Assessment of solid mineral to soil radioactivity contamination index in selected mining sites and their radiological risk indices to the public

Ezekiel Oghenenyerhovwo Agbalagba\textsuperscript{a}, Mohammed S. Chaanda\textsuperscript{b} and Stephen U. Egarievwe\textsuperscript{c}

\textsuperscript{a}Department of Physics, College of Science, Federal University of Petroleum Resources, Effurun, Nigeria; \textsuperscript{b}Department of Earth Sciences, College of Science, Federal University of Petroleum Resources, Effurun, Nigeria; \textsuperscript{c}Department of Electrical and Computer Engineering, Nuclear Engineering and Radiological Science Center, Alabama A&M University, Normal, AL, USA

\textbf{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 10 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 \(^{40}\text{K}\), \(^{226}\text{Ra}\) and \(^{232}\text{Th}\) are 100.22 Bq kg\(^{-1}\), 33.15 Bq kg\(^{-1}\) and 77.31 Bq kg\(^{-1}\), respectively. The \(^{226}\text{Ra}\) and \(^{40}\text{K}\) activities were found to be within the United Nation Scientific Committee on the Effects of Atomic Radiation acceptable permissible limit, but the \(^{232}\text{Th}\) mean value was above the permissible limit of 30 Bq kg\(^{-1}\) for the public. In comparison, \(^{40}\text{K}\), \(^{226}\text{Ra}\) and \(^{232}\text{Th}\) 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\(^{-1}\), 68.40 nGyh\(^{-1}\), 83.65 $\mu$Svy\(^{-1}\) and 454.70 $\mu$Svy\(^{-1}\) for the Radium Equivalent (Ra\textsubscript{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 ($I_\text{r}$) and Excess Life Cancer Risk (ELCR) were 0.41, 0.50, 1.06 and 0.29 $\times$ 10\(^{-3}\), respectively. The statistical analysis shows positive skewness.

\textbf{1. Introduction}

Assessment of background radioactivity level plays a significant role in the protection of man from excessive radiation exposure [1]. 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 syntgenetic 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 [2]. Radioactivity levels are evaluated as part of national and international survey at different areas and countries of the world for radiation protection [3,4].

\textbf{CONTACT}

Ezekiel Oghenenyerhovwo Agbalagba \textsuperscript{a} agbalagba.ezekiel@fupre.edu.ng

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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 [5]. 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 [6,7]. Other problems associated with high exposure to ionising radiations to health include alteration in the structure and functions of the cells and organs, deterministic effect, stochastic effects, irritations, sensitisation, embryonic effects, etc. The knowledge of radionuclide distribution in the environment is therefore of immense benefit in assessing the effects of radiation exposure; thus, the monitoring of radioactive materials are of primary importance to man and for the protection of the environment [5–9].

Most mineral deposits are associated with radionuclide like uranium, thorium and its' progenies. Primary uranium ore minerals when weathered, oxidised 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 [10]. They are also found in conglomerates, shale, limestone, sediment and hydrocarbon [11]. 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 [4,12]. Measurement of the radioactivity level of some rock samples, potential sources rocks of hydrocarbons have been conducted in different parts of the world [13,14].

It has been established from previous studies that some of these soil and minerals such as monazite, pyrochlore and xenotime that are obtained as byproducts of tin mining are radioactive [15–18]. Exposure to radiations emitted by some of these radioactive minerals is a major source of health hazards [19,20]. 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 [12,21–25] and some countries of the world for radiation protection [13,14,19,20,26–29,30,31]. In the Northern and Western Nigeria, a sizeable number of research work have been conducted in this regard [32–34], 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 [4,35–37]. 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.
2. Materials and methods

2.1. Location of study area

Mineral resources deposits abound in different part of Nigeria, due to the geology spread of these minerals in the country landscape as presented in Figure 1. This has given 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) as shown in Figure 3. 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 7161 km² with elevation ranging from 32.0 to 590.2 m above mean sea level [38]. 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, Awdh/Ndeabor Shale group, Asata/Nkporo Shale group and parts of Mamu Formation [38].

