ASSESSMENT OF RADIOACTIVE AND TRACE ELEMENTS IN MAIGANGA COAL OF UPPER BENUE TROUGH, NORTHEASTERN NIGERIA

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ABSTRACT

Maiganga coal is one of the recently discovered coal deposits in Gombe state, north-eastern Nigeria of which no published information is known about the concentrations of its naturally occurring radioactive materials and trace elements. Though it is currently being mined and used for firing the kilns of a leading cement factory in Nigeria, a comprehensive radiological and trace elements characterization of this deposit is needed for the protection of both the coal workers and the community from the effects of ionizing radiation and metal pollution associated with coal use. In this research, gamma spectrometric technique which employs the HPGe detector was employed to qualitatively and quantitatively assess the natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) present in Maiganga coal samples. Radiological hazard indices due to these primordial radionuclides were computed with a view to establishing the radiological health implications on both the miners, the immediate community of the mine and the coal users. The trace elements concentrations were determined using inductively coupled plasma-mass spectrometry (ICP-MS). The levels of lead (Pb), chromium (Cr), cobalt (Co), copper (Cu), zinc (Zn), barium (Ba), beryllium (Be), nickel (Ni) and vanadium (V) were analysed, and correlated with their respective Clarke values for world low-rank coals. The overall results were compared with literatures from other parts of the world.

Keywords: Maiganga coal; HPGe detector; Natural radioactivity; Absorbed dose rates; ICP-MS; Trace elements.

INTRODUCTION

The recent population growth and simultaneous industrialization in Nigeria has resulted in high demand for power generation and energy consumption. It is becoming difficult for the nation's sizable oil and gas reserves to support the growing national economy in a sustainable pace. Alternative sources such as coal are thus attracting increasing attention as supplementary energy source. Coal which remains the most abundant and most accessible natural source of fossil energy [1, 2], contain varying amounts of naturally occurring radioactive materials particularly ^{238}U, ^{232}Th and their radioactive progenies, and ^{40}K [3]. Coal is also contains many non-radioactive but toxic elements in trace amounts [4]. Coal exploitation and utilization constantly redistributes these radioactive and trace elements thereby enhancing their concentrations above background levels in the environment [5, 6].

According to Lu et al. [2], this can result in serious health consequences for the coal workers, the environment and the end users. Detailed information on the radiological characteristics of coal is thus an indispensable tool for determining the exposure levels of coal workers and the environment. Data on the elemental constituents of coal are also needed for comprehensive assessment of human health and environmental impacts of coal exploitation and utilization.

In this study, activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Maiganga coal has been evaluated using HPGe γ-ray spectrometer. Elemental concentrations were also assessed using the ICP-MS technique. The results of this study are of paramount importance, not just for the health of the workers and the environment but also to help the Nigerian government in developing a safer and environmentally friendly technology for a coal energy-driven economy.

MATERIALS AND METHODS

Study Area. Maiganga is a local community located between latitude 10^0 02` to $10^0$ 05` and longitude 11^0 06` 50`` to 11^0 08` in Akko local government area of Gombe state, northeast Nigeria (Fig. 1). Maiganga coal is one of the recently discovered coal deposits whose seam thickness measure about 3 m. It is among the coal deposits targeted for power generation by Nigerian government. The coal extracted from this mine currently serve as the primary energy source for firing the cement kilns of one of Nigeria's major cement factory in north-eastern Nigeria.

Sample Collection. A total of 7 coal samples, namely MCS1 to MCS7 were collected at random from Maiganga coal mine. After collection, the samples were thoroughly cleaned to rid them of all possible contaminants, then sun-dried for 72 h. The dried samples were neatly packed in well labelled polythene bags and transported to the radiation laboratory, department of physics, University of Malaya for analysis.

