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Assessment of Heavy Metal Contamination of Coal Deposits in Kogi State, Nigeria using Instrumental Neutron Activation Analysis

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Abstract

In this study, the concentration of heavy metal contaminants in the coal samples from selected coal mining sites in Okobo, Odagbo, Ofugo and Opoko-Obido, within Kogi State, Nigeria were identified and determined using the Instrumental Neutron Activation Analysis Technique. Four samples were collected from the mining sites at 0 – 5 cm, and 5 – 10 cm below the soil surface. The range of the concentrations of heavy metal in the samples are V (6.03 ± 0.66 to 18.40 ± 1.10), Mn (6.06 ± 1.17 to 273 ± 2), As (0.18 ± 0.02 to 0.33 ± 0.04), U (0.57 ± 0.05 to 0.74 ± 0.05), Cr (7.14 ± 1.62 to 16.60 ± 1.80), Co (1.49 ± 0.16 to 9.10 ± 0.33), Sr (312 ± 71 to 383 ± 107), Sb (0 to 0.17 ± 0.05), Ba (0 to 174 ± 19) and Th (1.30± 0.09 to 2.39 ± 0.11). These values of Mn, Sr and Sb are slightly higher than the National Environmental Standards and Regulations Enforcement Agency and the United States Environmental Protection Agency permissible limits of 150 mg/kg, 300 mg/kg and 3.0 mg/kg respectively. The study revealed that the coal from the mines was contaminated with heavy metals especially Sr, Ba, and Ti thereby posing stern environmental and health concerns.

Keywords: Coal; heavy metals; Neutron Activation Analysis.

I. INTRODUCTION

Coal is one of the natural resources found in abundance across the globe and is derived from ancient plants that have been buried for millions of years under layers of rock and soil. It is characterized as a carbon-rich sedimentary rock, with a dark hue that varies from black to brownish-black and exhibits high combustibility [1]. Its versatility makes it a widely used resource for various applications, such as in power generation, steel production, and heat purposes. However, coal also produces effluence that is hazardous to humankind.

The burning process of coal produces sulfur and nitrogen that reacts with the atmospheric moisture to produce sulfuric and nitric acids, which constitute pollution [2]. Exposure to these pollutants is associated with severe adverse impacts in critical human organs. The various stages of coal utilization including mining, transportation, combustion processes, and disposal of the coal wastes, put humans and the environment at greater health risk.

Heavy metals are elements that are dense and have a high atomic number. They are typically hard, shiny, and have a high melting point with exceptional ability to absorb and scatter X-rays and other forms of electromagnetic radiation. These heavy metals can have harmful effects on the environment and human health if not handled and disposed of properly. They can accumulate in the body and cause damage

to organs and tissues, leading to various health problems [3].

Heavy metals are categorised into toxic metals (Mercury, Lead, Arsenic, Cadmium, Chromium, and Thallium); essential metals which are necessary for human health in small amounts (Iron, Zinc, Copper, Manganese, Cobalt and Strontium) and radionuclides (Silver, Gold, Uranium, Thorium and Plutonium). Heavy metals in coal are of concern due to the potential environmental and health impacts when released during combustion and mining activities [3].

Coal mining and exploitation within and outside Nigeria have been on the increase in recent times. While there have been studies on coal and sediments from the study sites as well as several other coal mining sites within and outside Nigeria [4], [5], [6], [7], [8], [9], [10] and [11], the purpose of this study is to extensively investigate the toxic heavy metal pollution levels across the major coal sites in Okobo, Odagbo, Ofugo and Opoko-Obido, in Kogi State, Nigeria, with the view to ascertain the potential health effects of the metal's effluence caused by coal.

II. MATERIALS AND METHODS

A. Study Area

Okobo coal mining site with coal reserves of up to 380 million tonnes, located in Enjema district about 16 km east of Ankpa town, approximately 200 km North of Enugu lies between latitude 7˚22'14" N and longitude 7˚37'31"E [12], while Opoko Obido coal mine located in a small village in Egabada community of Igalamela-Odolu local government area (LGA), is bordered by River Niger in the west and Enugu State in the east. The mine has an area of 26.2 km^2 with abundant coal deposits and lies within latitude 7^0 2' 36" N and longitude 7^0 1' 15" E [13].

