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Natural radionuclide concentration and associated health hazard indices in coal samples from Okobo Coal Mine, Nigeria

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Abstract
Background: Mining activities of coal have the capacity to raise the level of natural radionuclide concentration in our environment. This exposes human population to ionizing radiations with their associated health hazards. This study was aimed at measuring natural radionuclide concentration of ^{226}Ra, ^{232}Th and ^{40}K, and their associated health hazard indices in coal samples from Okobo coal mine.
Materials and Methods: The survey design adopted for this study was the cross sectional type. Coal samples totaling ten (10) in number, were randomly collected from the coal mine. The sampled coal was analyzed at the University of Ibadan, in the laboratory of the National Institute of Radiation Protection and Research (NIRPR), using high resolution spectrometry technique, with high purity germanium (HPGe) detectors.
Results: The obtained results revealed that the radionucide concentration of ^{226}Ra in the sampled coal recorded slightly higher value (21.34±7.94 Bq/kg) than global average of 20 Bq/Kg. However, the recorded values for ^{232}Th (13.61±5.08 Bq/kg) and ^{40}K (20.91±15.77 Bq/kg) fall below the global average value of 20 and 50 Bq/Kg respectively. All calculated radiological hazard parameters recorded lower values than the global permissible value but for annual gonadal dose equivalent (AGDE) which recorded significantly higher value (407.74 mSv/yr) than the world acceptable limit of 300 mSv/yr·
Conclusion: The activity concentration of ^{226}Ra for Okobo coal sample recorded a unit mean value higher than the global average, however the recorded values for ^{232}Th and ^{40}K fall below global permissible values. All radiological health hazards recorded lower values than the precautionary safe limit but for annual gonadal equivalent dose which recorded significantly higher value than the global permissible limit. This implies that both staff and residents of the community could be exposed to radiological threat, which might affect the functionality of their sensitive organs if prolonged and cumulative exposure is not avoided.

Keywords: Radionuclide concentration, Coal, Radiological hazard indices, Okobo

Introduction
Coal is one of the nonrenewable energy sources used for industrial purposes such as power generation, transportation, cement production, aluminum, iron and steel production [1]. Among the fossil fuels, coal is the leading source of energy used globally because of its low cost and efficiency [2]. Using coal as a means of power generation contributes about 40.8% of the world's electricity [3], however, such potential has not been fully harnessed in Nigeria. The quest for civilization and industrialization has generally increased the demand for use of coal for power generation world over and has led to continued and increasing need for coal exploitation and utilization. Coal, like most earth materials contain varied level of concentration of natural radioactive isotopes including ^{238}U, ^{232}Th and ^{40}K [4, 5] which are of most concern because of their radio-toxicity. Though their concentrations are generally lower than that of the earth crust, higher concentrations can be found sometimes as a result of leaching from uranium and thorium – rich volcanic rocks [6]. Their concentrations are generally dependent on geological formation of the coal and are closely related to the overall radioactivity of the earth crust [3, 7, 8, 9].

Mining and processing activities of coal are associated with enhancement and redistribution of natural radionuclide materials in the environment, leading to increased background radiation from terrestrial sources. Coal mining operations commenced in Okobo community in the year 2011 with estimated coal reserve of over 380 million tones [10]. The underlying goal was to provide alternative source of power generation by using coal as the main source of fuel to drive industrial processes. However, the environmental degradation and radiological threat associated with coal exploitation and utilization cannot be ignored [11]. Series of coal mining activities beginning from its excavation to loading and unloading produces radiation emitting dusts which has a direct negative impact on the ecology and heath of the local population [12]. These activities have the capacity to relocate and redistribute radionuclides in the soil, air and water bodies in and around the mining area [13]. During excavation, drilling and blasting, NORMs are relocated from normally inaccessible locations to areas where humans are present [14]. Dust and inhalable particulate matters are released causing air pollution in the vicinity of the coal mine. Water bodies such as rivers, streams, lakes, and underground water are also not spared from the negative impact of pollution and contamination with possible radiological hazard to consumers of such water [15]. This, thereby raise radiation levels beyond normal background in the environment[8, 16] resulting in increased dose received not only to coal miners but also the residents of the local community with associated health hazard [17, 18, 9]. Coal dusts inhalation in an amount large enough to exceed the clearing function of the lungs can cause some diseases among coal workers such as black lung disease [19, 20, 9]. Potential risk of developing cancer due to external gamma radiation exposure to coal in the mining environment has also been reported among coal miners and human population in the vicinity of mining sites [19, 21, 17, 20].