Sample Preparation. In the laboratory, the samples were further air-dried for 24 h to attain constant weight. The dried samples were crushed into fine powder using the agate mortar and homogenized via sieving process. An amount of 370 ± 1 g of the homogenized samples were packed into sealable, well labelled marinelli beakers. All the samples were finally sealed tightly and stored for about 5 – 6 weeks in preparation for radiological analysis. This was necessary to maintain radioactive secular equilibrium between the daughter nuclides and their respective long-lived parents [7-11].

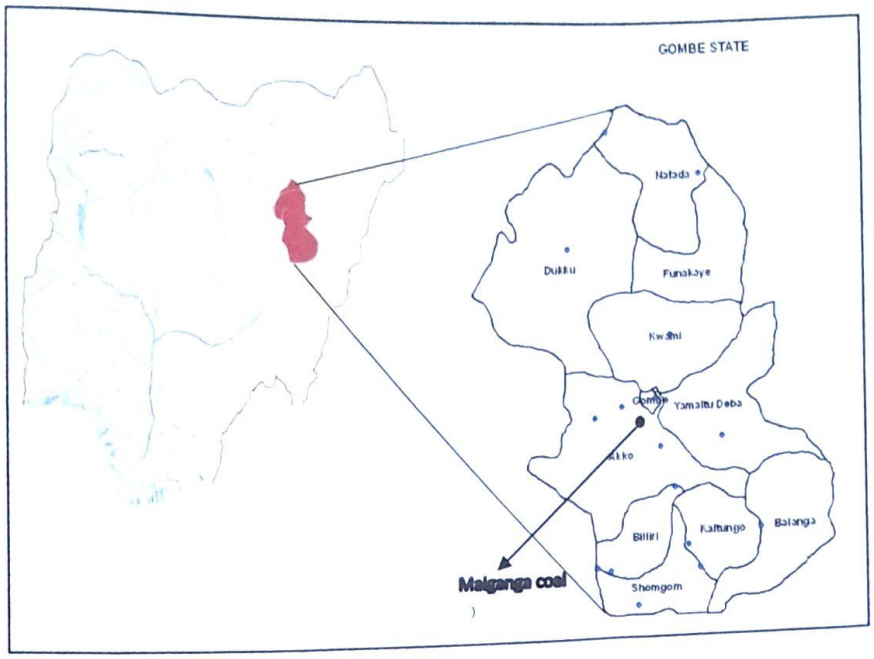

Figure 1: Map of Gombe state, Nigeria, showing the project site

Sample preparation for elemental analysis involved acid digestion procedure whereby 0.50 g of fine, homogenized coal samples was treated with a mixture of concentrated HNO_3, HF and $HClO_3$. The solution was evaporated to near dryness on a hot plate and further treated with 10 mL of 5 M HNO_3. The final digestate was diluted to 50 mL with deionized water and stored in refrigerator for analysis.

Gamma Spectrometric Measurements. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in all the samples was assessed using a P-type Coaxial ORTEC, GEM-25 HPGe gamma ray detector with 57.5 mm crystal diameter, 51.5 mm thickness and 28.2% relative efficiency [12]. The detector which has a FWHM energy resolution of 1.67 keV at 1.33 MeV peak of ^{60}Co gamma ray line [13, 14], was coupled to ADCM data acquisition system with PCAII multi-channel analyser. The detector was enclosed in a good cylindrical lead shield with a fixed bottom in order to reduce the interference of background radiation from terrestrial and extra-terrestrial sources with the measured spectrum [15-17]. Before the measurement, the detector was calibrated for energy and efficiency using a cylindrical multi-nuclide gamma ray source with homogenously distributed activity in the same container geometry as the samples. The calibration source which contained the nuclides: ^{241}Am (59.541 keV), ^{109}Cd (88.040 keV), ^{57}Co (122.061 keV, 136.474 keV), ^{203}Hg (279.195 keV), ^{113}Sn (391.698 keV), ^{85}Sr (514.007 keV), ^{137}Cs (661.657 keV), ^{88}Y (898.042 keV, 1836.063 keV), and ^{60}Co (1173.22 keV, 1332.492 keV), was supplied by Isotopes products laboratories, Valencia, CA 91355.