Odagbo coal mine located in the Okaba district, lies between latitudes 7^0 28' 30" N to 7^0 29' 00" N and longitudes 7^0 43' 30" E to 7^0 44' 00" E at an altitude of about 275 m above mean sea level and is positioned on the North-eastern part of the Anambra Basin [14], while Ofugo coalmine is a locality in the district of Enjema in Ankpa LGA and lies on latitude 7^0 33' 36.9" N (7.56024000) and longitude 7^0 37' 24.7" E (7.62354000) [13].

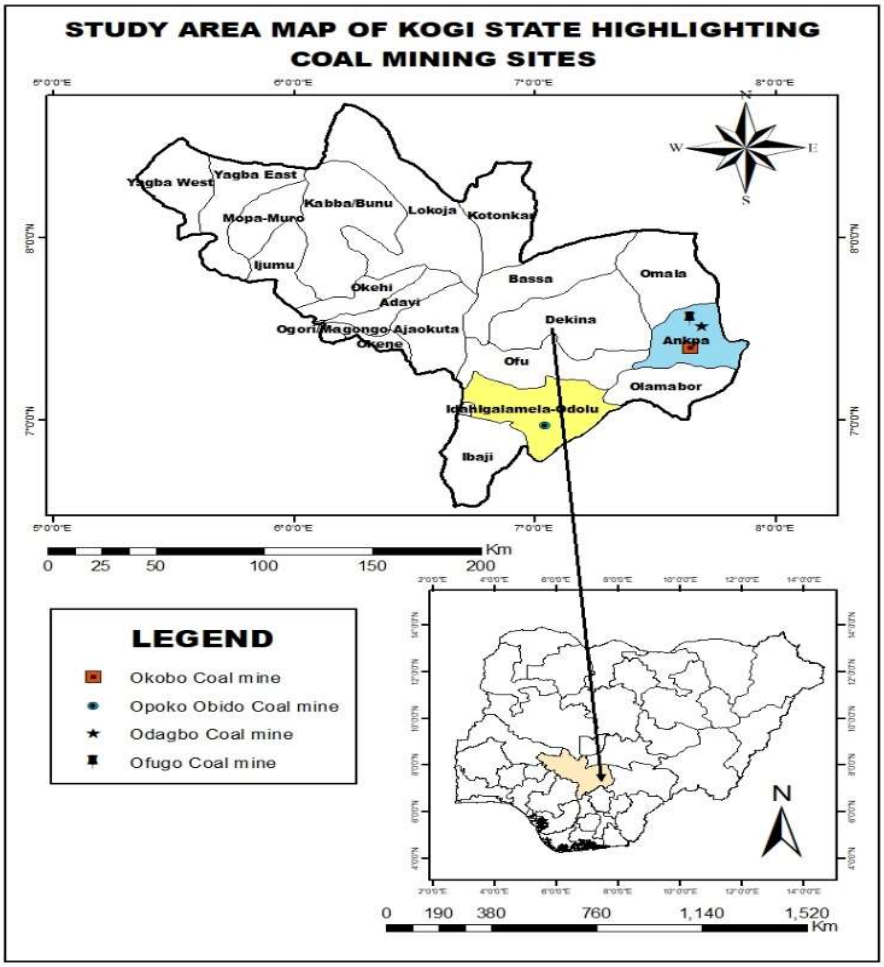

Fig. 1. Study Area Map of Kogi State, Nigeria Showing Coal Mining Sites

B. Materials

The materials used for this study are aluminum foil and polythene bag for sample collection, a grinder for sample pulverization, a mesh of 2 mm for sieving, tissue paper and acetone for cleaning to avoid cross-contamination of samples, plastic containers, masking tape, marker, polythene vial and a miniature neutron source reactor for irradiation of samples, and high purity germanium detector for detecting and measuring gamma radiation.