Although documented reports exist on the radioactivity concentration of some Nigeria coal mines such as the Maiganga coal mine in Gombe State [8], Barakinladi coal mine in Plateau state and Enugu coal mine in Enugu state [16], very few research reports exist about Okobo coal mine. A study conducted to evaluate the outdoor ambient radiation hazard indices in selected solid minerals mining Sites in Kogi state noted that the measured value for background ionizing radiation in the mineral deposition sites, including Okobo was above global permissible standard [22]. But adequate information was lacking with respect to the source of emission and the radionuclides responsible for such high emission in the area. This study was aimed at measuring natural radionuclide concentration their associated radiological health hazard indices in Okobo coal mine. Radiological hazard parameters calculated for this study include Radium equivalent activity (Raeq), Absorbed Dose rate (D), Annual Effective Dose Equivalents and Annual Gonadal Dose Equivalent, Representative Gamma Index (Iγr), External (Hex) and internal (Hin) hazard indices, and Excess Life Time Cancer Risk (ELCR). These were meant to determine the level of threats posed by opencast mining operation in the coal mining site.

Materials and Method

Study Area

Okobo coal mining site is located between latitude N07^0 51' and longitude E07^0 70' in Okobo village of Enjema district of Ankpa local government area in Kogi state. Two neighboring states of Benue and Enugu bordered the area in the east and South respectively. Sandstones, mudstones, shales and sandy shales with coal seams arranged in alternating horizons constitute the geology of the area [23].

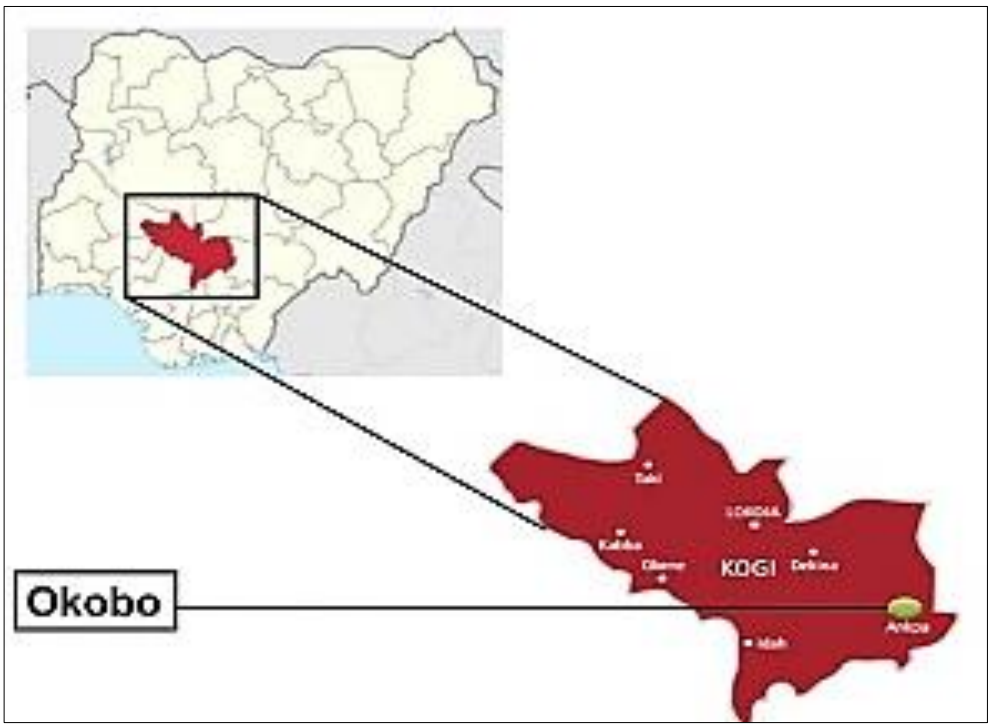

Fig 1: Maps showing the location of the study area

Sample collection and preparation

Coal samples totaling ten (10) in number, were randomly chosen from the Mine. The samples were carefully collected from two stock piles of coal in the mining site using manual sampling method. They were cautiously picked in such a way as to acceptably represent the entire coal lot in the

mining site. In the laboratory, the samples were dried in the air for 72 hours. This was done to ensure that regular weight of the sample was obtained before counting [24]. The dried samples were further crushed and reduced to fine particles. The fine particles were made uniform using a sieve of about 0.2 mm mesh screen. This also helped to get rid of foreign bodies within the sample. About 400 g of the uniform samples were carefully measured and poured into calibrated 500 ml plastic containers (Marinelli beakers). The counting containers were covered with their lids and properly sealed to ensure there was no room for escape of any radioactive gas, like radon. The sealed samples were allowed to reach circular equilibrium by keeping them away for the period of about 28 days before counting [9].

