

## Quantitative Assessment of Heavy Metals Contamination in Nigerian Coal Using Instrumental Neutron Activation Analysis



\*<sup>1</sup>Uloko, F. O., <sup>2</sup>Agomuo, J. C. and <sup>2</sup>Agu, M. N.

<sup>1</sup>Department of Physics, Federal University Lokoja, Kogi State

<sup>2</sup>Department of Physics, Nigerian Defence Academy, Kaduna

\*Corresponding author's email: [felix.uloko@fulokoja.edu.ng](mailto:felix.uloko@fulokoja.edu.ng)

### ABSTRACT

The mining, processing and use of coal result in the accumulation of coal and coal ash, posing environmental and human health risks. In this study, the concentration of heavy metal distribution in the coal samples from selected coal mining sites (Maiganga, Gboko, Onyeama, Okobo, Opoko-Obido, Odagbo and Ofugo) in Nigeria were determined using the Instrumental Neutron Activation Analysis. The concentrations (mg/kg) of heavy metals ranged from  $4.69 \pm 0.92$  to  $18.40 \pm 1.10$  for V,  $4.82 \pm 0.29$  to  $185 \pm 2$  for Mn,  $0.11 \pm 0.01$  to  $0.33 \pm 0.04$  for As,  $0.19 \pm 0.04$  to  $0.74 \pm 0.05$  for U,  $7.14 \pm 1.62$  to  $16.60 \pm 1.80$  for Cr,  $1.49 \pm 0.16$  to  $9.10 \pm 0.33$  for Co,  $312 \pm 71$  to  $383 \pm 107$  for Sr,  $0.12 \pm 0.03$  to  $296 \pm 23$  for Sb,  $92 \pm 25$  to  $296 \pm 22$  for Ba, and  $0.26 \pm 0.04$  to  $2.39 \pm 0.11$  for Th. The concentrations of Mn, Sr, and Sb were slightly higher than the permissible limits given by the Nigerian National Environmental Standards and Regulations Enforcement Agency (NESREA) and the United States Environmental Protection Authority (USEPA). The study revealed that the coal from the mines was contaminated with heavy metals especially Mn and Ti thereby posing stern environmental and health concerns. The study therefore recommends that regulations and emission control technologies should be implemented to mitigate the release of these toxic metals during coal mining, processing, combustion, and waste management into the environment. In addition, the study suggests that workers should receive regular training on the proper use, maintenance, and disposal of personal protective equipment such as respirators, gloves, and eye protectors to maximize its effectiveness in reducing exposure.

### Keywords:

Coal,  
Metals,  
Neutron Activation  
Analysis,  
Samples.

### INTRODUCTION

Coal is a solid, combustible, sedimentary rock composed primarily of carbon, hydrogen, oxygen, nitrogen, sulfur, and minerals. It is a fossil fuel formed from the remains of ancient plants, mainly from the Carboniferous period, 300 – 360 million years ago (Benedict *et al.*, 2022). Coal is used for electricity generation, for steel manufacturing, cement production and as cooking fuel. Coal combustion contributes to climate change (greenhouse gas emissions), air pollution (particulates, sulfur dioxide, nitrogen oxides), water pollution (acid mine drainage), and land degradation (Al-areq *et al.*, 2008).

Coal is found in deposits called seams that originated through the accumulation of prehistoric plants remains in marshy environments that has undergone physical and chemical changes under intense temperature and pressure condition (Affolter *et al.*, 2015). Coal is

classified into four ranks: anthracite, bituminous, sub-bituminous and lignite. The ranking depends on the carbon contents and the amount of heat energy the coal can produce.