C. Sample Collection and Preparation

Four coal seams were collected from four active coal mines namely, Okobo, Opoko-Obido Odagbo, and Ofugo, all in Kogi State, Nigeria. These sites were selected due to the historical coal depositions and the reported cases of indiscriminate mining activities, poor water quality supply and incessant acid mine drainage pollution from underground coal mines which require further investigations that prompted this current study. Three samples taken at 0 – 5 cm, and 5 – 10 cm below the surface were collected from each site to form the composite sample. The samples were collected using aluminium foil, then wrapped and packed in polythene bags and transported safely into the laboratory for preparation.

The coal samples were pulverized into fine powder, sieved with a mesh of diameter 2 mm, and homogenized at the Nigerian Geological Survey Agency (NGSA) laboratory in Kaduna State, Nigeria. Each time a sample was pulverized; the plate inside the machine was washed and dry-cleaned with acetone and tissue paper to avoid cross-contamination of the samples. Then the powdered samples were packed in well-labelled plastic containers and properly sealed to avoid cross-contamination and for ease of identification. These samples were then taken to the Centre for Energy Research and Training (CERT), Ahmadu Bello University (ABU) Zaria for the instrumental neutron activation analysis (INAA).

The homogenized samples of coal were weighed (0.15 g), transferred to a 0.5 mL polythene vial (irradiation capsule) and sealed. The same process was done at 0.10 g for the standard reference material (SRM) NIST 1633c Coal Fly Ash. Subsequently, the coal samples and the primary standard (NIST 1633c) were put into cladding and irradiated accordingly through the instrumental neutron activation analysis method.

D. Methods

The Nigeria Research Reactor -1 (NIRR-1), a miniature neutron source with low enriched or natural uranium core as fuel, light water as moderator and beryllium as reflector [15] was used for the analysis. It is specifically used for elemental analysis and limited radioisotope production. Before the analysis, the spectrometer was calibrated using the standard reference material (coal fly ash 1633c), to generate the correction factor, which was used in calculating the concentration of the heavy metals in the coal.

The Coal and standard reference samples were encapsulated in heat-sealed polythene quartz vials and irradiated. Using the comparative method, the different elements were determined simultaneously.

Two irradiation schemes were used: one for short half-life elements and the other for long half-life elements. For the short half-life elements, irradiation was done for 1 minute and counted immediately for 10 minutes, with the activity of the sample kept at less than 40 Bq and the corresponding dead time of 10%. After counting, the sample was allowed to decay for two to three hours and then counted again for another 10 minutes. The short irradiation was done for 1 minute because geological samples have higher thermal neutron cross-section, so neutrons easily activate them. Elements such as magnesium (Mg), aluminium (Al), calcium (Ca), titanium (Ti), vanadium (V), sodium (Na), potassium (K), Manganese (Mn), europium (Eu) and dysprosium (Dy) were all detected after the short irradiation.

For elements with a long half-life, the samples were irradiated for 6 hours, and the first count was done after 3 days for 30 minutes. Then the second count was done after 10 days (i.e. 7 days after the first count) of irradiation for 1 hour. Elements such as arsenic (As), bromine (Br), lanthanum (La), samarium (Sm), ytterbium (Yb), uranium (U), scandium (Sc), chromium (Cr), iron (Fe), cobalt (Co), zinc (Zn), rubidium (Rb), antimony (Sb), barium (Ba), caesium (Cs), lutetium (Lu), hafnium (Hf), tantalum (Ta) and thorium (Th) were detected.

Table I presents a summary of the irradiation and counting procedure for the INAA technique used in this study.

E. Heavy Metals Pollution of the Coal Samples

We assess the quality of the coal samples and the level of heavy metal contamination using inferential statistical techniques such as enrichment factor, pollution index/contamination factor and pollution load index.

1) *Enrichment factor (EF)*

The degree of concentration of certain elements in coal, rocks, and soil samples relative to a reference or background value was calculated using (1).

$$EF = \frac{(C_x/C_f)sample}{(C_x/C_f)crust} \quad (1)$$

Where C_x and C_f represent concentrations of the element x and reference element f in the coal and reference sample [16].

2) *Pollution index (PI)*

A pollution index is a quantitative measure used to assess the level of pollution in a particular environment, such as air, water, or soil [17]. It provides a numerical representation of the concentration of harmful pollutants in the coal found in the environment. The heavy metals pollution index in coal serves as a valuable tool for assessing and monitoring the environmental impact of coal-related activities, as well as for safeguarding human health and the environment from the adverse effects of toxic metals contamination [17].

