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Heavy Metal Contamination in Abuni and Adudu Lead and Zinc Mines in Southern Nasarawa State, Nigeria

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ABSTRACT
Heavy metal pollution of the environment is becoming a significant issue and a major concern due to the negative consequences it has globally. Artisanal mining, the rapidly expanding mining industries, agriculture, the metal industries, inappropriate waste management, and the use of fertilizers and pesticides are all contributing factors to the release of these inorganic pollutants into the land, water, and environment. This study investigated the concentrations of some heavy metals (Al, Cd, Cr, Cu, Fe, Mn, and Zn) in soil samples from Abuni and Adudu lead and zinc mines, Southern Nasarawa State, Nigeria. Two spots were chosen from each of the mining sites, and soil samples were taken at the topsoil, one foot, and two feet above the ground level, respectively, to achieve a fully representative sampling of the area. The soil samples were processed and analyzed for each metal using instrumental neutron activation analysis. The concentrations of the metals were in the order: Cd < Cu < Cr < Mn < Al < Zn < Fe in Abuni mine and Cd < Cu < Cr < Mn < Al < Fe < Zn in Adudu mine. The heavy metals, Al, Cr, and Cu, fall within the recommended limits. The levels of Cd, Fe, Mn, and Zn in Abuni and Adudu mines were higher than the permitted limits of 3 mg/kg, 50,000 mg/kg, 200 mg/kg, and 421 mg/kg, respectively, set by the Food and Agricultural Organization of the United Nations (FAO) and the Nigerian National Environmental Standards and Regulations Enforcement Agency (NESREA) for Nigerian soils. The study revealed that the soil samples from the mines are contaminated with heavy metals, especially Cd, Fe, and Mn, raising serious environmental and health concerns. It is worth noting that the extremely noticeable values for Zn in all the evaluated indices simply confirm its existence in commercial quantities, as both sites are mining zinc metal.

INTRODUCTION

Human activities such as industrial, mining, logging, petroleum activities, smoke from motor vehicles, coal power plants, domestic waste, and agricultural activities cause contamination of metals and rare earth elements into the environment. On the other hand, anthropogenic and naturally occurring processes such as weathering of rocks and volcanic eruptions have a significant effect in enriching the ecosystem with metals (Veena *et al.,* 1997). Heavy metals are prominent environmental contaminants because of their toxicity, propensity to persist in the atmosphere, and capacity for bioaccumulation within the human body. Toxic heavy metal pollution of terrestrial and aquatic ecosystems is a serious environmental issue with health implications for the general public (Saikat *et al.,* 2022). Generally, heavy metals are described as inorganic metallic elements and metalloids, which are characterized by being above 5 g/cm^3 in density. Those that are commonly mentioned and considered daily include titanium, vanadium, chromium, manganese, iron, cobalt, nickel, copper, zinc, arsenic, molybdenum, silver, cadmium, tin, platinum, gold, mercury, and lead (Briffa *et al.,* 2020). Heavy metals can be classified into three categories: radionuclides (silver, gold, uranium, thorium, and plutonium); vital metals (iron, zinc, copper, manganese, cobalt, and strontium) that are required in trace levels for human health; and toxic metals (mercury, lead, arsenic, cadmium, chromium, and thallium). Given that mining operations may have negative effects on the environment and human health, heavy metal contamination of the soil is a cause for concern. Additionally, high exposure to antimony and chromium promotes carcinogenicity (Sun *et al.,* 2015; Sunder and Chakravarty, 2010), lead poisoning causes intellectual abnormalities in children (Hou *et al.,* 2013), mercury toxicity causes Minamata disease, and cadmium poisoning causes Itai-Itai disease. The risks of heavy metals are generally greater than their benefits. Therefore, individuals must relocate away from industrial locations with high levels of heavy metal pollution (Saikat *et al.,* 2022). Over 20 million hectares of land are affected by heavy metals, including Zn, Pb, Ni, As, Hg, Cu, Cd, and Cr (Liu *et al.,* 2018). Pollution of the environment by heavy metals hinders the efficient activities of microbes, plants, animals, humans, and the agricultural ecosystem (Elanga *et al.,* 2022).