The burning process of coal produces sulfur and nitrogen that reacts with the atmospheric moisture to produce sulfuric and nitric acids (acid rain) which constitutes pollution (Ozoko, 2015). Pollutants from coal are known to have very harmful effects on major organs of the human body (Greenpeace, 2015). Similarly, the various stages of coal utilization ranging from mining of the coal to transportation and its combustion processes to disposal of the coal wastes, put humans and the environment at health risk. The use of coal fired power plants for example, leaves some high carbon footprints which leads to climate change, because they are the largest source of man-made CO<sub>2</sub>

emissions (Hannef *et al.*, 2016), and contributes over 83% of the CO<sub>2</sub> emission since 1990 (EIA, 2012). These negative environmental effects can be exacerbated by the presence of other impurities in the coal deposits such as heavy metals and radionuclide which are both bio-toxic and radioactive (Svnivasa *et al.*, 2005). Heavy metals are a class of elements distinguished by their high atomic numbers ( $Z > 30$ ) and masses, resulting in high densities, and a unique ability to absorb and scatter electromagnetic radiation, including X-rays (Jorgensen *et al.*, 2018). Even at relatively low concentrations, these heavy metals have been found to exhibit toxic properties and can cause a range of health effects (NIHES, 2020).

Heavy metals toxicity can lower energy levels and damage the functioning of the human brain, lung, kidney, heart, liver, blood composition, and other important organs (Engwa *et al.*, 2019). The diseases and health problems associated with these heavy metals poisoning due to coal includes: hypertension, cardiovascular problems, respiratory system issues, reproductive problems, decreased blood cell count, immune system deficiencies, decreased white blood cell count, hair loss, skin disorder, poor memory, blurred vision and sensitivity to light, physiological impairment, etc (Pyatha *et al.*, 2022). Examples of these pollutants are: arsenic (As), cadmium (Cd), lead (Pb), chromium (Cr), mercury (Hg), strontium (Sr), cobalt (Co), copper (Cu), zinc (Zn), antimony (Sb), manganese (Mn), nickel (Ni), and thallium (Tl) (Roy *et al.*, 2018).

Despite the potential environmental and health impacts, there are limited information on the levels of heavy metal contamination in Nigerian coal deposits. The exposure to these coal ash piles can cause health effects, making it a pressing concern. The present study has been able to determine the susceptibility of humans to heavy metal concentration in the coal deposits in Nigeria.

## MATERIALS AND METHODS

### Sample Collection

Seven (7) coal samples were collected from some active coal mines (Maiganga, Gboko, Onyeama, Okobo, Opoko-Obido, Odagbo, and Ofugo) in Nigeria. These sites were selected due to the reported cases of indiscriminate mining activities, contaminated water supply and continuous acid mine drainage contamination from underground coal mines. These regions in Nigeria are known for coal mining activities in the last decades and the coals are constantly transported from these mines to other parts of the country for the purpose of heat and electricity generation, cement production and other industrial processes.

Three samples taken at 0–5 cm, 5–10 cm, and 10–15 cm below the surface were collected from each sites of Maiganga, Gboko, Onyeama, Okobo, Opoko-Obido, Odagbo, and Ofugo to form the composite sample. The coal samples were collected using aluminium foil, wrapped and packed in polythene bags and transported safely to the laboratory at the Nigerian Geological Survey Agency (NGSA), Kaduna, Kaduna State for preparation.

These coal mines presented in the location map of Figure 1 has been carefully labelled using the format below:

Maiganga coal mine in Gombe State (S<sub>1</sub>)

Gboko coal mine in Benue State (S<sub>2</sub>)

Onyeama coal mine in Enugu State (S<sub>3</sub>)

Okobo coal mine in Ankpa local government area of Kogi State (S<sub>4</sub>)

Opoko Obido coal mine in Igalamela-Odolu local government area of Kogi State (S<sub>5</sub>)

Odagbo coal mine in Ankpa local government area of Kogi State (S<sub>6</sub>) and

Ofugo coal mine in Ankpa local government area of Kogi State (S<sub>7</sub>).

