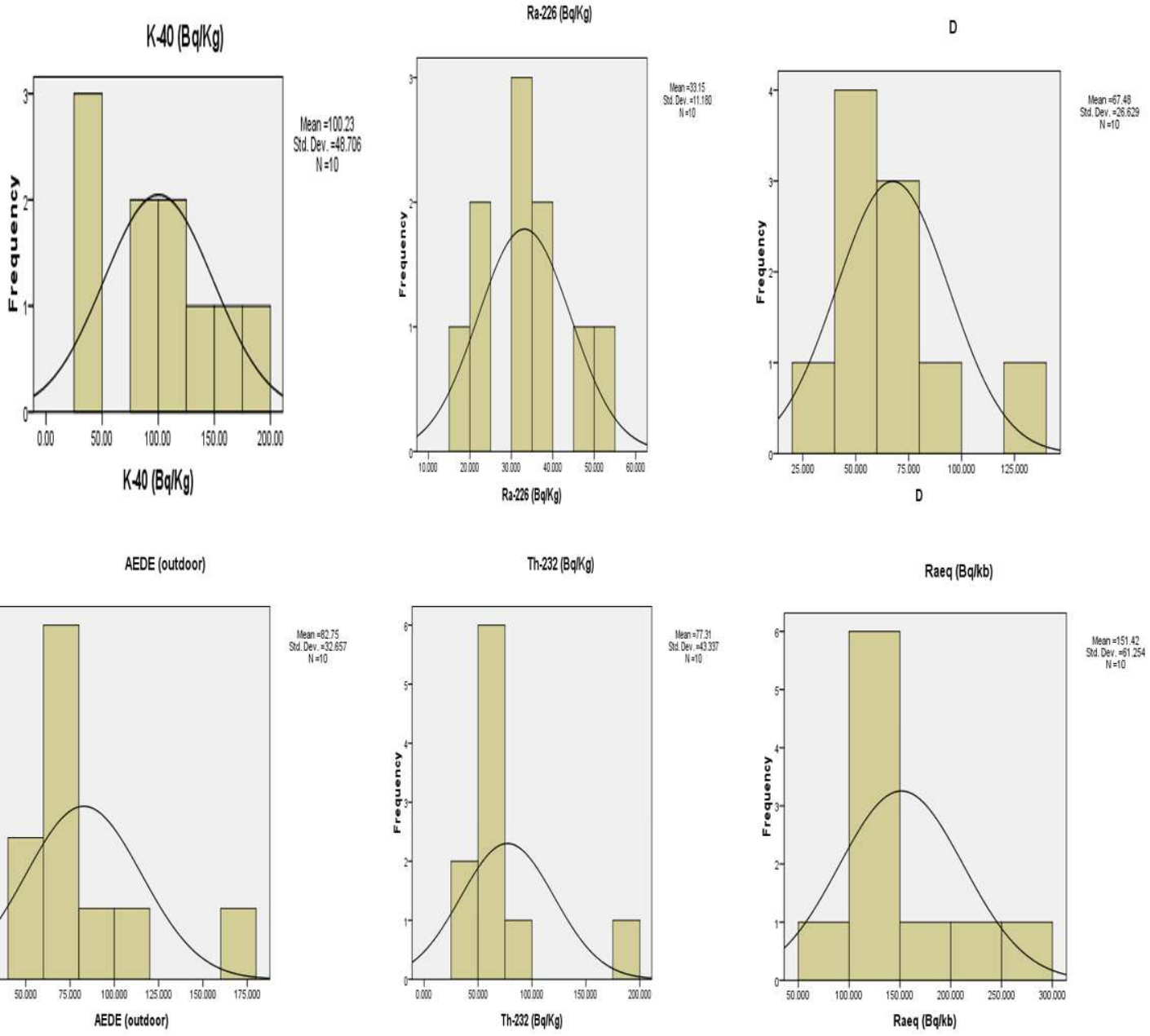

Figure 3

Histogram and Skewness plot of some Radiation parameters.