Each sample was counted for 86,400 s. The net count rate of the primordial radionuclides was obtained by subtracting the respective count rate from the background spectrum acquired for the same counting time. The activity concentration of ^{226}Ra was estimated from the weighted average gamma peaks of ^{214}Pb (351.93 keV, 35.6%) and ^{214}Bi (609.32 keV, 45.49%); while that of ^{232}Th was estimated from the weighted average gamma peaks of ^{212}Pb (238.63 keV, 46.6%) and ^{208}Tl (583.19 keV, 99.2%). The ^{40}K activity concentration was determined from its 1460.822 keV, (10.66%) single characteristic gamma

line. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in all the samples investigated were calculated using the expression [14, 17-22];

$$A(Bqkg^{-1}) = \frac{CPS \times 1000}{\varepsilon_\gamma \times I_\gamma \times W} \quad (1)$$

where A (Bq kg^{-1}) is the specific activity, CPS is the net counts per second for each sample investigated, ε_γ (E) is the detector photo-peak efficiency at respective gamma-ray peak, I_γ is the corresponding gamma ray intensity and W the mass of sample in gm.

Trace Elements Analysis. The trace elements analysis was done at the ICP-MS laboratory, Chemistry department, University of Malaya using the Agilent Technologies 7500 Series inductively-coupled plasma mass spectrometry (ICP-MS). Before analysis, the instrument was externally calibrated with a commercially available multi-element standard having analyte concentration of 10 ppm and the results corrected to weight of bulk rock [23].

RADIATION INDICES

Radium Equivalent Activity (Ra_{eq}). ^{226}Ra, ^{232}Th and ^{40}K are known to be non-uniformly distributed in virtually all materials on the earth surface. The overall activity of any material containing different elements of these primordial radionuclides is expressed as a weighted sum of their individual activities. This weighted sum, called the Radium equivalent activity (Ra_{eq}) is defined based on the assumption that 370 Bq kg^{-1} of ^{226}Ra or 259 Bq kg^{-1} of ^{232}Th or 4810 Bq kg^{-1} of ^{40}K produce the same gamma ray dose [8, 18]. It is calculated from the equation [24, 25]:

$$Ra_{eq}(Bqkg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_K are the respective specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}.

Absorbed Dose Rate (D_R). The absorbed dose rates (D_R) due to gamma radiations in air, 1m above the ground were estimated using the conversion factors published in [25] as

$$D_R(nGyh^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \quad (3)$$

where A_{Ra}, A_{Th} and A_K are the specific activities measured in Bqkg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively.

Annual Effective Dose Equivalent (AEDE). Two radiation parameters have been provided by UNSCEAR [25] which are critical in the estimation of annual effective dose in air. These are the conversion coefficient from absorbed dose in air to effective dose, given to be 0.7 Sv Gy^{-1}; and the outdoor occupancy factor, given to be 0.2, with the view that an individual spends an average of 80% of his time indoors. The annual effective dose equivalent (AEDE) in outdoor air, measured in mSv yr^{-1} is thus calculated as [25]:

$$AEDE(mSvy^{-1}) = D_R(nGyh^{-1}) \times 24 \times 365.25 \times 0.2 \times 0.7 \times 10^{-6} \quad (4)$$

External Hazard Index (H_{ex}). Radiation hazard incurred due to external exposure to gamma rays is quantified in terms of external hazard index (H_{ex}) given by UNSCEAR [25]:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{48\ 10} \quad (5)$$

where A_{Ra}, A_{Th}, and A_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K respectively.

UNSCEAR [25] provided that the value of the above index must be less than unity for the radiation hazard to be regard as insignificant.

RESULTS AND DISCUSSION

Activity concentrations in Bq kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K in Maiganga coal were assessed using Eq. (1) and the results presented in Table 1 with their respective uncertainty levels of $\pm\sigma$. Fig. 2 shows an independent pictorial representation of the activities of the radionuclides. Table 2 shows the comparison of the results with similar studies around the world and with the world average values documented by [25].