The pollution index (PI) of heavy metals is defined as the quotient of the concentration of the element of interest in the sample to the maximum permissible level of the element. This can be determined using (2).

Table I. Summary of INAA conditions used in this study.

Procedure	Element	Nuclide	Half-life	γ-ray energies (keV)
	Mg	^{27}Mg	9.4 min.	1014.4
Short irradiation - 1 minute	Al	^{28}Al	2.25 min.	1779.0
	Ca	^{49}Ca	2.5 hour	3084.5
Cooling: 2 - 3 hours	Ti	^{51}Ti	5.76 min.	320.1
	V	^{52}V	3.74 min.	1434.1
Counting - 600s	Na	^{24}Na	15 hour	1368.6
	K	^{42}K	12.4 hour	1524.6
	Mn	^{56}Mn	2.58 hour	846.8
	Eu	^{152}Eu	13.5 year	841.6
	Dy	^{165}Dy	2.334 hour	94.7
Long irradiation - 6 hours	As	^{76}As	26.24 hour	559
	Br	^{82}Br	33.9 hour	776.5
Cooling: 3 - 7 days	Fe	^{59}Fe	45.1 days	1099.3
	Cr	^{51}Cr	27.80 days	320.1
Counting :1800s - 3600s	Ba	^{131}Ba	12 days	496.3
	Sb	^{122}Sb	2.70 days	564.2
	Zn	^{65}Zn	244.00 days	1115.6
	U	^{239}Np	2.35 days	277.6
	Cs	^{134}Cs	2 years	795.8
	Sc	^{46}Sc	84 days	889.3
	Th	^{233}Pa (Th)	27.00 days	312
	Co	^{60}Co	5.25 year	1173.2
	Yb	^{169}Yb	32 days	198.0
	Sr	^{90}Sr		

$$PI = \frac{C_i}{S_i} \quad (2)$$

where PI is the pollution index corresponding to each sample, C_i is the concentration of each metal (mg/kg), and S_i is the maximum permissible level or background value (mg/kg) of the element.

Suppose the value of the pollution index of an element is greater than unity, it implies that the contamination of the sample by the element is high and may be toxic at the level it is in the sample [17].

The purpose of the pollution index evaluation is to provide policymakers, scientists, environmentalists, and the public with information about the environmental quality and health risks associated with coal pollution. By quantifying pollution levels, indices can help prioritize pollution control efforts, inform regulatory decisions, evaluate the effectiveness of pollution mitigation measures, and raise awareness about environmental issues.

3) *The Pollution load index (PLI)*

The Pollution load index (PLI) developed by [18] is used to quantify the amount of metal contamination in the examined coal samples. The PLI is represented as the nth root of the product of the n-contamination factor (CF). The Pollution load index (PLI) can be determined by (3).

$$PLI = (CF_1 * CF_2 * CF_3 * \dots\dots * CF_N) \quad (3)$$

where n is the number of heavy metals studied, and CF is the contamination factor [18].

4) *Contamination factor (CF)*

This is used to determine the degree of contamination status of the coal.

The contamination factor, CF was determined using (4).

$$CF = {}^{H_c}/_{H_b} \quad (4)$$

Where H_c = concentration of the metal at the contaminated site and H_b = background value of the metal.

PLI values >1, and < 1, mean deterioration of the site quality and no pollution respectively [17].

III. Results and Discussions

The results of the concentration of toxic heavy metals in the analyzed coal samples are presented in Table II.

The results of the heavy metals concentrations (mg/kg) obtained from the instrumental neutron activation analysis of the collected samples from Okobo coal, Opoko-obido coal, Odabgo coal, and Ofugo coal deposits showed the presence of ten (10) notable heavy metals (V, Mn, As, U, Cr, Co, Sr, Sb, Ba and Th). The concentrations and standard error (mg/kg) of

these heavy metals ranged from 6.03 ± 0.66 to 18.40 ± 1.10 for V, 6.06 ± 1.17 to 273 ± 2 for Mn, 0.18 ± 0.02 to 0.33 ± 0.04 for As, 0.57 ± 0.05 to 0.74 ± 0.05 for U, 7.14 ± 1.62 to 16.60 ± 1.80 for Cr, 1.49 ± 0.16 to 9.10 ± 0.33 for Co, 312 ± 71 to 383 ± 107 for Sr, 0 to 0.17 ± 0.05 for Sb, 0 to 174 ± 19 for Ba and 1.30 ± 0.09 to 2.39 ± 0.11 for Th.