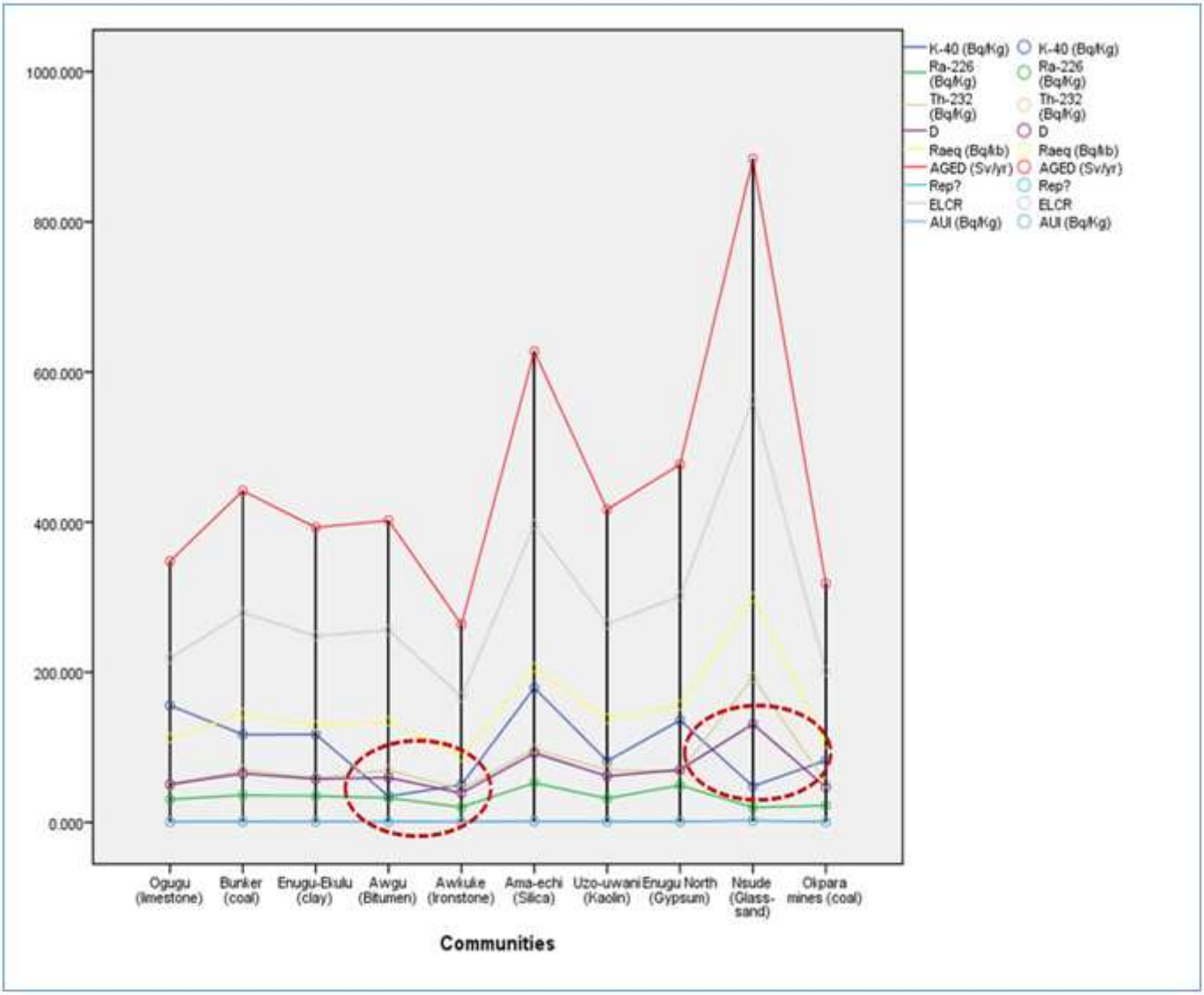

Figure 4

Sequence Chart of the Statistical Analysis.