Table 1: Activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th, ^{40}K, and the radiological hazard indices of Maiganga coal

Sample ID	Activity concentrations (Bq kg^{-1})			Radiation hazard indices			
	^{226}Ra	^{232}Th	^{40}K	Ra_{eq} (Bq kg^{-1})	D_R (nGy h^{-1})	AEDE (mSv y^{-1})	H_{ex}
A12	8.0 ± 0.5	7.1 ± 0.4	13.3 ± 0.7	19.2	8.5	0.01	0.05
A14	6.5 ± 0.3	6.2 ± 0.3	9.3 ± 0.5	16.1	7.1	0.01	0.04
A16	6.7 ± 0.3	5.7 ± 0.3	9.0 ± 0.5	15.5	6.9	0.01	0.04
A21	2.3 ± 0.2	1.1 ± 0.1	6.1 ± 0.4	4.4	2.0	0.00	0.01
A23	1.5 ± 0.1	1.4 ± 0.1	4.8 ± 0.4	12.7	5.7	0.01	0.03
A24	5.0 ± 0.2	4.8 ± 0.3	11.1 ± 0.6	12.7	5.7	0.01	0.03
A26	3.0 ± 0.2	1.5 ± 0.1	6.5 ± 0.4	5.7	2.6	0.00	0.02
Min	1.5 ± 0.1	1.1 ± 0.1	4.8 ± 0.4	3.8	1.7	0.00	0.01
Max	8.0 ± 0.5	7.1 ± 0.4	13.3 ± 0.7	19.20	8.55	0.01	0.05
Mean	4.7 ± 0.3	4.0 ± 0.2	8.6 ± 0.5	11.1	4.9	0.01	0.03

Table 2: Comparison of activity concentrations (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K of Maiganga coal with world average values and those of similar studies

Country	^{226}Ra	^{232}Th	^{40}K	Reference
Hong Kong, China	17	20	24	Tso and Leung [26]
Lodz, Poland	10.4 – 28.4	8.5 – 20.1	43.9 – 180.3	Bem, Wieczorkowski [27]
Kolaghat, India	25.0 – 49.9	39.3 – 55.2	120.8 – 151	Mandal and Sengupta [28]
Greece	309 – 395	19 – 24	148 – 207	Karangelos, Petropoulos [29]
Baoji, China	26.3	36.6	99.8	Lu, Jia and Wang [30]
Cayrrhan, Turkey	14.55	11.12	123.01	Cevik, Damla [1]
World average	35	30	400	UNSCEAR [25]
Maiganga, Nigeria	1.5 – 8.0	1.1 – 7.1	4.8 – 13.3	Present study

