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Radiological Impact Assessment of Mining on Soil, Water and Plant Samples from Okobo Coal Field, Nigeria

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Abstract

Anthropogenic, including mining activities leaves the environment contaminated with potentially toxic substances and remote hazards if not properly checked. The quest in this study is the levels of naturally occurring radionuclides in Okobo coal, Nigeria and their possible distribution in coal mine vicinity soils, water and plants (cassava). Samples were characterized for levels of radionuclides and radiological detriments using high resolution gamma spectrometer, Gamma ray liquid scintillation and applicable radiological hazard indices. The range of mean activity concentrations ($Bq \cdot kg^{-1}$) for the environmental samples are as follows: ^{226}Ra (8.39 ± 1.0 to 77.6 ± 4.0), ^{232}Th (0.470 ± 0.4 and 77.8 ± 2), and ^{40}K (29.1 ± 0.4 and 289 ± 6), with their respective mean values of 32.7 ± 2.1, 54.0 ± 1.5 and 158.8 ± 3.1 ($Bq \cdot kg^{-1}$). Radiological detriments including radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), internal hazard index (H_{in}), radioactivity level index or gamma index (I_γ) and the ELCR for coal sample is 96.94 $Bq \cdot kg^{-1}$, 0.26, 0.30, 0.69, and 1.56 respectively. Reported values were below the safety limits stipulated by UNSCEAR and implied that the environment is relatively safe with low levels of natural radioactivity. Overall, this background study has demonstrated that Okobo coal mine is a less radio-hazard contributor to environmental samples. Exceptions to this generalization are representative gamma index (I_{yr}) and annual effective dose equivalent values for some cassava and soil samples, which may call for future impact monitoring.

Keywords

Coal, Radiological Detriments, Hazard, Okobo,
Activity Concentrations Gamma-Ray Spectrometry

1. Introduction

In spite of its abundant deposits in Nigeria, Coal does not presently contribute to Nigeria's electricity generation. The problem with mining activities in Nigeria is the negligence of the stakeholders (miners, businessmen and the government) to provide best global mining practices.

The unsuspected populace that constitute these communities where coal mining activities take place, do not know their environmental obligations under the minerals and mining act. This affects their rights including right to education, infrastructures, life, security, health, adequate standard of living, liberty (Sambo et al., 2012).

Threats, linked to the residual and temporal side effects of coal mining on the environmental and agricultural dimensions are well documented (Bergh et al., 2011; Odunayo et al., 2016). There are strong indications of coal's impact on human health and the environment. Air pollution generated by coal mining and combustion in power plants (Din et al., 2013) can affect the environment and human health. Key pollutants from coal mining of adverse impacts on the environment and health include oxides of C, S, and N. Others include particulate matter, heavy metals (Ikwuagwu, 2017) and organic pollutants (PAHs, PCBs among others) even at low concentration pollute the environment.

Coal mining impact on groundwater quantity and quality, as it also impacts on river flows and consequential impact on agricultural land uses cannot be overemphasized (Ikwuagwu, 2017). Environmental impacts for coal mining range from mining subsidence, changes in ground water regimes and mining hydrology. There is also the release of methane into the atmosphere, the release of contaminated water and the generation of solid waste products.

Naturally occurring radionuclides are reported in varying proportions in rocks and soil of several geo-formations, including coal mine. These radionuclides may be deposited in sediment and dissolve into drinking water, thereby leading to human exposure (Raymond et al., 2013).

Uranium (^{238}U), Thorium (^{232}Th) and their decay products (^{226}Ra, ^{212}Pb, etc.) and Potassium isotope (^{40}K) are the natural radionuclides observed as inherent soil contents with known contribution to the radiation exposure and emission of gamma ray and subsequent ingestion through inhalation and the food chain (Mujahid & Hussain, 2011).

The potential radiological hazards parameters investigated in this study include the radium equivalent activity (Ra_{eq}), the annual effective dose rate (H_R), external hazard (H_{ex}) and internal hazard (H_{in}) indices, Representative Gamma Index ($I_{\gamma r}$) and Excess Lifetime Cancer Risk (ELCR). Others (not considered in this study) are the activity utilization index (I), and the alpha index (I_α). The knowledge of radionuclides distribution and radiation levels in the environment is important for assessing the effects of radiation exposure due to both terrestrial and extra-terrestrial sources (Rajesh et al., 2013). If the values of the hazard indices are below the recommended levels; therefore, it is concluded that the environment is relatively safe.

2. Study Area

Okobo community, is a small town in Enjema district of Ankpa Local Government Area (7˚22'14"N, 7˚37'31"E) in Kogi state with reserves of up to 380 million tonnes of coal.

Studies on radiological hazards and detriments are widely documented (Bachama et al., 2017; Alausa et al., 2019; llori & Alausa, 2019). In this study, we focus on the levels of radiation exposure due to the natural radionuclides (^{238}U, ^{232}Th and ^{40}K) in Okobo coal and their vicinity soils, water and cassava plants. The study, for the first time unveiled the radiological detriments associated with the impact of coal mine activities within the study area.

3. Materials and Methods

Analytical grade reagents were used in this study. Instrument employed for analysis include high resolution gamma spectrometer at the Center for Energy Research and Training (CERT), Ahmadu Bello University Zaria, consisting of high purity germanium (HPGe) detector, by Ortec Inc., connected to an Ortec series multichannel analyzer (MCA) through a preamplifier base and coupled to a personal computer. Water samples were analyzed using the liquid scintillation counter (Tri-CarbLSA1000).