Table 2. Heavy metals concentration in the coal samples.

Elements	S1	S2	S3	S4
V	13.80 ± 0.90	14.59 ± 1.24	6.03 ± 0.66	18.40 ± 1.10
Mn	237 ± 2	34.10 ± 0.70	6.06 ± 1.17	185 ± 2
As	0.18 ± 0.02	0.33 ± 0.04	0.28 ± 0.04	0.30 ± 0.03
U	0.57 ± 0.05	0.66 ± 0.07	0.61 ± 0.09	0.74 ± 0.05
Cr	16.20 ± 1.50	16.60 ± 1.80	10.50 ± 1.60	15.90 ± 1.60
Co	2.42 ± 0.19	9.10 ± 0.33	3.86 ± 0.28	1.49 ± 0.16
Sr	312 ± 71	BDL	383 ± 107	BDL
Sb	BDL	0.17 ± 0.05	BDL	BDL
Ba	BDL	BDL	131 ± 19	174 ± 19
Th	1.87 ± 0.09	2.16 ± 0.12	1.30 ± 0.09	2.39 ± 0.11

The results showed that the heavy metal concentrations varied within the coal samples and between locations. The concentration of manganese in samples *S1 (237 mg/kg)* and *S4 (185 mg/kg)* were higher than the National Environmental Standards and Regulations Enforcement Agency (NESREA) and the United States Environmental Protection (USEPA) permissible limits of 150 mg/kg. Similarly, strontium concentrations in samples *S1 (312 mg/kg)* and *S3 (383 mg/kg)* were higher than the NESREA and USEPA limits of 300 mg/kg.

Table III presents the enrichment factor of the heavy metal distribution in the coal samples analyzed.

Table III. Enrichment factor

Elements	S1	S2	S3	S4
Ti	1.4941	6.1743	16.2141	4.8981
V	0.0738	0.1688	0.1979	0.1062
Mn	1.5097	0.4701	0.2369	1.2721
As	0.0015	0.0059	0.0141	0.0027
U	0.0943	0.2363	0.6193	0.1321
Cr	0.0961	0.2130	0.3822	0.1018
Co	0.0863	0.7024	0.8450	0.0574
Zn	0.0801	0.3438	0.8392	0.0000
Sr	0.5298	0.0000	3.9921	0.0000
Sb	0.0000	0.0658	0.0000	0.0000
Ba	0.0000	0.0000	10.9258	2.5524
Th	0.1244	0.3110	0.5308	0.1716

Five contamination categories are generally recognized based on the enrichment factor: EF<2, depletion to minimal enrichment; 2≤EF<5, moderate enrichment; 5≤EF<20, significant enrichment; 20≤EF<40, very high enrichment; and EF>40, extremely high enrichment [16].

As shown in Table III, variation occurs in the enrichment factors values. Titanium reveals numerical values that range between minimal, moderate and significant enrichment while barium showed moderate and significant enrichment. Similarly, strontium and manganese indicated only moderate minimal enrichment respectively. However, V, As, U, Cr, Co, Zn and Th with mean values less than 1 indicated no enrichment. These variations could be due to natural and anthropogenic sources, such as natural weathering of the earth's crust, mining and agricultural practices (use of fertilizers).

The enrichment factor provides a quantitative measure of how much more abundant the element in the sample is compared to the background level. A value greater than 1 indicates enrichment, meaning the substance is more concentrated in the sample than in the background, while a value less than 1 indicates depletion [16]. It is used to assess the extent of pollutants in the coal.

Table IV presents the results of the pollution index/contamination factors in the coal deposits from the different mines in Kogi State, Nigeria.