Although several heavy metals contamination studies have been conducted, the majority of these studies mainly focused on dumpsites (e.g. Ojiego *et al.,* 2022, Akoto and Anning, 2021, Karkarna and Mujahid, 2021, Opaluwa *et al.,* 2012, Nyiramigisha *et al.,* 2021, Nasir and Rakiya, 2015, Raymond and Felix, 2011, Parvez *et al.,* 2023, Bykowszezenko *et al.,* 2006, Ho *et al.,* 2010). Almost no information is available from the published literature discussing metal contamination in the Southern Nasarawa mining districts. Therefore, to further provide baseline information on their environmental concerns, this study was conducted to assess the amounts of heavy metals in soil samples taken from the lead and zinc mines at Abuni and Adudu in southern Nasarawa State, Nigeria.

MATERIALS AND METHODS

The study area

Nasarawa State is located in North Central Nigeria. It is bordered to the East by the States of Taraba and Plateau, to the North by Kaduna, to the South by the States of Kogi and Benue, and to the West by the Federal Capital Territory (FCT), Abuja. The State is on Latitude 8° 34' 13.854''N and Longitude 8° 18' 31.8384''. Nasarawa State is known and considered Nigeria’s home of solid minerals. It is endowed with so many natural mineral resources such as barite, copper, granite, tantalite, zinc, gold, quartz, limestone, diamond, talc, gypsum, calcite, topaz, apatite, and a host of other minerals. Abuni and Adudu lead and zinc mines are located in Awe and Obi local government areas in Nasarawa South Senatorial District. Their geographical coordinates are 8° 13' 30.9''N, 009° 00' 59.8''E and 8° 13' 59.4''N, 009° 01' 09.3''E respectively.