3.1. Sampling

Water sampling at Okobo coal field was carried out from the months of November to December, 2018 and January, 2019. Method, described in **Table 1** for the first stage of this study (Itodo et al., 2020) was followed. Water samples were collected at a depth of 2 - 10 cm at the water surface using a 1litre plastic container with a screw cap, with respect to the coal mining operation site. Water samples were collected between 7 am and 10 am) in triplicates from 3 locations (50 cm upstream, midstream and downstream) with pre-cleaned glass bottles. The samples in 1-L amber bottles were adjusted to pH < 2 using 6 M of hydrochloric acid (Adeniji et al., 2019).

Table 1. Sampling codes and description.

S/N	Sample ID	Description
1	OC	Okobo coal
2	SS	Soil Sample
3	CSS	Control soil sample
4	WS	Water sample
5	CWS	Control water sample
6	CS	Cassava sample
7	CCS	Control cassava sample

Soil sampling method reported by Usikalu et al. (2017) was adopted. The vicinity of Okobo coal mine was divided into four quadrants. Soil samples were obtained from each of these quadrants which was 200 m apart from one another and the harmonized into a representative sample. The samples were collected by digging the ground to at least 3 cm. Five soil samples were taken from different points at each location, using composite sampling method. The samples were then kept in airtight bags and labelled accordingly. The samples were dried at 110˚C in an oven to ensure complete removal of moisture until constant weight was attained. Dried samples were grinded and passed through a 2 mm sieve. 100 g of each sample were placed in a radon-tight plastic vessel. The vessels were weighted and sealed for 30 days to allow equilibrium in the ^{238}U and ^{232}Th with their respective progeny.

Coal collection was based on methods documented by Odunayo et al. (2016) and Querol et al. (1996) were carefully followed. A sample grid was established in which five samples of mass 20 g of coal was obtained from a split which was taken each 50 m away from the grid and then harmonized as one sample by means of a hand trowel and a hammer at the coal site. A gross sample of 60 g was obtained after homogenisation of the five samples. The gross coal samples were air-dried, milled and split carefully to obtain a representative 10 g sub-sample of particle size < 250 μm. Coal sample preparation involved crushing, screening and storage. In crushing of coal, samples were reduced to smaller sizes by mechanical means, followed by pulverising in a rotary mill. The powdered samples were passed through a 210 micron sieve to prepare a representative 50 g sub-sample for analysis. The sample was tagged as Okobo coal (OC).

Cassava samples were randomly collected as donations based on farm basket approach. Control samples for the duo (soil and cassava) were collected from Ogaji district (6 - 7 km away from Okobo), following similar protocol. Control water sample is a commercial and registered potable water.

3.2. γ-Ray Spectrometric NaI(Tl) Analysis

The soil, coal and cassava samples were dried and crushed to fine powder with the use of pulverizer. Packaging of the samples into radon impermeable cylindrical plastic containers was selected based on the space allocation of the detector vessel. To prevent radon-222 escape, the packaging in each case was triple sealed, which included smearing of the inner rim of each container lid with Vaseline jelly, filling the lid assembly gap with candle wax to block the gap between lid and container and tight-sealing lid container with masking adhesive tape. Radon and its short lived progenies were allowed to reach secular radioactive equilibrium by storing the samples for 30 days prior to gamma spectroscopy measurement.

The γ-ray spectrometer NaI(Tl) is equipped with NaI(Tl) detector that measures the natural radio-nuclides activity (count rate in the environmental samples). It was calibrated using known source such as ^{60}Co and ^{137}Cs point sources. In order to calculate the radionuclide activity concentration (activity per unit mass)

for each gamma ray photo-peak, we rely on the secular equilibrium between parents and daughters in the samples, the Equation (1) was used (Alashrah & Taher, 2017).

The analysis was carried out using a 76 × 76 mm NaI (Tl) detector crystal optically coupled to a photomultiplier tube (PMT). The assembly has a preamplifier incorporated into it and a 1 kilovolt external source. The detector is enclosed in a 6 cm lead shield with cadmium and copper sheets. This arrangement is aimed at minimizing the effects of background and scattered radiation. The samples were measured for a period of 29,000 seconds, for each sample. The peak area of each energy in the spectrum was used to compute for the activity concentrations in each sample by the use of the following equation:

$$C\left(\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right)=\frac{C_n}{C_{fk}} \tag{1}$$

where:

C = activity concentration of the radionuclides in the sample given in $\mathrm{Bq}\cdot\mathrm{Kg}^{-1}$;

C_n = count rate (counts per second).

$$\text{Count per second}\left(\text{cps}\right)=\frac{\text{Net Count}}{\text{Live Time}} \tag{2}$$

C_{fk} = Calibration factor of the detecting system.

The measuring time for gamma-ray spectra was 43,200 s. To obtain the same gamma dose rate, the activity concentration from the three radionuclides assuming to be 370 $\mathrm{Bq}\cdot\mathrm{kg}^{-1}$ from ^{226}R, 259 $\mathrm{Bq}\cdot\mathrm{kg}^{-1}$ from ^{232}Th and 4810 $\mathrm{Bq}\cdot\mathrm{kg}^{-1}$ from ^{40}K. This is the definition of radium equivalent and is given as Equation (3):

$$Ra_{eq}=A_{\mathrm{Ra}}+1.43\times A_{\mathrm{Th}}+0.077\times A_{\mathrm{K}} \tag{3}$$

where A_{Ra}, A_{Th} and A_{K} are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in $\mathrm{Bq}\cdot\mathrm{kg}^{-1}$, respectively.