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 48 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 0–10 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 steel 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 for between 6 and 8 hours to attain a constant weight.

The dried samples were then grounded using mortar and pestle to pulverise form and then filtered using 100-mesh sieve. At each interval of pulverisation, the pestle and mortar were clean using methylated spirit to avoid crossed contamination. The dried homogeneously pulverised 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 [4,5,39–41].

2.3. Radioactivity analysis of samples

The soil samples analysis for the natural radionuclide concentration were carried out using a computerised $\gamma$-ray spectrometry system with high purity germanium (HpGe) p-type (model: BE2020 and Serial number: b15168) detector at the National Institute of Radiation Protection and Research, University of Ibadan, Ibadan, Nigeria. The relative efficiency of the detector system was 39% and resolution of 1.8 kev at 1.33 MeV of Co-60. Energy calibration of broad Energy Germanium detector was done with point sources of $^{241}$Am, $^{137}$Cs and $^{60}$Co. The reference material used for the efficiency calibration was IAEA-414 with a cylindrical geometry of equal dimensions as the sample vessels. The spectrometer was attached to conventional electronics connected to a multichannel analyser (MCA) card installed in a laptop computer. MAESTRO-32 software program was deployed to accumulate and analyse the data of the natural radionuclides present in the samples. The detector is located inside a cylindrical lead shield of 5 cm × 24 cm × 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 $\gamma$-rays of the analysed samples. The radioactivity levels of the uranium series were obtained using $\gamma$-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 $\gamma_p$. 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 [5,39,41].

The radioactivity content ($A_c$) in Bq kg$^{-1}$ of the radionuclides were computed after decay correction was made using the expression [42].

$$A_c = \frac{N_p}{\varepsilon_f J t M_x T_x \gamma_{p}} \text{Bq kg}^{-1} \tag{1}$$

Where: $A_c$ is the sample activity concentration, $N_p$ is the net peak area of a peak at energy, $\varepsilon_f$ is the efficiency of the detector for a $\gamma$-energy of interest, $M_x$ is the sample mass, $T_x$ is the total counting time and $\gamma_{p}$ 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 [38]. 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 [12]. The index is very useful in regulating safe allowable standards and is estimated using the expression [31]:

\[
Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{k}
\]  

(2)

where \(Ra_{eq}\) (Bqkg\(^{-1}\)) 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 [40].

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 [43,44]:

\[
D_{R} = 0.462C_{Ra} + 0.621C_{Th} + 0.0417C_{k}
\]  

(3)

where \(D_{R}(\text{mGy} h^{-1})\) is the outdoor dose rate, and \(C_{k}, C_{Ra}, C_{Th}\) are the activity content levels in (Bq kg\(^{-1}\)) for \(^{40}\)K, \(^{226}\)Ra and \(^{232}\)Th, respectively.

UNSCEAR [44] 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 [44,45]. The AGED is estimated using Equation 4:

\[
AGED = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_{k}
\]  

(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.

2.4.4. Hazard index (external \(H_{ex}\))
The hazard index (\(H_{ex}\)) 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 (\(H_{ex}\)) index is computed applying the expression [46]:

\[
H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_{k}/4810
\]  

(5)

where \(H_{ex}\) is the external hazard index (Bqkg\(^{-1}\)), and \(C_{k}, C_{Ra}\) and \(C_{Th}\) are the radioactivity levels in (Bqkg\(^{-1}\)) for \(^{40}\)K, \(^{226}\)R and \(^{232}\)Th, respectively.
2.4.5. Internal hazard index ($H_{in}$)

The internal index ($H_{in}$) is estimated as [31]:

$$H_{in} = C_{Ra}/185 + C_{Th}/259 + C_{k}/4810$$  \hspace{1cm} (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, $^{226}$Ra 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 ($I_{\gamma}$)

The representative gamma was formulated to estimate the $\gamma$-radiation risk linked to a specific natural radionuclide samples being investigated. It is an analytical tool for categorising samples that might cause radiological implications if deployed for construction [12]. Values of $I_{\gamma} \leq 1$ correspond to 1.0 mSv, 1.0 mSv, while $I_{\gamma} \leq 0.5$ is within an annual effective dose of 0.3 mSv0.3 mSv[40].