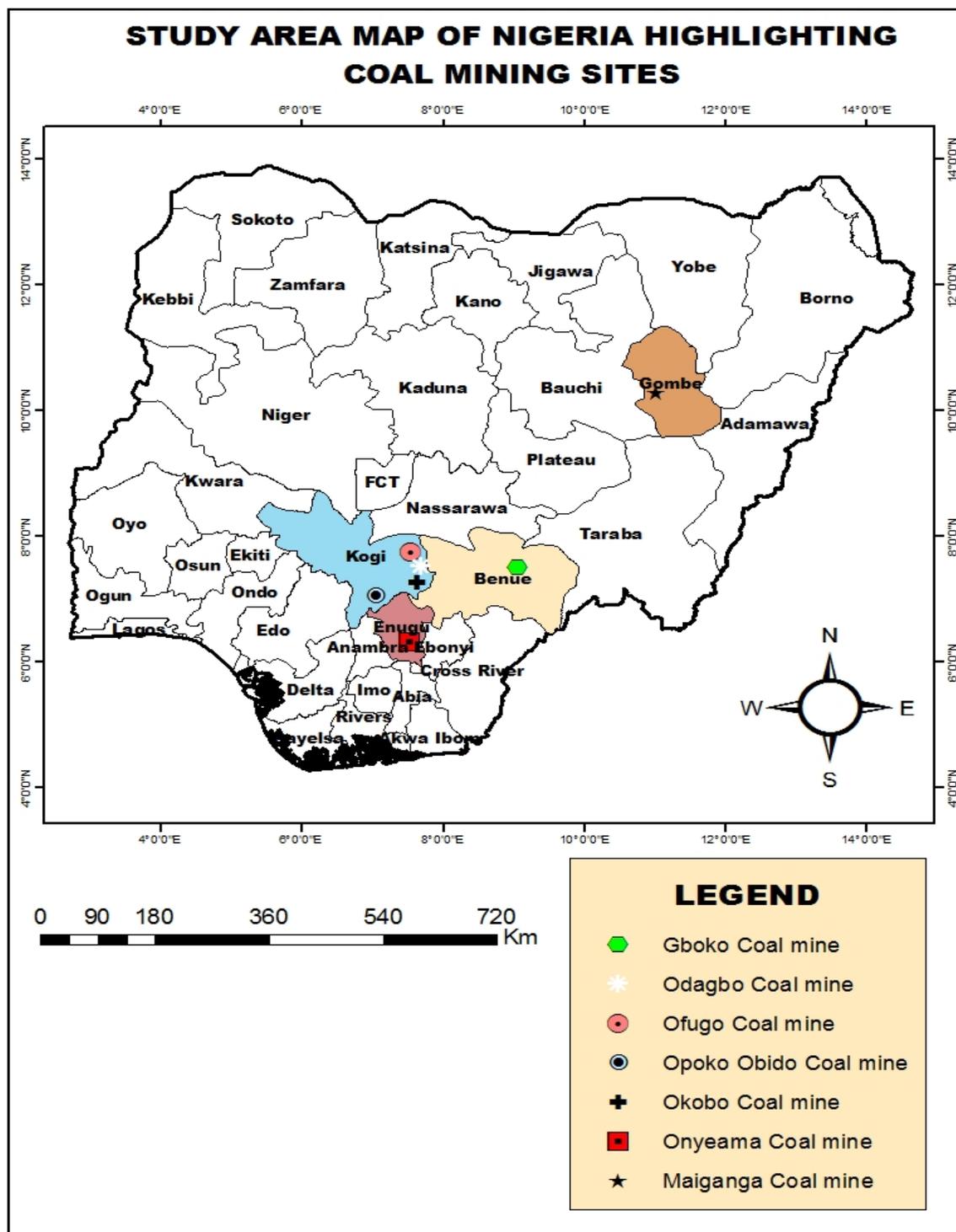


Figure 1: Study Area Map of Nigeria Showing Coal Mining Sites (Uloko *et al.*, 2024)

**Description of the Study Areas**

Maiganga is a community located between latitude 10° 02' to 10° 05' and longitude 11° 06' to 11° 08' in Akko local government area of Gombe State, northeast Nigeria (Kolo *et al.*, 2018). The Maiganga coal is

situated within the late Cretaceous Gombe formation, in the Gongola sub-basin of the Northern Benue Trough of Nigeria (Kolo *et al.*, 2016). The Maiganga coal deposit is a low rank, sub-bituminous coal resource, identified

by the Nigerian government as a key target for future power generation initiatives.