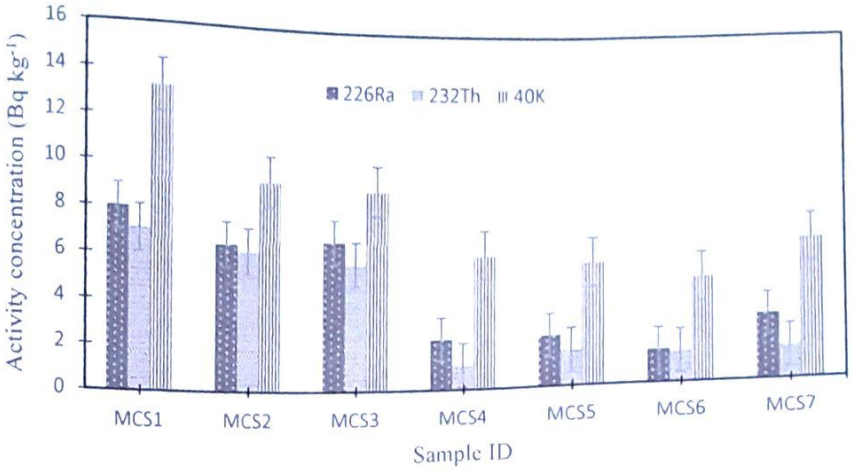

Figure 2: Distribution of ^{226}Ra, ^{232}Th and ^{40}K in the coal samples

As seen in Table 1, the range of activity concentration of ^{226}Ra is between 1.5 ± 0.1 Bq kg^{-1} and 8.0 ± 0.5 Bq kg^{-1} with an average value of 4.7 ± 0.3 Bq kg^{-1}. ^{232}Th varies in activity values from 1.1 ± 0.1 Bq kg^{-1} to 7.1 ± 0.4 Bq kg^{-1} with a mean activity of 4.0 ± 0.2 Bq kg^{-1}, while ^{40}K recorded a mean activity value of 8.6 ± 0.5 Bq kg^{-1} for all the studied coal samples. These values are lower than those reported for similar studies from several parts of the world. They are also below the world mean values of 35, 30 and 400 Bq kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K respectively documented by UNSCEAR [25] for low rank coals (Table 2).

To effectively assess the health and environmental effects due to Maiganga coal exploitation, radiation hazard parameters were calculated using Eqs. (2) – (5) and the results are presented in Table 1. The calculated parameters include Ra_{eq}, D_R, AEDE and H_{ex}, respectively.

The mean value of Ra_{eq} as seen in Table 1 was 11.1 Bq kg^{-1}, while the average value of 0.03 was recorded for the external radiation hazard, H_{ex}. These mean values for Ra_{eq} and H_{ex} fall far below the world maximum limit of 370 Bq kg^{-1} and 1 respectively provided by [25]. The mean relative contributions of ^{226}Ra, ^{232}Th and ^{40}K to Ra_{eq} are 43.3%, 51.5% and 5.9% respectively as seen in Fig. 3a.

The absorbed dose rate, D_R in air due to terrestrial gamma rays at 1 m above the ground ranged from 1.7 to 8.5 nGy h^{-1} with a mean value of 4.9 nGy h^{-1}. The corresponding mean outdoor annual effective dose rate, AEDE for Maiganga coal samples was 0.01 mSv y^{-1}. These values were lower than the UNSCEAR [25] world average provisions of 55 nGy h^{-1} and 0.460 mSv y^{-1} respectively. Major contributions of 44.4% and 48.4% to the absorbed dose rate were from ^{226}Ra and ^{232}Th respectively, while ^{40}K contributed about 7.3% as seen in Fig. 3b.

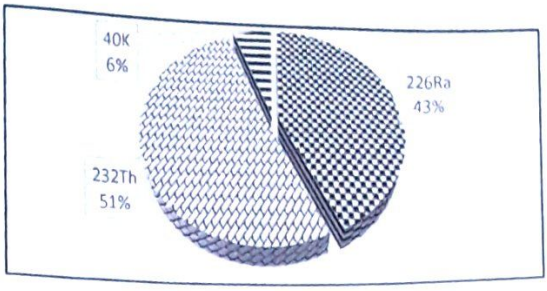

Figure 3a: Relative contribution to Ra_{eq} from ^{226}Ra, ^{232}Th and ^{40}K

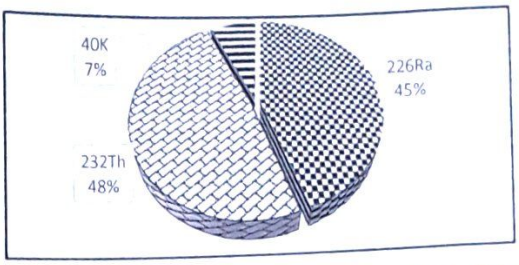

Figure 3b: Relative contribution to D_R from ^{226}Ra, ^{232}Th and ^{40}K

The overall results of the hazard indices affirmed that the exploitation of Maiganga coal pose no significant radiological impact for both the mine workers, the immediate community and the environment in general.