3.3. Liquid Scintillation Counting

Water sample (10 mL) was added into a scintillation vial containing 10 mL of installgel scintillation cocktail and analyzed using the liquid scintillation counter (Tri-CarbLSA1000). Initial Counting was carried out immediately the prepared samples were brought to the laboratory. Two litters of water sample treated with 10 mL of concentrated Nitric acid were used for each sample. Evaporation was achieved using beaker (500 mL) set on hot plate at a steady temperature below boiling point to avoid boiling of the water sample. This prevents loss of much residue. The volume evaporated was taken and recorded. After surface drying, residual volume was transferred into Petri-dish for further drying to give final dried residue. Residue was weighed and transferred into planchet, followed by addition of drops of acetone and vinyl-acetate. The vinyl-acetate helps in removing the moisture content and serves as a binder while the acetone was used as sterilizer. Then the sample is ready for counting.

3.4. Calibration and Calibration Efficiency

Table 2 represents the spectral energy calibration data used in the analysis. The beta standards are Strontium-90 beta sources of diameter 38 mm and active film of 12 mg/m^3 thick. The alpha standards are Plutonium-239 alpha sources of diameter 38 mm in an oxidable disc of 3 mm thick. 10 mL of concentrated nitric acid were added to the water sample immediately after collection so as to reduce the pH, minimize precipitation and prevent the absorption on the wall of the container. The calculation formulae for the count rate and activity are:

$$\text{Count rate}:(\alpha,\beta)=\frac{\text{Raw count}}{\text{Count Time}} \tag{4}$$

Calibration of the system for energy and efficiency were done with two calibration point sources, Cs-137 and Co-60. These were done with the amplifier gain that gives 72% energy resolution for the 66.16 Kev of Cs-137 and counted for 30 minutes.

Table 3 stands for quality control parameters for quantitative spectral analysis. The standards used to check for the calibration are the IAEA gamma Spectrometric reference materials RGK-1 for K-40, RGU-1 for Ra-226 (Bi-214 peak) and RTG-1 for Th-232 (Ti-208). The background count rate was done for 29,000 seconds just as those of the samples.

Gamma ray spectroscopic systems were used to analyze the samples collected. The radiation source was placed close to the detector in order to increase the intensity reaching the detector hence; the counting system may exhibit high detection efficiency (Bello et al., 2014). The net count was obtained using the expression:

$$N_S = N_g - N_b \tag{5}$$

where N_s = Sample counts (Net counts), N_b = Background counts, N_g = Gross counts.

Table 2. Spectral energy windows used in the analysis.

Radio-isotope	Gamma Energy (Kev)	Energy Window (Kev)
R-226	1764.0	1620 - 1820
Th-232	2614.5	2480 - 2820
K-40	1460.0	1380 - 1550

Table 3. Quality control parameters for quantitative spectral analysis.

	Calibration Factor			Detection Limit	
Radio-Isotopes	10^{-3} (cps/ppm)	10^{-4} (cps/ppm)	Conversion Factor (Bq·Kg^{-1})	ppm	Bq/kg
^{40}K	0.026	6.431	0.032	454.54	14.54
^{226}Ra	10.500	8.632	12.20	0.32	3.84
^{232}Th	3.612	8.768	4.12	2.27	9.08

The activity concentration ($Bq{\cdot}kg^{-1}$) of each radionuclide was obtained using:

$$\text{Sample activity}(C) = \frac{N_s}{\varepsilon ytm} \tag{6}$$

where ε = efficiency detector, y = gamma yield, m = mass of the sample, t = counting time.

4. Results and Discussion

4.1. Visual Inspection of Samples

The sampling sites for cassava, soil and water samples were presented in Figure 1. The physical outlay of cassava farm land from Okobo coal-mining areas physically shows varieties of stunted growth of cassava samples. Outburst of suspected polluted water from the coal mining regions in both downstream and upstream water samples from Okobo vicinity were well observed as coloured due to coal deposits and leachates. This is the only source of water for industrial, agricultural and domestic uses in Okobo.

4.2. Radiological Study

Results of radiological study for soil, coal and cassava samples from Okobo coal mine is presented in Tables 4-6. Table 4 shows the K-40 radiological data of water samples measured with liquid scintillation counter. K-40 counts do not follow a regular trend. Values recorded for upstream water samples (49.95 - 111.32 cpm) exceed those reported for mid and down streams. This is an indication that the upstream water is more contaminated with K-40 radionuclide.

4.3. Activity Concentrations of K-40 in Water Samples

The radiological analysis of water samples by Gamma ray liquid scintillation at 60-minute count rate shown in Table 4 was converted to activity concentrations (Table 5) using the conversation factor of $cps/Bq{\cdot}Kg^{-1}$ = 0.000643 for K-40. Table 5 revealed that the control water sample (CWS1 and CWS2) has high K-40 content, with mean activity concentration range of 96.15 - 1129.69 $Bq{\cdot}kg^{-1}$. Next to this value is the upstream K-40 mean activity concentration of 934.86 $Bq{\cdot}kg^{-1}$. The trend of k-40 activity concentration follows the order of US > MS > DS as shown in Figure 2. K-40 isotope values estimated for the analytical water samples are below those recorded for the IAEA reference water samples (785 - 1150.51 $Bq{\cdot}kg^{-1}$).