The $I_{\gamma}$ is expressed as [43]:

$$I_{\gamma} = C_{Ra}/150 + C_{Th}/100 + C_{k}/1500$$  \hspace{1cm} (7)

where $I_{\gamma}$ is the representative gamma index (Bq kg$^{-1}$), and $C_{k}, C_{Ra}, C_{Th}$ are the radioactivity content values (Bq kg$^{-1}$) for $^{226}$Ra, $^{232}$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 [5,39]:

$$AEDE(\text{Outdoor}) = DRx8760 \times 0.7 \times 0.2 \times 10^{-3}$$  \hspace{1cm} (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 Sv Gy$^{-1}$, 8760 hrs is in a year and $0.2 \times 10^{-3}$ 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 [47]:

$$ELCR = AEDE \times DL \times RF$$  \hspace{1cm} (9)

where $ELCR$hnounits. $AEDE$ 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 [47].

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 [48]:

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

$$D_{ext} = \sum A_i C_{ext} T_e$$

(10)

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

$$D_{inh} = \sum A_i C_{inh} \eta_{inh} D_f T_e$$

(11)

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

$$D_{ing} = \sum A_i C_{ing} \eta_{ing} T_e$$

(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$^{-1}$, $\eta_{inh}$ 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$^{-1}$, $\eta_{ing}$ is the ingestion rate for adults, measured in kgh$^{-1}$ and $T_e$ is the exposure duration in years [49,50].

3. Results and discussion

3.1. Results of radioactivity analysis

The results of the soil $\gamma$-ray spectroscopy analysis in the 10 solid mineral mine sites of the five eastern states of Nigeria are presented in Table 1. Table 2 presents the summary of the analysed radionuclides and the radiation risk indices, while Table 3 presents 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 analysed 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 [44,64]. The activity concentration range for the kaolin mine sites soil samples in Imo State are 18.19–40.72 Bq kg$^{-1}$,
Table 1. Soil activity concentration (Bq kg⁻¹) of \(^{40}\text{K}\), \(^{226}\text{Ra}\), \(^{232}\text{Th}\) around solid mineral mining areas in South-Eastern Nigeria.

| S/N | Geographical location | Mining State | Mineral found | \(^{40}\text{K}\) (Bq kg⁻¹) Mean (Range) | \(^{226}\text{Ra}\) (Bq kg⁻¹) Mean (Range) | \(^{232}\text{Th}\) (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 | \(100.22 \pm 8.20\) | \(33.15 \pm 3.31\) | \(77.31 \pm 6.10\) |
|-------------------|----------------------|---------------------|----------------------|
| Control (Mean)    | 28.60                | 19.63               | 25.92                |
| Commended Value [44] | 400                  | 3                   | 30                   |
Table 2. Risk Estimate for workers in solid mineral mining site.

| Variables       | K-40 (Bq kg$^{-1}$) | Ra-226 (Bq kg$^{-1}$) | Th-232 (Bq kg$^{-1}$) | Raeq (Bq kg$^{-1}$) | D (Gy h$^{-1}$) | $\eta Gy h^{-1}$ | AEDE (µSv y$^{-1}$) | AGED (µSv y$^{-1}$) | Hex | Hin | $\xi$ | ELCR $\times 10^{-6}$ |
|-----------------|---------------------|-----------------------|-----------------------|---------------------|----------------|----------------|---------------------|---------------------|-----|-----|-----|------------------------|
| 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                  |
| Skewness        | 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          | 3.26          | 3.67                | 3.67                | 3.34 | 3.48 | 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) | \(D_{ex}\) (mSv y⁻¹) | \(D_{inh}\) (mSv y⁻¹) | \(D_{org}\) (Sv y⁻¹) | Total Eff. Dose (mSv 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               |

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 127.08–289.79 Bq 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 [50]. 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 were 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 [4]. 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 [34,40,51].