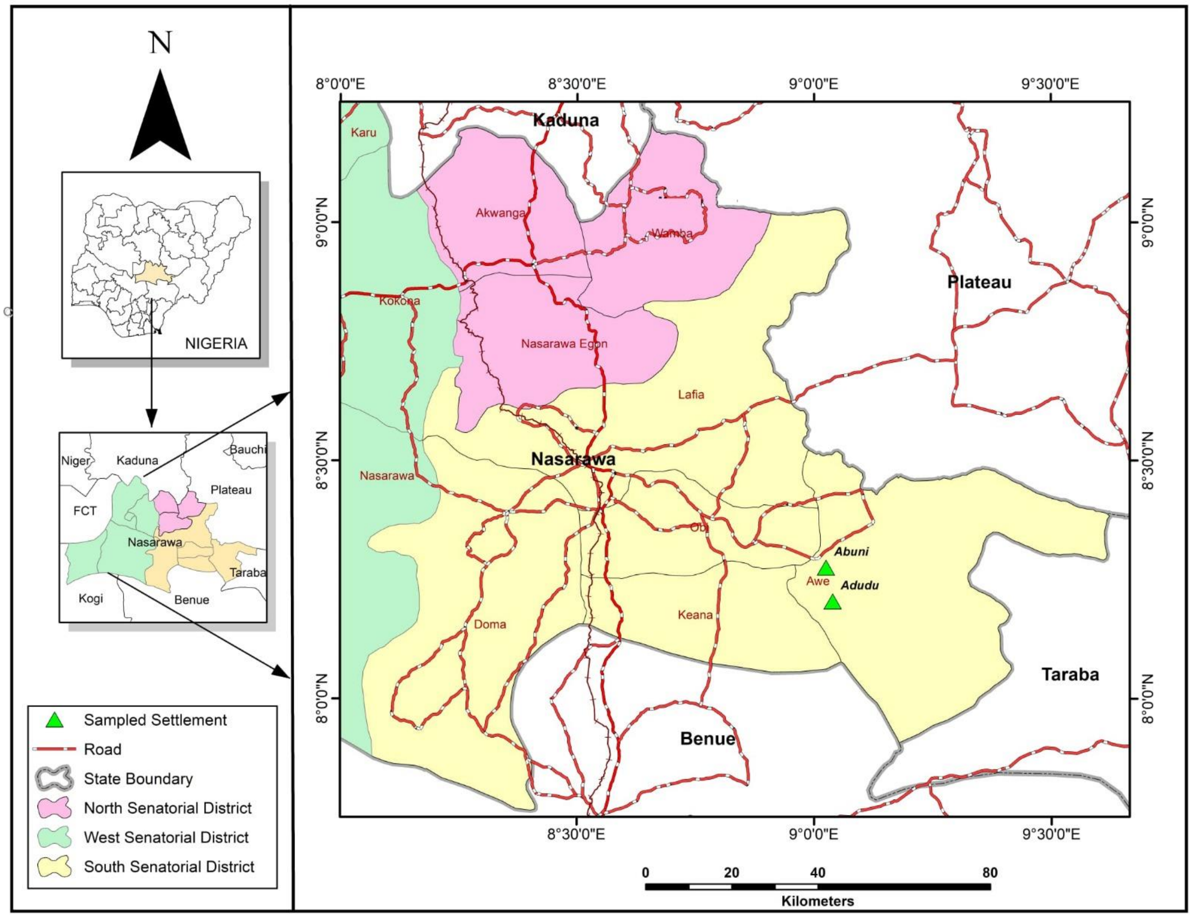

Figure 1: Study Area Map of Nasarawa State, Nigeria, showing Abuni and Adudu mines

Materials

For this study, the following materials were used: plastic containers, masking tape, marker, polythene vial, a miniature neutron source reactor for irradiating samples, a high purity germanium detector for detecting and measuring gamma radiation, tissue paper and acetone for cleaning to prevent cross-contamination of samples, a polythene bag and aluminium foil for collecting samples, a mortar and pestle, and a 2 µm sieve.

Sample Collection, Preparation, and Irradiation

A total of six (6) soil sample from each of the mines was collected from two (2) points at the topsoil, 1 foot, and 2 feet to form a composite sample. These sites were selected due to the increasing mining activities which has created a mini-settlement with a small market where the locals and miners gather every day without observing any safety measure, poor hygiene and incessant use of dynamite explosives to break the rocks and other earth formation in search of these precious minerals thereby increasing the level of both air and land pollution that necessitates additional research and led to the current study. The samples, after being gathered using aluminium foil, were securely transported to the Centre for Energy Research and Training (CERT), Ahmadu Bello University (ABU), Zaria, where they were prepared and subjected to instrumental neutron activation analysis (INAA). The collected samples were rid of stones, pebbles, and shrubs. It was dried under ambient conditions, then ground into a fine powder using a mortar and pestle and sieved with a 2 µm sieve, and labelled. The homogenized soil samples were weighed (0.15 g), transferred to a mL polythene vial, and sealed. The same encapsulation process was repeated for the standard reference material (SRM) NIST 1633c. Subsequently, the soil samples and the SRM (NIST 1633c) were put into an irradiation capsule and irradiated through the instrumental neutron activation analysis method.

Method

The analysis was conducted using the Nigeria Research Reactor-1 (NIRR-1), a miniature neutron source that uses beryllium as a reflector, light water as a moderator, and 13% low-enriched uranium as fuel (Anas *et al.,* 2023). It is especially employed in limited radioisotope generation and elemental analysis. The International Atomic Energy Agency (IAEA) Lake sediment, IAEA 158 (treated in the same way as the sample), was used to calibrate the NAA computational software (Winspan Ver 2.0) before the analysis. This produced the relative calibration factor, which was then used to determine the concentration of the elements of interest. A short-lived element irradiation scheme and a long-lived element irradiation scheme were employed. Irradiation was applied for 60 seconds, and 10 seconds was immediately counted for the short-lived elements. Following the counting, the sample was left to decompose for two to three hours before being tallied once more for 600 seconds. Following the brief irradiation, the elements magnesium (Mg), aluminium (Al), calcium (Ca), titanium (Ti), vanadium (V), manganese (Mn), and dysprosium (Dy) were found. For elements having a long half-life, the samples were exposed to radiation for six hours, counted for 1800 seconds after three days, and then allowed to decay for seven more days before being recounted for 3600 seconds. Among the elements found were sodium (Na), potassium (K), 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), europium (Eu), and thorium (Th).