Gamma Spectrometric Measurements

The measurement of radionuclides concentration in the samples was performed using gamma spectrometry technique as adopted by IAEA technical document [24]. This method was used because of its advantage of being able to directly measure gamma emitters without involving chemical separation. The detector used was a high purity germanium (HPGe) type, because of its unparalleled spectra energy resolution. The relative efficiency of the detector is about 80% with active volume of 172cm3. The detector was connected to 18 k-channel analyzer software called Genie-2000 which was successfully used to analyze the spectra of the samples. The software performs the function of matching various gamma energy peaks to a library of possible radionuclides and hence estimated the concentrations of ^{238}U, ^{232}Th and ^{40}K in the samples. The detector is shielded in a cylindrical container made of lead and iron to reduce gamma ray background interference. Before counting, the background radiation of the laboratory environment was estimated with the same plastic container (Marinelli beaker) to be used in the sample measurement. Each of the samples was counted for a period of 5 hrs [24]. The activity concentration of ^{226}K, ^{232}Th and ^{40}K in Bq/Kg were calculated using the expression [25]:

$$A_{ci} = \frac{NCi}{E \times yi \times M \times T} \quad (1)$$

where A*ci* represents the activity concentration of the ith radionuclide in the sample given in Bq/Kg. NCi on the other hand stands for the net count rate of *i*th radionuclide under the corresponding peak, which is obtained by subtracting the background count value in count per second (cps) from the value of the sample count in count per second (cps). E = Detection efficiency. yi = gamma ray yield per transformation of a particular nuclide. M represents the mass of the measured sample in kilogram (kg), while T stands for the counting time in seconds which in this case was about 5 hrs.

Radiation Hazard Estimation

Based on the estimated activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, from the sampled coal, the health hazard indices to the exposed population were calculated through the following radiation hazard parameters:

Air-Absorbed Dose Rates

Determination of absorbed dose rate in air is the most important step to take when evaluating radiation exposure to a given population emitted by gamma radiation sources [26]18. The exposure to ionizing radiation due to activity concentration of natural radionuclides in soil is given by the absorbed dose rate in the air at one metre above the ground surface/ level. The mean radionuclide concentration of ^{226}Ra, ^{232}Th and ^{40}K in a given samples such as coal is generally used to calculate the absorbed dose rate available in the air. The conversion factors of 0.462, 0.604 and 0.0417 for ^{226}Ra, ^{232}Th and ^{40}K, were used as provides by UNSCEAR [6, 27,28].20

$$D(nGy/h) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_k \quad (2)$$

Where D = dose rate (nGy h^{-1}) at one metre above the surface of the ground due to ^{226}Ra ^{232}Th and ^{40}K in a given samples. $A_{Ra,}$ A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively. The population weighted values of absorbed dose rate in air outdoor from terrestrial gamma radiation is given a value of 59 nGyh^{-1}[29]32.

Radium Equivalent Activity (Ra_{eq})

Radium equivalent activity (Ra_{eq}) is one of the radiation hazard indices often used to estimate the output of gamma radiation from varying mixtures of ^{226}Ra, ^{232}Th and ^{40}K in different samples [30, 31] This allows comparison of the specific activity of materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K to be calculated through the relation according to Beretka and Mathew [27] and UNCEAR [6] as follows;

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (3)$$

where, A_{Ra}, A_{Th} and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively in Bqkg^{-1}.This will be calculated based on the estimation that 370B q/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K are all producing the same gamma ray dose rate [25]. The maximum value of radium equivalent activity (Raeq) must be equal to or less than 370 Bq/kg in order to keep the external dose less than 1.5mSv [32].