The Gboko coal mine is situated in Benue State, Nigeria, within the Gboko Local Government Area. Geographically, it lies at  $7.3162^{\circ}$  N latitude and  $8.9017^{\circ}$  E longitude (Kankara *et al.*, 2020). The region is notable for its rich mineral deposits, including limestone, granite, barite, and alluvial clay (Kankara *et al.*, 2020).

The Onyeama coal mine is geographically situated within the latitudes  $6^{\circ} 29' - 6^{\circ} 34' N$  and longitudes  $7^{\circ} 30' E$ , and it is located within Enugu coal field, specifically within the catchment area of the Ekulu River (Ozoko, 2015). The Onyeama coal is situated approximately 6.5 kilometers northwest of Enugu city, in Enugu State, southeastern Nigeria (Ozoko, 2015). The Onyeama coal mine is an underground mining operation that extracts sub-bituminous coal from the seams 2, 3, and 4 of the Mamu Formation, located at the base of the Enugu Escarpment (Ozoko, 2015).

Similarly, the geographical location of the Okobo coal mine lies between latitude  $7^{\circ} 22' 14'' N$  and longitude  $7^{\circ} 37' 31'' E$  in Enjema district area of Ankpa local government which is about 200 km North of Enugu having coal reserves amounting to 380 million tonnes (Itodo *et al.*, 2020), while in Igalamela-Odolu local government area of Kogi State, precisely in Egabada community which is surrounded to the East by Enugu State and to the west by the Niger River is the Opoko-Obido coal mine with a longitude of  $7^{\circ} 1' 15'' E$  and a latitude of  $7^{\circ} 2' 36'' N$  having an abundance of coal deposits covering approximately  $26 \text{ km}^2$  (Uloko *et al.*, 2024).

Consequently, in Ankpa local government specifically Okaba district, the Odagbo coal mine is situated in the North-eastern part of the Anambra Basin with an altitude of about 275 m, longitudes  $7^{\circ} 43' 30'' E$  to  $7^{\circ} 44' 00'' E$  and a latitudes  $7^{\circ} 28' 30'' N$  to  $7^{\circ} 29' 00'' N$ . The mine in this district is about 0.8 m thick of bituminous coal with an overburden of between 3 m to 6 m. The coal is characterized by a dark colour, having a light grey silty shale that over-lies (Momoh *et al.*, 2021). Similarly, in Ankpa local government, is the Ofugu coalmine located and has a longitude of  $7^{\circ} 37' 24.7'' E$ , latitude of  $7^{\circ} 33' 36.9'' N$  and an altitude of 390m (Uloko *et al.*, 2024).

### Sample Preparation

The coal samples were pulverized into fine powder, sieved with a mesh of diameter 2mm, 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 coal samples were transported to the Centre for Energy Research and Training (CERT), Ahmadu Bello University (ABU) Zaria for the Instrumental Neutron Activation Analysis (INAA)

### Method of Analysis

In the laboratory at the Centre for Energy Research and Training (CERT) of Ahmadu Bello University (ABU) Zaria, the homogenized samples of coal were weighed (0.15 g), transferred to a 0.5 mL polyethylene vial (irradiation capsule) then sealed. The same process was done for 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 encapsulated in a cladding and subjected to neutron irradiation using the Instrumental Neutron Activation Analysis (INAA) technique. At the Centre for Energy Research and Training (CERT), Ahmadu Bello University, Zaria, Kaduna State, we used a miniature neutron source (NIRR-1) reactor with low enriched uranium core, light water as moderator and beryllium as reflector. The NIRR-1 was operated using the control console, the microcomputer control system, and the rabbit system through which the samples were introduced into the irradiation position, which is in the moderator. The maximum thermal neutron flux in the core is approximately  $1 \times 10^{12} \text{ n. cm}^{-2} \cdot \text{s}^{-1}$  and the neutron flux at the irradiation position where the samples were irradiated is  $5 \times 10^{11} \text{ n. cm}^{-2} \cdot \text{s}^{-1}$  at half power.