(a) (b) (c)

Figure 1. Study area showing (a) Cassava plants (with Stunted Growth), (b) Outburst of water from coal mine site, (c) Downstream water samples in Okobo.

Table 4. Radiological data (counts per minutes) of K-40 in water samples from Okobo coal mine, using gamma ray liquid scintillation counter.

S/n	Sample I.D	Radiological Data (cpm)		
		Channel A	Channel B	Channel C
1	CWS 1	66.08	61.17	134.25
2	CWS 2	54.22	48.95	108.90
3	US	55.13	49.95	111.32
4	MS	47.85	43.42	96.33
5	DS	47.98	39.07	91.08
6	BW	64.22	45.57	117.22
7	IAEA 423(R1)	39.65	49.27	92.93
8	IAEA427(R2)	70.83	61.57	133.92
9	IAEA 431(R3)	53.08	50.52	106.43

CPM/K = K-40 Count Per Minute Channel A, IAEA423 = International Atomic, Energy Agency 423 Calibration Factor of Liquid Scintillation's counter is 13.37, BW= Background water.

Table 5. Activity concentrations of K-40 in water samples.

S/n	Sample id	Activity concentration ($Bq \cdot kg^{-1}$)			
		Channel A	Channel B	Channel C	Mean values
1	CWS 1	856.40	792.77	1739.89	1129.69
2	CWS 2	702.70	634.40	1411.35	916.15
3	US	714.49	647.36	1442.72	934.86
4	MS	620.14	562.73	1248.44	810.46
5	DS	621.82	506.35	1180.40	769.52
6	BW	832.30	590.59	1519.18	980.69
7	IAEA 423 (R1)	513.87	638.54	1204.38	785.60
8	IAEA427 (R2)	917.96	797.95	1735.61	1150.51
9	IAEA 431 (R3)	687.92	654.74	1379.34	907.33

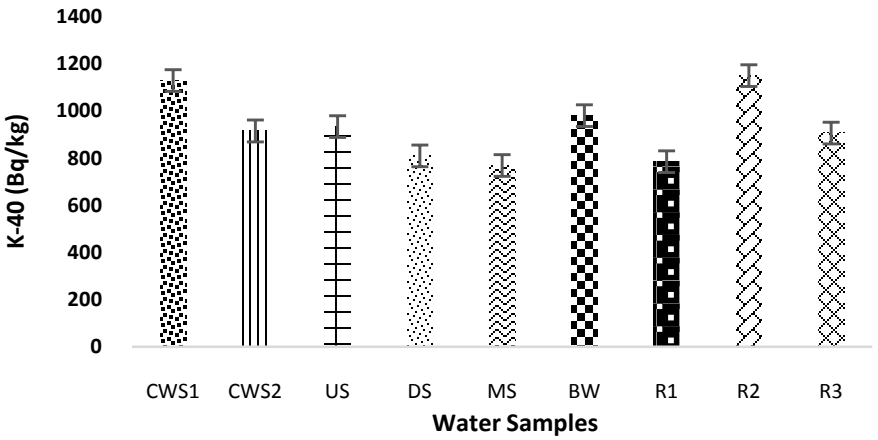

Figure 2. Mean activity concentrations (Bq/Kg) of K-40 in water samples from Okobo coal mine.

Table 6 represents the radiological data (counts) for Okobo coal, soil and cassava samples. Activity concentration levels of naturally available K-40, Ra-226 and Th-232 were presented in Table 7. This study shows that the control soil from

Table 6. Radiological data (counts per second) for soil, coal and cassava samples from Okobo coal Mine site.

S/N	Sample ID	Radiological Data (cps)					
		K-40	SD	Ra-226	SD	Th-232	SD
1	CSS	0.0452	0.006	0.0983	0.0022	0.0010	0.0008
2	SS 1	0.0948	0.0010	0.0232	0.0034	0.0433	0.0022
3	SS 2	0.1208	0.0017	0.0146	0.0020	0.0466	0.0019
4	SS 3	0.0846	0.0022	0.0184	0.0021	0.0682	0.0014
5	SS 4	0.0774	0.0012	0.0669	0.0010	0.0545	0.0011
6	SS 5	0.0693	0.0030	0.0376	0.0025	0.0660	0.0013
7	CCS	0.1493	0.0032	0.0433	0.0010	0.0534	0.0011
8	CS 1	0.0733	0.0026	0.0078	0.0012	0.0560	0.0010
9	CS 2	0.1860	0.0026	0.0415	0.0035	0.0449	0.0015
10	CS 3	0.1662	0.0033	0.0072	0.0016	0.0583	0.0012
11	CS 4	0.1609	0.0060	0.0951	0.0023	0.1113	0.0023
12	CS 5	0.1187	0.0039	0.0123	0.0013	0.0316	0.0013
13	OC	0.1014	0.0012	0.0134	0.0022	0.0424	0.0017

CPS = Count per second; Bq/Kg = Becquerel per Kilogram; Calibration Factors: $\times 10^{-4}$; Conversion Factor: cps/Bq·Kg^{-1}; K-40 = 0.000643, Ra-226 = 0.000643, Th-232 = 0.000877.

Table 7. Activity concentrations of samples from Okobo coal mine vicinity.