Table IV. Pollution index/Contamination factor of INAA results

Elements	S1	S2	S3	S4
Ti	0.0968	1.8646	1.7265	2.9655
V	0.0482	0.0510	0.0211	0.0643
Mn	0.9867	0.1420	0.0252	0.7702
As	0.0010	0.0018	0.0015	0.0016
U	0.0616	0.0714	0.0659	0.0800
Cr	0.0628	0.0643	0.0407	0.0616
Co	0.0564	0.2121	0.0900	0.0347
Zn	0.0523	0.1038	0.0894	0.0000
Sr	3.4628	0.0001	4.2508	0.0001
Sb	0.0012	0.0199	0.0012	0.0012
Ba	0.0001	0.0001	1.1634	1.5453
Th	0.0813	0.0939	0.0565	0.1039

To evaluate the magnitude of contamination with heavy metals in the soil, the pollution index (PI) was introduced for

each metal. The pollution indices were calculated using the background value of the heavy metals.

The mean PI at the Okobo mine indicated that the soil was not polluted (PI<1) by Ti, V, As U, Cr, Co, Zn, Sb, Ba and Th, but moderately polluted (2<PI≤3) by Sr, while at Opoko-Obido, the mean pollution index reveals that the soil was not polluted (PI<1) by V, Mn, As, U, Cr, Co, Zn, Sr, Sb, Ba and Th, but slightly polluted (1<PI≤2) by Ti.

Also, at Odabgo, the mean PI showed that the soil was not polluted (PI< 1) by V, Mn, As, U, Cr, Co, Zn, Sb and Th, but mildly polluted (1<PI≤2) by Ti and Ba, and highly polluted (PI>3) by Sr, while at Ofugo, the mean pollution index suggests that the soil was not polluted (PI< 1) by V, Mn, As, U, Cr, Co, Zn, Sr, Sb and Th, but slightly polluted (1<PI≤2) by Ba, and moderately polluted (2<PI≤3) by Ti.

The pollution loads index for the different sampling points located at Okobo, Opoko-Obido, Odabgo and Ofugo mines are shown in Table V for the Instrumental Neutron Activation Analysis results. The pollution load index values in Table V are seen to be less than 1 (PLI < 1). This means there is no contamination in the soil of the sampling locations.

Table V. Pollution load index of INAA results.

S4	S5	S6	S7
0.081321	0.029606	0.070359	0.030723

IV. Conclusion

The elemental concentrations of toxic heavy metal impurities in coal samples obtained from the Okobo, Opoko-Obido, Odagbo and Ofugo coal mines in Kogi State, Nigeria have been determined using the Instrumental Neutron Activation Analysis (INAA) technique. The heavy metal concentrations vary significantly within the samples and between locations. The trend for the heavy metals concentrations was as follows: Sr > Mn > Ba > Cr > V > Co > U > Th > As > Sb for the INAA method. The concentration of Manganese (Mn) in samples S1 and S4 were slightly higher than the permissible limits of 150 mg/kg given by the Nigerian National Environmental Standards and Regulations Enforcement Agency (NESREA) and the United States Environmental Protection Authority (USEPA). Prolonged exposure to this manganese emission from the coal could cause memory loss, mood changes, cognitive impairment, respiratory and reproductive issues.

Similarly, the concentrations of Strontium (Sr) in S1 and S3 was slightly higher than the National Environmental Standards and Regulations Enforcement Agency (NESREA) limit of 300 mg/kg and lower than the United States Environmental Protection Authority (USEPA) and European Union limits of 670 and 500 mg/kg respectively. Variations in the enrichment factor values occur based on the enrichment factors. The numerical values of Ti and Ba have shown moderate to significant enrichment. Similarly, the Sr and Mn indicated only moderate minimal enrichment respectively. V, As, U, Cr, Co, Zn and Th with mean values less than 1 indicated no enrichment. These variations could be from natural and anthropogenic sources, such as natural weathering of the earth's crust, mining and agricultural practices (use of fertilizers and organic manure). In addition, the pollution index (PI) has shown that the soils around the coalmines were highly polluted by strontium (Sr), and slightly and moderately polluted by manganese (Mn), titanium (Ti) and barium (Ba). The enrichment factor and pollution index results are indications of the crustal origin and anthropogenic contribution that might partly result from weathering materials, coal mining, soil erosion and agricultural activities.

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