Annual Effective Dose Equivalent (AEDE)

The absorbed dose in the air at 1 meter above the ground surface does not directly provide any idea about the radiation hazard to which an individual is exposed. The annual effective dose equivalent was estimated from the absorbed dose rate by applying dose conversion factor of 0.7 Sv/Gy as recommended by UNSCEAR [33] with outdoor and indoor occupancy factors of 0.2 and 0.8 respectively. The annual effective dose due to natural activity concentration in the mining environment was determined using the following equations:

$$\text{AEDE (Outdoor) (mSv/y)} = \text{(D) nGy/h} \times 8760\text{h} \times 0.7\ \text{Sv/Gy} \times 0.2\times10^{-6} \quad (4)$$

where D = Dose rate (nGy h^{-1}), 8760h = period of exposure in hours per year and 10^{-6} = Factor used to convert nanogray to milligray, 0.2 = Outdoor occupancy factor, 0.7 (Sv/Gy) = The Factor used to convert the absorbed dose in air to the effective dose received by an adult person. 0.07mSv was used as the average background outdoor effective dose rate in the mining site for an adult person [6].

External Hazard index (H_{ex})

When the decay of naturally occurring radioactive material (NORMs) takes place, they give rise to external radiation field which has the potential to expose humans [34]. The decay of radioactive materials occurs naturally and when these happen they produce external radiation field which expose humans. The external health hazard index as a result of this exposure represents to these three major radionuclides External hazard index represents the hazard incurred due to exposure radiation from ^{226}Ra, ^{232}Th and ^{40}K in the coal mining environment. It was derived from the same expression of Ra_{eq} with the supposition that its maximum value corresponds to the upper limit of 370 Bq/kg. It was calculated from the equation bellow [27, 6];

$$H_{ex} = \frac{ARa}{370} + \frac{ATh}{259} + \frac{AK}{4810} \leq 1 \quad (5)$$

where H_{ex} = the external hazard index and ARa, ATh and AK are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The value of external hazard index must be less than unity in order to keep the radiation hazard insignificant. The prime objective of this index is to limit the radiation dose to the accepted dose limit of 1mSv/yr.

Internal Hazard Index (H_{in})

Radon and its short-lived daughter nuclei endanger the respiratory organs such as the lungs when inhaled. So the internal exposure to radon and subsequent production of its short lived daughters is quantified by an internal hazard index and expressed mathematically by UNSCEAR [6] and Beretka and Mathew [27] as:

$$H_{in} = \frac{ARa}{185} + \frac{ATh}{259} + \frac{AK}{4810} \leq 1 \quad (6)$$

where H_{in} = internal hazard index and A_{Ra}, A_{Th} and A_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The internal hazard index must be less than unity for radiation hazard incurred due to inhalation of radon contaminated air to be negligible [35].

Gamma index

The radiation hazard associated with natural radionuclides contained in the investigated coal samples were estimated using representative gamma index (Iγr). Gamma index was also used as a screening index that confirms the conformity of environmental samples to dose standards set for building construction materials [36]. According to the European Commission (EC), gamma activity concentration index (Iγr) helps to identify whether dose standard is met [37] or not and this was estimated using the formula:

$$I\gamma r = \frac{ARa}{150} + \frac{ATh}{100} + \frac{AK}{1500} \leq 1 \quad (7)$$

Where ARa, ATh and AK are the specific activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K respectively in Bq/kg. In order to satisfy the given dose criteria representative gamma index (Iγr) must be less than or equal to unity which corresponds to an annual effective dose of ≤ 1mSv [38, 36].

Annual Gonadal Dose Equivalent

According to UNSCEAR [39], the reproductive organs and bone marrows are considered as organs of interest. Therefore the Annual Gonadal Equivalent Dose (AGDE) for the workers/residents of the study area due to exposure to ^{226}Ra, ^{232}Th and ^{40}K was estimated using the equation given by Arafa [40] as:

$$AGDE = 3.09\text{ARa} + 4.18\text{ATh} + 0.314\text{AK} \quad (8)$$

Where ARa, = Activity concentration of ^{226}Ra, ATh = Activity concentration of ^{232}Th and
AK = Activity concentration of ^{40}K.

Excess life time cancer risk (ELCR)

The possibility of developing cancer by any of the mine workers or residents of the study area who will probably spend majority or all their life time in the study environment can be evaluated using the excess life time cancer risk (ELCR). The calculated annual effective dose (AEDE) is used to estimate the excess lifetime cancer risk (ELCR) using 70 years as the average duration of life for human beings according to the equation [36, 41]:

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

where AEDE = The annual effective dose equivalent in mSv/year , DL = Average duration of life (Estimated to be 70 years average), and RF = Risk factor (Sv^{-1}), i.e fatal cancer risk per Sievert. For low dose background radiations which are considered to produce stochastic effects, ICRP uses 0.05 threshold value for the public [41].