S/N	Sample ID	Activity concentrations (Bq·kg^{-1})					
		K-40	SD	Ra-226	SD	Th-232	SD
1	CSS	29.07	0.35	47.10	1.00	0.47	0.35
2	SS 1	147.48	1.61	26.93	4.00	49.35	2.52
3	SS 2	187.91	2.63	16.86	2.36	53.08	2.20
4	SS 3	131.60	3.38	21.38	2.48	77.77	1.57
5	SS 4	120.45	1.82	77.56	1.20	62.12	1.22
6	SS 5	107.74	4.72	43.55	2.88	75.26	1.53
7	CCS	232.26	4.99	50.23	1.16	60.91	1.26
8	CS 1	113.96	3.97	9.03	1.44	63.85	1.10
9	CS 2	289.32	4.02	48.11	4.04	55.79	1.73
10	CS 3	258.54	5.09	8.39	1.80	66.49	1.38
11	CS 4	103.55	0.39	45.58	1.12	52.53	1.10
12	CS 5	184.53	6.01	14.26	1.52	36.02	1.53
13	OC	157.67	1.88	15.58	2.60	48.40	1.97
	Min	29.07	0.35	8.39	1.00	0.47	0.35
	Max	289.32	6.01	77.56	4.04	77.77	2.52
	Mean	158.78	3.14	32.66	2.12	54.00	1.50

CPS = Count per second, Bq/Kg = Becquerel per Kilogram, Calibration Factors: $\times 10^{-4}$, Conversion Factor: cps/Bq·Kg^{-1}, K-40 = 0.000643, Ra-226 = 0.000643, Th-232 = 0.000877.

the non-coal region records the least activity concentrations of K-40 (29.07), Ra-226 (47.18) and Th-232 (0.47) Bq·kg^{-1}. A corresponding high level was observed for control cassava samples. This could be attributed to the soft cassava tissue and their affinity for the sorption of radionuclides at short period.

4.4. Activity Concentrations of Radionuclides in Coal Sample

The range of activities obtained for the studies showed the mean concentrations as: ^{226}Ra (8.39 ± 1.00 to 77.56 ± 4.04 Bq·kg^{-1}), ^{232}Th (0.47 ± 0.35 to 77.77 ± 2 Bq·kg^{-1}) and for ^{40}K (29.07 ± 0.35 to 289.32 ± 6.01 Bq·kg^{-1}), with respective mean values of 32.66 ± 2.12, 54.00 ± 1.50 and 158.78 ± 3.14 Bq·kg^{-1} respectively. The coal sample itself has values of 15.58 ± 2.60 Bq·kg^{-1} for ^{226}Ra, 48.40 ± 1.97 Bq·kg^{-1} for ^{232}Th and 158.78 ± 3.14 Bq·kg^{-1} for ^{40}K. The activity concentrations of ^{226}Ra is below the 20 Bq·kg^{-1}, while those of ^{232}Th, and ^{40}K are above the 20 Bq·kg^{-1} and 50 Bq·kg^{-1} above the world average values for coal (UNSCEAR, 1982).

4.5. Radiological Data for Soil and Cassava Samples

Figure 3 indicted activity concentration trend of K-40 > Th-232 > Ra-226. The affinity of the control (non-coal region) cassava sample for Th-232 is higher compared to the corresponding soil sample, CSS. The high radionuclides activity concentrations of analytical samples compared to the coal itself could be attributed to bioaccumulation and sample size considered in the study.

4.6. Hazard Indices

Table 8 represent the radiological detriments attributed to radionuclides estimated in Okobo coal and their near environmental samples. Table 8 contains calculations of Radium equivalent activity (Ra_{eq}), External hazard index (H_{ex}), internal hazard index (H_{in}), Representative gamma index (I_{yr}) and Excess lifetime cancer risk (ELCR).

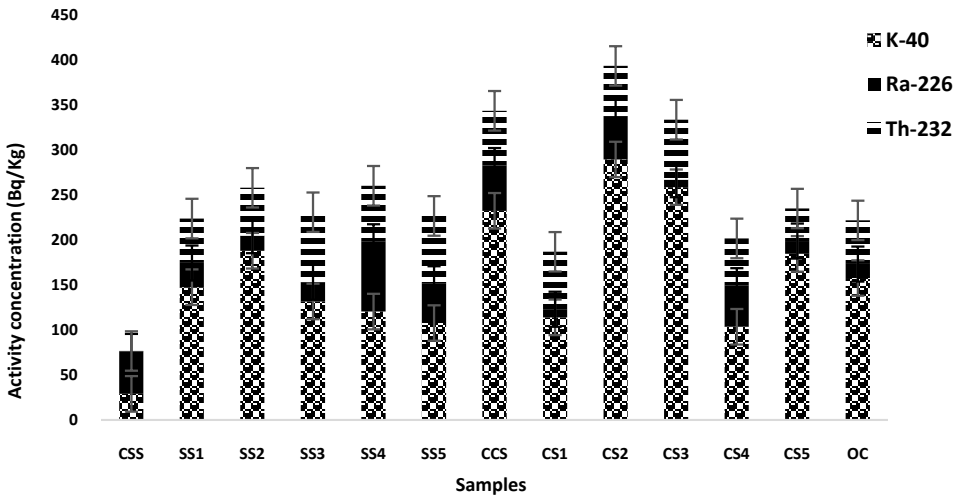

Figure 3. Mean activity concentrations (Bq/Kg) of radionuclides (^{40}K, ^{226}Ra and ^{228}Th) of coal, soil and plant samples from Okobo coal mine site.