The procedure used for the Instrumental Neutron Activation Analysis (INAA) at the Centre for Energy Research and Training (CERT), Zaria was encapsulating the coal samples and the standard reference sample in heat sealed polyethene quartz vials and the comparative method was used to determine many elements simultaneously by irradiating the standard reference material (NIST 1633c coal fly ash) and the coal samples in the same neutron flux and were counted in the same geometry. The other standard reference material (NIST 1633c coal fly ash) was used to calibrate the spectrometer machine to generate the correction factor used for the calculation of the concentration of the elements in the coal samples.

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), aluminum (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.

All measurements for the Instrumental Neutron Activation Analysis (INAA) were performed using a high purity germanium (HPGe) semiconductor detector with relative efficiency of 43.4% and a resolution of

1.80 keV (FWHM) at 1332.5 keV. Photopeaks were analysed using Genie-2000 (Canberra, Inc) software that smooth the spectral data and determined the net areas of gamma ray photopeaks. The program then translated the area into count rates (counts per minute). This program was capable of resolving overlapping of complex photopeaks energy regions, like the decay time differences, electronic dead time losses and unresolved interferences.

## RESULTS AND DISCUSSION

The elemental concentration (mg/kg) of heavy metals distribution in the coal samples obtained from the Instrumental Neutron Activation Analysis (INAA) of the samples from the Maiganga coal, Gboko coal, Onyeam coal, Okobo coal, Opoko-obido coal, Odabgo coal and Ofugo coal, that are of health concern to humans because of their toxicity are presented in Table 1.

**Table 1: Heavy Metals Concentration (mg/kg) in the Coal Samples using INNA Technique**

Elements	Locations						
	S1	S2	S3	S4	S5	S6	S7
Ti	BDL	646 ± 123	605 ± 132	707 ± 109	1350 ± 138	1250 ± 105	2147 ± 150
V	BDL	10.16±1.68	4.69±0.92	13.8±0.9	14.59±1.24	6.03±0.66	18.4±1.1
Mn	164±2	162±25	4.82±0.29	237±2	34.1±0.7	6.06±1.17	185±2
As	0.33±0.02	0.15±0.02	0.11±0.01	0.18±0.02	0.33±0.04	0.28±0.04	0.30±0.03
U	0.29±0.04	0.48±0.06	0.19±0.04	0.57±0.05	0.66±0.07	0.61±0.09	0.74±0.05
Cr	7.14±1.62	12.2±1.4	BDL	16.2±1.5	16.6±1.8	10.5±1.6	15.9±1.6
Fe	6985±188	4813±164	953±80	6856±178	3168±146	1117±115	6351±178
Co	BDL	3.58±0.27	4.17±0.22	2.42±0.19	9.10±0.33	3.86±0.28	1.49±0.16
Zn	28.4±3.5	BDL	BDL	12.3±2.9	24.4±3.7	21±3	BDL
Sr	326±86	BDL	BDL	312±71	BDL	383±107	BDL
Sb	296±23	0.12±0.03	BDL	BDL	0.17±0.05	BDL	BDL
Ba	296±22	92±25	BDL	BDL	BDL	131±19	174±19
Th	1.10±0.10	1.9±0.10	0.26±0.04	1.87±0.09	2.16±0.12	1.30±0.09	2.39±0.11

BDL means Below Detection Limit.

The reported uncertainty was calculated from counting statistics and is not the normal standard deviation on replicate analyses.