An overview of the irradiation and counting process for the INAA technology employed in this investigation is shown in Table 1.

Table 1: Overview of INAA Conditions Used in this Study

Procedure	Elements	Nuclide	Half-life	γ-energies (keV)
	Mg	^{27}Mg	9.4 min.	1014.4
Short irradiation (60 s)	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	K	^{42}K	12.4 hrs	1524.6
	Mn	^{56}Mn	2.58 hrs	846.8
	Eu	^{152}Eu	13.5 year	841.6
	Dy	^{165}Dy	2.334 hrs	94.7
Long irradiation(1800s)	Na	^{24}Na	15 hrs	1368.6
	As	^{6}As	26.24 hrs	559
Cooling: 3 - 7 days	Br	^{82}Br	33.9 hrs	776.5
	Fe	^{59}Fe	45.1 days	1099.3
Counting1800s-3600s	Cr	^{51}Cr	27.80 days	320.1
	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

Data Analysis

The overall data obtained for the two sets of samples were interpreted using appropriate descriptive and inferential statistical techniques such as the contamination factor (CF), enrichment factor (EF), geo-accumulation index (I_{geo}), and pollution index (PI).

Contamination factor (CF)

This is employed to ascertain the soil's level of pollution. Using (1), the contamination factor, CF, was calculated.

$$CF = \frac{C_m\ Sample}{C_m\ Background} \quad (1)$$

where C_m Sample = concentration of the metal at the contamination site C_m Background = background value of the heavy metal

CF values >1 and <1 indicate deterioration of the site soil quality and no pollution, respectively (Asubiojo *et al.,* 2012).

Enrichment factor (EF)

To determine the level of concentration of specific elements in rocks and soil samples in comparison to the reference or background value, (2) was used

$$EF = \frac{\left(C_m/C_r\right)sample}{\left(C_m/C_r\right)crust} \quad (2)$$

where C_m and C_r represent concentrations of the metal and reference metal in the soil sample and reference sample (Ho *et al.,* 2010). In this study, Fe is chosen as the normalizing (reference) element while determining EF-values since in wetlands, it is mainly supplied from soil or sediments and is one of the widely used reference elements Chakravarty and Patgiri, 2009 and Addo *et al.,* 2012). However, Ekaete *et al.* (2015) posited that several researchers have also used Si and Al as reference elements for calculating the enrichment factor (EF).

Geo-accumulation Index (I_{geo})

The enrichment of metal concentration above baseline concentration was calculated using the equilibrium equation (3) as proposed by Nikolaidis *et al.* (2010).

$$I_{geo} = \log_2\left(\frac{C_m}{1.5B_n}\right) \quad (3)$$

where C_m is the concentration of the metal in the soil samples, B_n is the background value and 1.5 is the background matrix correction factor due to lithogenic effects.

Pollution Index (PI)

According to Asubiojo *et al.,* (2012), a pollution index is a numerical metric used to evaluate the degree of pollution in a certain environment, such as the air, water, or soil. It gives the concentration of hazardous pollutants in the environment's soil a numerical representation. In addition to protecting the environment and human health from the negative impacts of toxic metal contamination, the heavy metals pollution index in soil is a useful instrument for evaluating and tracking the environmental impact of mining-related activities (Asubiojo *et al.,* 2012). The pollution index (PI) is the proportion of the concentration of the element of interest in the sample to the element's maximum permissible level. This can be evaluated using (4):

$$PI = \frac{C_i}{m_i} \quad (4)$$

where C_i is the concentration of each metal (mg/kg), m_i is the maximum permissible level or background value (mg/kg) of the metal.

If an element's pollution index value is higher than unity, it indicates that the element has contaminated the sample to a large degree and may be harmful at the level it is present (Asubiojo *et al.,* 2012). To inform regulatory decisions, assess the efficacy of pollution mitigation strategies, and increase public awareness of environmental issues, the PI evaluation aims to educate policymakers, scientists, environmentalists, and the general public about the health and environmental quality risks associated with soil pollution control efforts.