4.6.1. Radium Equivalent Activity (Ra_{eq})

^{226}Ra, ^{232}Th, and ^{40}K are not uniformly distributed in most environmental samples. This is shown in their respective radio hazard indices. A single parameter known as Radium equivalent activity (Ra_{eq}) in Table 8 is defined with respect to radiation exposure which compares the activity of materials containing different elements of primordial radionuclides. Its definition also considers external and internal effective dose from radon and its decay progeny (Huang et al., 2015). Ra_{eq} is measured in Bq·kg^{-1} and defined based on the assumption that specific activity of 370 Bq·kg^{-1} for ^{226}Ra uniformly distributed in any environmental sample can result in annual effective dose of 1 mSv at 1 m above ground level (Huang et al., 2015; Taskin et al., 2009). It is quantitatively expressed as (Beretka & Mathew, 1985; UNSCEAR, 2000).

$$Ra_{eq}\left(\mathrm{Bq}\cdot\mathrm{kg}^{-1}\right) = A_{\mathrm{Ra}} + 1.43A_{\mathrm{Th}} + 0.077A_{\mathrm{K}} \tag{7}$$

where A_{Ra}, A_{Th} and A_{K} are the respective specific activities of ^{226}Ra, ^{232}Th, and ^{40}K. The constants; 1, 1.43, and 0.077, represents the activity conversion rates for ^{226}Ra, ^{232}Th, and ^{40}K in sequence, which result in gamma dose rate at maximum permissible Ra_{eq} of 370 Bq·kg^{-1}. The result of Ra_{eq} (Bq·kg^{-1}) in this study (Table 8) revealed the contributions of coal mining to vicinity environmental samples. The values reported for CSS (50.01 Bq·kg^{-1}) is far less than those of the analytical soil samples (107.24 - 175.67 Bq·kg^{-1}) as well as their corresponding cassava samples (79.98 - 150.17 Bq·kg^{-1}). Figure 4 unveiled the difference between the Radium activity and detriment in cassava as higher compared to soils.

Table 8. Radiological detriments (Hazard Indices) of coal, soil and cassava samples from Okobo mine site.

S/N	Sample ID	Sample ID				
		Ra_{eq} (Bq·kg^{-1})	H_{ex}	H_{in}	I_{yr}	ELCR (×10^{-3})
1	CSS	50.01	0.14	0.26	0.34	0.89
2	SS 1	108.85	0.29	0.37	0.77	1.77
3	SS 2	107.24	0.29	0.34	0.77	1.73
4	SS 3	142.73	0.39	0.44	1.01	2.25
5	SS 4	175.67	0.47	0.68	1.22	2.89
6	SS 5	159.47	0.43	0.55	1.11	2.55
7	CCS	155.20	0.42	0.55	1.10	2.56
8	CS 1	109.12	0.29	0.32	0.77	1.71
9	CS 2	150.17	0.41	0.54	1.07	2.50
10	CS 3	123.38	0.33	0.36	0.89	1.98
11	CS 4	128.67	0.35	0.47	0.90	2.09
12	CS 5	79.98	0.22	0.25	0.58	1.31
13	OC	96.94	0.26	0.30	0.69	1.56
	Min	50.01	0.14	0.25	0.34	0.89
	Max	175.67	0.47	0.68	1.22	2.89
	Mean	122.11	0.33	0.42	0.86	1.98

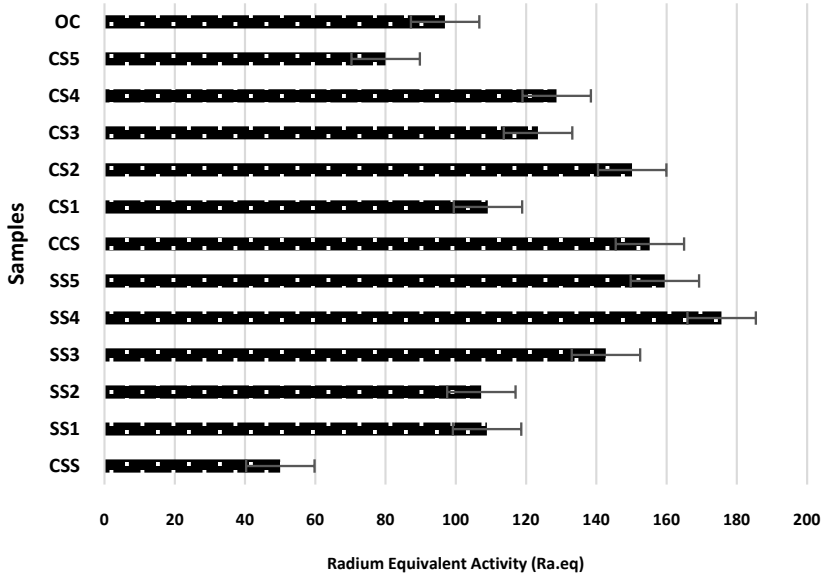

Figure 4. Radium equivalent activity (Ra_{eq}) of coal, soil and plant samples from Okobo coal mine site.

4.6.2. External Hazard Index (H_{ex})

Radiation hazard incurred due to external exposure to gamma rays is quantified in terms of the external hazard index (H_{ex}). The maximum permissible value for H_{ex} is unity, which corresponds to Ra_{eq} upper limit of 370 Bq·kg^{-1} (Huang et al., 2015; Oni, 2019). H_{ex} is calculated from the equation:

$$H_{ex} = \frac{A_{\mathrm{Ra}}}{370} + \frac{A_{\mathrm{Th}}}{259} + \frac{A_{\mathrm{K}}}{4810} \tag{8}$$

where A_{Ra}, A_{Th}, and A_{K} are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. It is assumed that 370 Bq·kg^{-1} of ^{226}Ra, 259 Bq·kg^{-1} of ^{232}Th, and 4810 Bq·kg^{-1} ^{40}K, produce the same gamma dose rate (Lu et al., 2006; Lu et al., 2012). Values of Hex (0.14 - 0.47) computed for coal, soil and cassava samples in this study were below unity, hence within the permissible limit with sample tagged as non-hazardous with reference to radionuclides.