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 [26]. 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 [45]. 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 [37]. 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 [34,40,51,65]. 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 [26,32,33,52–57].

### 3.2.2. Radiological hazard parameter statistical analysis

Table 2 shows the summary of the result of the statistically analysed specific activity levels and radiation hazard/ risk indices. The eight radiation risk parameters were computed using reported standard and internationally established equations in literatures [5,43,44,46,58,59]. 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 solid mineral mine site in south-western Nigeria and sampled soil valued obtained in some cities and towns in Nigeria [4,5,10,12,32,34,37,39]. The absorbed dose rate (\(D\)) has a minimum value of 38.96 nGy h\(^{-1}\) and a maximum measured value of 133.00 nGy h\(^{-1}\) with 68.40 nGy h\(^{-1}\) being the mean. The mean absorbed dose value recorded exceeded the recommended worldwide ambient value of 59 nGy h\(^{-1}\). The outdoor Annual Effective Dose Equivalent (\(AEDE\)) has its values varied from 48.26 to 160.78 µSv\(^{-1}\) with the mean and mode values of 83.65 and 48.26 µSv\(^{-1}\), respectively, across the study-area and a standard deviation value of 32.66 µSv\(^{-1}\). The estimated average value of the
outdoor Annual Effective Dose Equivalent of 83.65 $\mu$Sv$^{-1}$ 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 $\mu$Sv$^{-1}$ for outdoor [4, 12, 39, 66].

The estimated value of the annual gonadal equivalent dose (AGED) varied from 263.62 to 879.51 $\mu$Sv$^{-1}$ with a mean value of 454.70 $\mu$Sv$^{-1}$. The estimated value recorded is above the ambient level recommended world permissible value of 300 $\mu$Sv$^{-1}$ 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 [10, 39, 44, 60]. 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 ($I_{\gamma}$) 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 [2, 60, 61]. 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 ($I_{\gamma}$) value of 1.06 obtained was slightly above the UNSEAR recommended value for the general public [44]. These obtained estimated values of the Excess Life Cancer Risk (ELCR) result ranged from $165.45 \times 10^{-6}$ to $559.31 \times 10^{-6}$ with a regular value of $289.14 \times 10^{-6}$. This mean ELCR value of $289.14 \times 10^{-6}$ obtained is approximately the recommended ambient public permissible limit of $0.29 \times 10^{-3}$ [45]. 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 4, while Figure 5 shows the sequential chart distribution of the three natural radionuclides investigated with the radiological risk parameters examined. It was observed from Figure 4 that the histogram showing the spatial dispersion of specific activities of the three natural radionuclides ($^{40}$K, $^{226}$Ra and $^{232}$Th) in the analysed 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 [62]. From Figure 5, 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 [48]:

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 1.0 mSv y\(^{-1}\) for the public [49].

Table 3 of the Occupational Risk Estimate indicates that the external exposure to gamma radiation (\(D_{\text{ext}}\)) has the highest Occupational risk ranging from 0.51 mSv y\(^{-1}\) to 1.3 mSv y\(^{-1}\), followed by Internal exposure from inhalation of radiation from solid mineral dust and contaminated air (\(D_{\text{inh}}\)) 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 (\(D_{\text{ing}}\)) ranging from 0.08 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.

**Figure 4.** Histogram and Skewness plot of some radiation parameters.
4. 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 deployed 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 values 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 being 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

Figure 5. Sequence chart of the statistical analysis.
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. Egariewe, while Dr. Mohammed S. Chaanda did the geological identification and characterisation 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 analysed during the current study are not publicly available, but will be made available from the corresponding author on reasonable request.
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