The results of the heavy metals concentrations (mg/kg) obtained from the Instrumental Neutron Activation Analysis of the samples collected from Maiganga coal, Gboko coal, Onyeama coal, Okobo coal, Opoko-obido coal, Odabgo coal, and the 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 (mg/kg) of these heavy metals ranged from  $4.69 \pm 0.92$  to  $18.40 \pm 1.10$  for V,  $4.82 \pm 0.29$  to  $185 \pm 2$  for Mn,  $0.11 \pm 0.01$  to  $0.33 \pm 0.04$  for As,  $0.19 \pm 0.04$  to  $0.74 \pm 0.05$  for U,  $7.14 \pm 1.62$  to  $16.60 \pm 1.80$  for Cr,  $1.49 \pm 0.16$  to  $9.10 \pm 0.33$  for Co,  $312 \pm 71$  to  $383 \pm 107$  for Sr,  $0.12 \pm 0.03$  to  $296 \pm 23$  for Sb,  $92 \pm 25$  to  $296 \pm 22$  for Ba, and  $0.26 \pm 0.04$  to  $2.39 \pm 0.11$  for Th. The results

showed that the heavy metal concentrations varied within the coal samples and between locations.

The concentration of Manganese (Mn) in samples S1, S2, S4 and S7 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). However, it is lower than the European Union limit of 100 mg/kg. This high concentration of Mn in coal when compared with the findings of Al-areqi *et al.*, 2008, Ezemokwe *et al.*, 2016, and Chilikwazi, *et al.*, 2023, were in good conformity.

Similarly, the concentration of Strontium (Sr) in S1, S4 and S6 were 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 mg/kg and 500 mg/kg respectively. The findings of Damastitu *et al.*, 2020 and Srikanth *et al.*, 2018 showed similarity in terms of high concentration of Sr in coal.

Furthermore, Antimony (Sb) concentration in sample S1 was greater than the National Environmental Standards and Regulations Enforcement Agency (NESREA), United States Environmental Protection Authority (USEPA) and European Union permissible limits of 3.0 mg/kg, 6.0 mg/kg and 2.5 mg/kg respectively. This result is closely related to the findings of Altikulac *et al.*, 2022, and Chand *et al.*, 2019.

The distribution of heavy metals in the different sampling locations of Maiganga, Gboko, Onyeama, Okobo, Opoko, Odabgo, and Ofugo coal mines are shown graphically in Figure 2. At the Odagbo mining location, almost all the heavy metals were observed to be present at high concentrations. In addition, from the Figure 2, Sr, Sb and Ba, indicated homogeneity in

concentration in S1. Similarly, V and Cr showed homogeneity in S2, S4 and S5. In addition, Mn, V and Co showed homogeneity in S3. Also, V and Mn indicated homogeneity in S6. Similarly, Mn and Ba showed homogeneity in S7. while As, U and Th showed minimal variations with their maximum levels.