Results

Radioactivity concentration in coal

The measured activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K from the sampled coal was displayed on Table 1 below. The activity concentration of ^{226} Ra, ^{232}Th and ^{40}K range from 13.75±0.75 to 36.88±1.90 Bq/kg, 2.85±0.35 to 18.59±1.13 Bq/kg, and 0.41±0.05 Bq/kg to 50.07±2.70 Bq/kg with average values of 21.34±7.94 Bq/kg, 13.61±5.80 Bq/kg and 20.91±15.77 Bq/kg respectively.

Table 1: Show Radionuclide concentration in different coal samples in Bq/Kg

Sampling code	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K Bq/kg)
CS1	36.88±1.90	11.01±0.63	15.50±1.00
CS2	13.75±0.78	2.85±0.35	6.86±0.51
CS3	14.50±0.82	11.49±0.62	13.72±0.90
CS4	32.11±1.66	12.34±1.21	19.34±1.21
CS5	27.34±1.41	9.61±0.55	9.05±0.69
CS6	16.64±0.89	17.47±0.95	42.94±2.32
CS7	16.61±0.89	17.19±1.03	0.41±0.05
CS8	19.11±0.99	17.84±1.03	50.07±2.70
CS9	17.61±0.94	18.59±1.13	28.53±1.62
CS10	18.82±0.99	17.72±0.98	22.64±1.34
Min.	13.75±0.78	2.85±0.35	0.41±0.05
Max.	36.88±1.90	18.59±1.13	50.07±2.70
Mean	21.34±7.94	13.61±5.08	20.91±15.77

Radiological hazard indices

Table 2 below, shows the calculated values of radiological hazard parameters which include radium equivalent activity (Raeq), absorbed dose rate in air (D), annual effective dose equivalent (AEDE) and annual gonadal dose equivalent (AGDE). They recorded minimum and maximum values of 18.36 to 53.81 Bq/Kg, 8.39 to 24.32 nGy/h, 0.01 to 0.03mGy/yr, and 129.97 to 826.69 mSv/yr respectively, with average values of 42.01 Bq/Kg, 18.76 nGy/h, 0.03 mGy/yr and 407.74mSv/yr respectively.

Table 2: Various Radiological Hazard Indices Recorded in this Study

Sampling Code	(Ra_{eq}) Bq/kg	D (nGy/h)	AEDE (mGy/yr) (Outdoor)	AGDE (mSv/yr)	$I_{\gamma r}$ (mSv/yr)	(H_{ex})	(H_{in})	ECLR ($\times 10^{-3}$)
CS1	53.81	24.32	0.03	808.07	1.86	0.73	0.99	0.51
CS2	18.36	8.39	0.01	506.99	1.17	0.46	0.58	0.32
CS3	31.99	14.24	0.02	826.69	1.89	0.75	0.99	0.50
CS4	51.25	23.09	0.03	475.86	1.27	0.39	0.57	0.30
CS5	41.78	18.81	0.02	211.56	0.49	0.19	0.25	0.13
CS6	44.93	20.03	0.02	216.24	0.50	0.14	0.19	0.14
CS7	41.22	18.07	0.02	153.33	0.35	0.12	0.15	0.10
CS8	44.48	21.70	0.03	129.97	0.30	0.54	0.70	0.10
CS9	46.39	20.56	0.03	583.88	1.34	0.15	0.25	0.38
CSI0	45.90	18.33	0.02	164.85	0.37	0.05	0.09	0.10
Min.	18.36	8.39	0.01	129.97	0.30	0.05	0.09	0.10
Max.	53.81	24.32	0.03	826.69	1.89	0.75	0.99	0.51
Mean.	42.01	18.76	0.03	407.74	1.06	0.39	0.53	0.29

Key: Raeq: Radium Equivalent Activity, D: Absorbed Dose rate, AEDE: Annual Effective Dose Equivalents AGDE: Annual Gonadal Dose Equivalent, $I_{\gamma r}$: Representative Gamma Index, H_{ex}:: External Hazard Indices, and H_{in}: Internal Hazard Indices, ECLR: Excess Life Time Cancer Risk for Coal Sample

Table 3: Comparison of activity concentration of ^{226}Ra and ^{232}Th and ^{40}K in coal from the study area with data from elsewhere