4.6.3. Internal Hazard Index (H_{in})

The internal radiation exposure is quantified by the internal hazard index (Hin) given by UNSCEAR (Gawlik & Bidoglio, 2006).

$$H_{in} = \frac{A_{\mathrm{Ra}}}{185} + \frac{A_{\mathrm{Th}}}{259} + \frac{A_{\mathrm{K}}}{4810} \tag{9}$$

where, A_{Ra}, A_{Th}, and A_{K} are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. UNSCEAR (2000) provided that the value of the above indices must be less than unity for the radiation hazard to be regarded as insignificant as exemplified in this studies. H_{in} values ranged from 0.25 - 0.68 across soil, coal and cassava samples within the Okobo coal field and environment.

4.6.4. Representative Gamma Index ($I_{\gamma r}$)

The representative gamma index ($I_{\gamma r}$) is a screening parameter for materials of

possible radiation health challenge (Jibiri & Okeyode, 2012). It is calculated using the equation (Ravisankar et al., 2014).

$$I_{\gamma r} = \frac{A_{\mathrm{Ra}}}{150} + \frac{A_{\mathrm{Th}}}{100} + \frac{A_{\mathrm{K}}}{1500} \tag{10}$$

where A_{Ra}, A_{Th}, and A_{K} are the specific activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bq·kg^{-1}. The numerical denominators of 150, 100, and 1500, are specific exposure rates for ^{226}Ra, ^{232}Th, and ^{40}K, respectively that yield a sum of $I_{yr} < 1$, which corresponds to an annual effective dose of <1 mSv, to satisfy the dose criteria (Manigandan & Chandar-Shekar, 2014; Taskin et al., 2009). Samples estimated in this study gave Representative Gamma Index values, ranging betwee 0.34 - 1.22. This implied that there could be possible radiation health challenges resulting from some samples whose annual gamma effective dose tends to unity or <1 mSv. On the average, I_{yr} values for this study is 0.86.

4.6.5. Excess Lifetime Cancer Risk (ELCR)

Excess life-time cancer risk (ELCR) was estimated from annual effective dose equivalent using the equation (Ravisankar et al., 2014; Taskin et al., 2009)

$$\mathrm{ELCR} = \mathrm{AEDE} \times \mathrm{DL} \times \mathrm{RF} \tag{11}$$

where AEDE, DL, and RF are the annual effective dose equivalent, duration of life (70 years), and risk factor (0.05 Sv^{-1}), respectively (Ravisankar et al., 2014). This parameter defined the risk factor as fatal cancer risk per Sievert, which according to Taskin et al. (2009) is assigned a value of 0.05 by ICRP 60 for the public for stochastic effects. **Figure 5** of this study presents low ELCR values (0.89×10^{-3} to 2.89×10^{-3}) were reported and implied the coal mine at Okobo poses no fatal cancer risk to the Okobo populace at the time of this study.

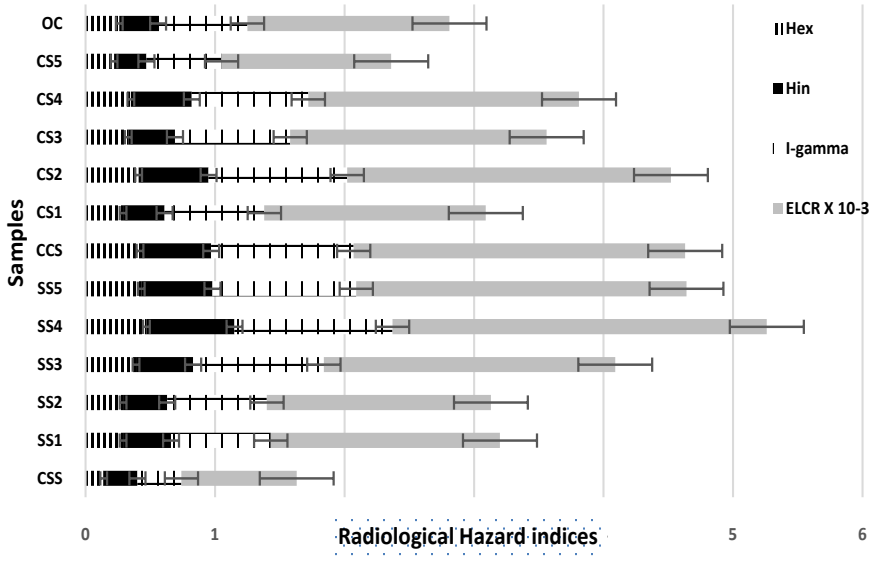

Figure 5. Radiological detriments of coal, soil and plant samples from Okobo coal mine site.

4.6.6. Absorbed Gamma Dose Rate (*D*)

This is a measure of the energy deposited in a medium by ionizing radiation. In the SI system of units, the unit of measure is joules per kilogram, and its special name is gray (Gy). Absorbed dose is used in the calculation of dose uptake in living tissue in both radiation protection and radiology. It is also used to directly compare the effect of radiation on inanimate matter.

The measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were converted into doses by applying the conversion factors 0.462, 0.604 and 0.0417 for uranium, thorium and potassium, respectively. These factors were used to calculate the total dose rate (nGy·h^{-1}) using the following equation:

$$D = 0.462A_{\mathrm{Ra}} + 0.604A_{\mathrm{Th}} + 0.0417A_{\mathrm{K}} \quad (12)$$

where, A_{Ra}, A_{Th} and A_{K} has been previously defined. In this study, absorbed gamma dose rate deduced for the cassava sample is in the range of 36.04 to 69.68 nGy·h^{-1}, with the control sample having the highest value.

4.6.7. Annual Effective Dose (AED)

The annual effective dose rate outdoors in units of (μSv/year) was calculated by the following formula:

$$\mathrm{AED} = \text{Absorbed dose}\left(\mathrm{nGy}\cdot\mathrm{h}^{-1}\right)\times 8760\ \mathrm{h}\times 0.7\ \mathrm{Sv}\cdot\mathrm{Gy}/\mathrm{year}\times 0.2\times 10^{-3} \quad (13)$$

From Table 9, the AED values for the cassava samples vary from 44.20 - 85.46 μSv/year. Some values for cassava sample (CCS, CS2 and CS4) were above the world average values at 70 mSv/year as observed (Diab, 2008). This calls for concern in some samples. On a mild note, an average of 68.098 mSv/year which poses no threat is more generalized for the cassava samples within Okobo coal field region.

Generally, the calculated values for Ra_{eq}, external hazard index; H_{ex}, internal hazard index and the H_{in} values for Okobo coal sample itself were 96.94 Bq·kg^{-1}, 0.26 and 0.30, respectively. These values were below the safety limits stipulated by UNSCEAR (2000). These implied that radioactivity of Okobo coal is non-hazardous and within safe limits. Similarly, the calculated mean values (Figure 6) for

Table 9. Absorbed gamma dose (D) and annual effective dose (AED) on cassava samples from Okobo mine site.

S/No	Cassava Sample	Radionuclides			Adsorbed Doses	
		K-40	Ra-226	Th-232	D (nGy·h^{-1})	AED (μSv/year)
1	CCS	232.26	50.23	60.91	69.68	85.46
2	CS 1	113.96	9.03	63.85	47.49	58.24
3	CS 2	289.32	48.11	55.79	67.99	83.38
4	CS 3	258.54	8.39	66.49	54.82	67.28
5	CS 4	103.55	45.58	52.53	57.10	70.03
6	CS 5	184.53	14.26	36.02	36.04	44.20

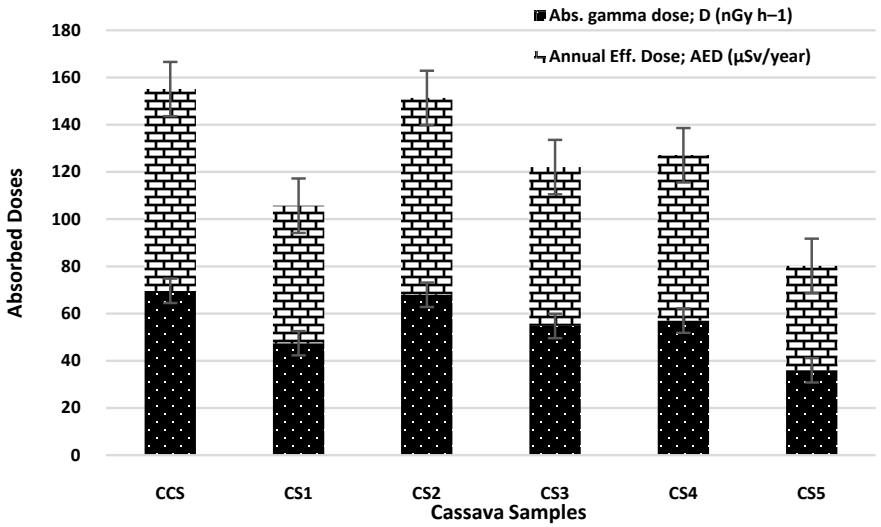

Figure 6. Absorbed gamma dose (D) and annual effective dose (AED) of cassava samples from Okobo mine site.

Radium equivalent activity (Ra_{eq}), External hazard index (H_{ex}) and internal hazard index (H_{in}), for the Okobo Soil Sample, Control Soil Sample, Okobo Harmonised Cassava Sample, Control Cassava Sample Okobo, were all within the desired safe limits of less than 370 Bq·Kg^{-1} (for Ra_{eq}), less than unity (for H_{ex}, H_{in}, and I_{yr}) and below 0.05 for ELCR. Thus, none of these samples pose any radiation risk. On the contrary, Representative gamma index (I_{yr}) and AEDE values for some samples calls for continual radiological impact monitoring.

5. Conclusion

The natural radiological activity concentrations of coal, cassava, water and soil samples within Okobo coal field have been estimated by gamma spectrometry. Radiological detriment and hazard indices values for Ra_{eq}, external hazard index H_{ex}, internal hazard index H_{in}, I_{yr} and the ELCR for Okobo coal and neighbouring environmental samples were below the safety limits stipulated by UNSCEAR. This implied that radioactivity of Okobo coal is non-hazardous and within safe limits for now. Exceptions to this generalization are representative gamma index (I_{yr}) and annual effective dose equivalent values for some samples, requiring continual radiological impact monitoring.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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