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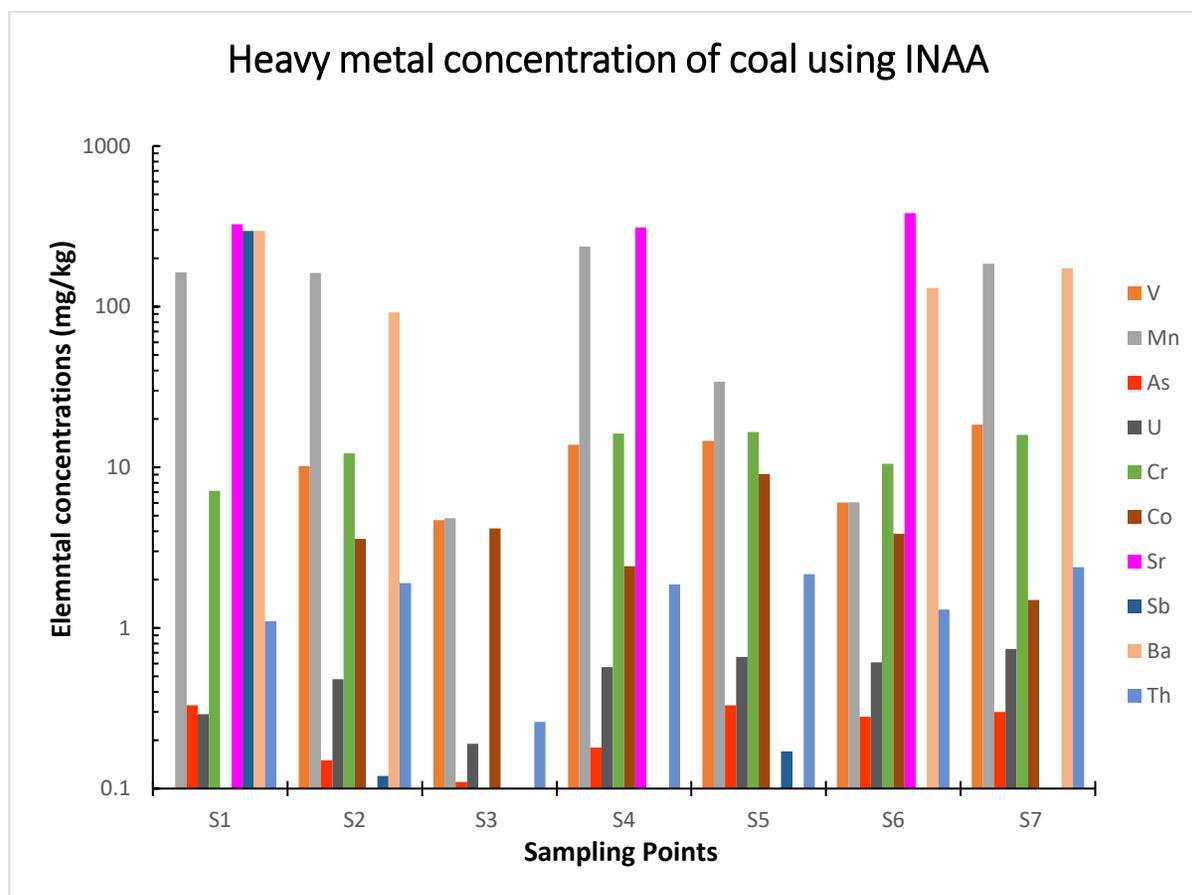


Figure 2: Heavy Metals Concentrations of Coal Using the INAA Technique

## CONCLUSION

The elemental concentrations of toxic heavy metal contamination in the coal obtained from the Maganga, Gboko, Onyeama, Okobo, Opoko-Obido, Odagbo and Ofugo coal mines in Nigeria have been determined

using the Instrumental Neutron Activation Analysis (INAA) nuclear technique. The results of the analysis showed that the concentration of Manganese (Mn) in samples S1, S2, S4 and S7 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). Similarly, the concentration of Strontium (Sr) in S1, S4 and S6 were slightly higher than the National Environmental Standards and Regulations Enforcement Agency (NESREA) limit of 300 mg/kg. Furthermore, Antimony (Sb) concentration in sample S1 was greater than the National Environmental Standards and Regulations Enforcement Agency (NESREA), the United States Environmental Protection Authority (USEPA) and European Union permissible limits of 3.0 mg/kg, 6.0 mg/kg and 2.5 mg/kg respectively. The study revealed that the coal from the mines was contaminated with heavy metals especially Mn and Ti thereby posing stern environmental and health concerns. The study therefore recommends that regulations and emission control technologies should be implemented to mitigate the release of these toxic metals during coal mining, processing, combustion, and waste management into the environment. In addition, the study suggests that workers should receive regular training on the proper use, maintenance, and disposal of personal protective equipment such as respirators, gloves, and eye protectors to maximize its effectiveness in reducing exposure.

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