Country	^{226}Ra	^{232}Th	^{40}K
Okobo Kogi State [present study]	21.34±7.94	13.61±5.08	20.91±15.77
Northwest Nigeria [9]	8.18	6.97	2.38
Kikiwira Tanzania [35]	41	37	293
Spain [34]	64	18	104
Greece [4]	133	18	108
World average [30]	20	20	50

Discussion

The result reveals that there were variations in the radionuclide concentration of ^{226}Ra, ^{232}Th, and ^{40}K in different coal sample. But the standard deviations of all three radionuclides appeared to be rather low showing that their concentration level in all the ten samples were close to a moderate extent. The mean value of ^{226}Ra (21.34 Bq/kg) concentration was found to be slightly higher than the world average value of 20 Bq/kg [39, 40, 41]. The slight increase may be due to leaching from upper layers and subsequent infiltration into coal beds. However the values of ^{232}Th (13.61 Bq/kg) and ^{40}K (20.91 Bk/kg) were found to be lower than the worldwide average value of 20 and 50 Bq/kg [39] respectively. When compared with other published studies as shown in table 3 above, it was discovered that the average activity concentration of ^{226}Ra was greater than the values reported by Kolo *et al.* [9] and Mehade Hasan *et al.* [42]. However it was lower than those reported from Spain and Greece [43, 4]. The activity concentration of ^{232}Th on the other hand was found to be greater than the values reported in Nigeria [9] and Bangladesh [42], but lower than the average values reported in Tanzania [44] and Spain [43]. The radionuclide concentration value recorded for ^{40}K in this study was greater than that reported by Kolo *et al.* [9] in Northeast Nigeria, but greater than the rest of the values reported in Tanzania, Spain and Greece as shown in table 3 above. This variation in radionuclide concentration may due to mineralization and geological formation of the area

The radiological hazard indices as shown above recorded values which were lower than the world average value of 370 Bq/kg, 59nGy/h, and 0.07mGy/yr [6] for Raeq, D, and AEDE. However, the AGDE recorded a value of 407.74mSv/yr which is significantly higher than the world permissible limits of 300mSv/yr [6]. This implies that both staff and residents of the community may be exposed to radiological hazard, which might affect the functionality of their sensitive organs most especially the reproductive organs, and the bone marrows [33, 36]. Representative gamma index ($I_{\gamma r}$), external hazard index (Hex), internal hazard index (Hin), and excess lifetime cancer risk (ELCR) all recorded values between 0.30 to 1.89 mSv/yr, 0.05 to 0.75, 0.09 to 0.99 and 0.10 to 0.51×10^{-3}, with average value of 1.06 mSv/yr, 0.39, 0.53 and 0.29×10^{-3}respectively, with average value of 1.06 mSv/yr, 0.39, 0.53 and 0.29×10^{-3}respectively, which are comparable to the minimum acceptable limit of 1.0 mSv/yr, 1, 1, and 0.29×10^{-3} [6] respectively. These values are within global acceptable limit but adequate precautionary measures should be put in place to avoid continued and long- time cumulative effects

Conclusion

The natural radionuclide concentration of ^{226}Ra, ^{232}Th and ^{40}K in the sampled coal from Okobo coal mine has been estimated using high resolution spectrometry technique. The radionuclide concentration of ^{226}Ra recorded a unit mean value higher than the global average, however the mean values recorded by ^{232}Th and ^{40}K fall below global permissible values. All calculated radiological health hazard indices recorded lower values than the precautionary safe limit but for annual gonadal dose equivalent which recorded significantly higher value than the global permissible limit. This implies that both staff and residents of the community could be exposed to radiologiscal threat, which might affect the functionality of their sensitive organs if continued and cumulative exposure is not avoided.

Limitation

Mechanical sampling from moving streams of coal is the preferred method of coal sampling. This helps to obtain more accurate and reliable results. Nevertheless manual sampling technique was used in this study because mechanical sampling facilities were not available in the mining site.

Recommendation

There should be regular environmental radiation monitoring program (area monitoring) using survey meters, which should cover the entire Okobo community to ascertain the extent of redistribution and enhancement of natural

radionuclide in the host community. Individual monitoring of mine workers to ensure that dose received are kept below specified annual limits or does not exceed global acceptable level. The use of personal protective equipment (PPE) such as overalls, safety glasses, dust mask, safety boots and gloves by mine workers is also advised.

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Conflict of interest
The authors declared no conflict of interest

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