Table 3 give the trace elements composition of Maiganga coal along with their respective Clarke values adopted from [31] and the estimated enrichment/depleting factors (EDF). The concentration of Cr in Maiganga coal varies between 0.9 mg kg^{-1} and 13.5 mg kg^{-1} with an average of 5.9 mg kg^{-1}. This value is lower than the Clarke value for low- rank coals. Pb varies in concentration from 0.7 mg kg^{-1} to 6.1 mg kg^{-1} with a mean concentration value of 3.2 mg kg^{-1}. Although the values are found to be lower than the world average concentration of Pb in coals, its accumulation over a long period, coupled with the subsequent combustion of coal could become a threat to man and the environment. The mean concentration for Ni is 4.5 mg kg^{-1} which is low compared with the average value for Ni in world low-rank coals. The estimated EDF of 0.3 shows the level of depletion of Ni in Maiganga coal. Furthermore, Cu, Zn and Co have mean concentration values of 5.2, 1.3 and 2.0 mg kg^{-1} respectively.

Table 3: Trace Elements Concentrations (mg kg^{-1}) of Maiganga Coal

Sample	Cr	Co	Ni	Pb	Cu	V	Zn	Ba	Be
A12	13.5	3.7	8.5	6.1	10.2	28.5	3.9	59.4	1.1
A14	7.3	2.6	4.2	4.7	7.2	15.6	1.1	50.4	1.5
A16	9.8	2.7	5.1	5.2	8.3	20.4	3.5	52.8	1.5
A21	1.5	0.4	3.2	0.7	1.2	1.8	0.0	30.2	0.3
A23	0.9	0.8	3.2	0.7	1.5	0.8	-0.3	39.8	0.3
A24	6.4	3.5	5.5	3.8	5.8	5.6	1.3	57.8	1.0
A26	1.8	0.1	2.1	0.9	2.0	2.2	-0.1	43.5	0.3

Min	0.9	0.1	2.1	0.7	1.2	0.8	-0.3	30.2	0.3
Max	13.5	3.7	8.5	6.1	10.2	28.5	3.9	59.4	1.5
Mean	5.9	2.0	4.5	3.2	5.2	10.7	1.3	47.7	0.9
Clarke	15 ± 1	4.2 ± 0.3	9 ± 0.9	6.6 ± 0.4	15 ± 1	22 ± 2	18 ± 1	150 ± 20	1.2 ± 0.1
EDF	0.4	0.5	0.5	0.5	0.3	0.5	0.1	0.3	0.7

A general assessment of the elemental concentrations of Maiganga coal shows that the mean concentration values for all the studied trace elements are below the world average values for low rank coals. Furthermore, the calculated EDF for the trace elements varies between 0.1 and 0.7 (Table 3). These values are below unity which suggests that Maiganga coal is generally depleted of trace elements including those elements considered to be environmentally harmful. Thus, Maiganga coal pose no environmental or health challenge either to the coal workers or the immediate community around the mine. It is therefore safe for exploitation and utilization

CONCLUSION

The natural radioactivity and trace elements concentrations of Maiganga coal was determined to assess the human health and environmental implications of its exploitation. The mean activities of ^{226}Ra, ^{232}Th and ^{40}K are 4.7 ± 0.3, 4.0 ± 0.2 and 8.6 ± 0.5 Bq kg^{-1} respectively with a mean external absorbed dose rate of 4.9 nGy h^{-1}. These values are below the values obtained from similar studies around the world. They were also found to be lower than the world average values for coal documented by UNSCEAR. The results showed that radiation resulting from the mining and use of Maiganga coal is of no significant consequence to coal workers or the environment. Furthermore, the trace elements concentrations of Maiganga coal were far lower than their respective Clarke values for low rank coals, indicating high depletion level of the elements. The results of this study thus confirm that Maiganga coal is safe for exploitation and utilization from the human health and environmental protection perspective.

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