RESULTS AND DISCUSSION

Concentration of Heavy Metals in Soil Samples

For many reasons, including normalizing neutron flux variability, irradiation and decay times, accounting for detection efficiency and geometry, and making it possible to solve for the unknown sample concentration straightforwardly, it is essential to analyze both the sample and a standard in the relative NAA method to ensure accurate and reliable quantification of elemental concentrations. In essence, the standard reference material serves as a calibrated reference and a methodology for evaluating experimental variables. The concentrations of heavy metals in soil samples from Abuni and Adudu lead and zinc mines in Southern Nasarawa State, Nigeria, as presented in Table 2, revealed the presence of all the tested metals at varying concentrations. The order of heavy metals concentration at Abuni and Adudu mines was Cd < Cu < Cr < Mn < Al < Zn < Fe and Cd < Cu < Cr < Mn < Al < Fe < Zn, respectively. This result indicates that the highest and lowest metals in both study sites were Fe and Cd, and Zn and Cd, respectively.

Table 2: Concentration of Heavy Metals in Soil Samples from Abuni and Adudu Mines

Metal (mg/kg)	Abuni mine	Adudu mine
Al	9762 ± 499	12,880 ± 116
Cd	4.04 ± 0.11	3.78 ± 0.12
Cr	21.60 ± 2.30	21.00 ± 2.20
Cu	14.50 ± 1.60	17.60 ± 1.80
Fe	118,200 ± 946.00	104,100 ± 833.00
Mn	4,704 ± 9.00	6,257 ± 13.00
Zn	27,640 ± 27.00	250,300 ± 2503

Contamination Factor of the Heavy Metals

The contamination factor (CF) values as presented in Table 3 are described using the following terms: CF < 1 suggests a low contamination of the soil; 1 ≤ CF < 3 suggests a moderate contamination; 3 ≤ CF < 6 suggests a considerable contamination, and CF > 6 suggests a very high contamination.

Table 3: Contamination Factor in Soil Samples from Abuni and Adudu Mines

Elements	Abuni mine	Adudu mine
Al	0.12	0.16
Cd	48.89	42.00
Cr	0.24	0.23
Cu	0.50	0.63
Fe	3.02	2.66
Mn	10.73	14.27
Zn	412.54	3735.82

As shown in Table 3, variation occurs in the contamination factor values. Al, Cr, and Cu reveal no contamination of the soil. Fe reveals moderate-to-considerable contamination of the soil. Cd and Mn reveal a very high contamination of the soil. The skyrocketed numerical value for zinc only confirms its abundance in commercial quantities, as both sites are lead and zinc mines.

Enrichment Factor of the Heavy Metals

The enrichment factor values as presented in Table 3 are interpreted as suggested by Ho *et al.,* (2010) where EF ≤ 1 indicates no enrichment; 1 < EF ≤ 3 indicates minor enrichment; 3 < EF ≤ 5 indicates moderate enrichment; 5 < EF ≤ 10 indicates moderately severe enrichment; 10 < EF ≤ 25 indicates severe enrichment; 25 < EF ≤ 50 indicates severe enrichment, and EF > 50 indicates extremely severe enrichment.

Table 4: Enrichment Factor in Soil Samples from Abuni and Adudu Mines

Elements	Abuni mine	Adudu mine
Al	0.04	0.05
Cd	16.21	13.93
Cr	0.08	0.08
Cu	0.17	0.21
Fe	1.00	1.00
Mn	3.57	0.47
Zn	13.68	1238.95

As shown in Table 4, variation occurs in the enrichment factor values. Mn and Cd reveal numerical values that range between moderate enrichment to severe enrichment. However, Al, Cr, and Cu with values less than 1 indicate no enrichment. The high enrichment values for Zn confirm its abundance in commercial quantities whilst it is been mined and processed in both mines. EF 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 element is more concentrated in the sample than in the background, while a value less than 1 indicates depletion. EF is also used to assess the extent of pollutants in the soil (Srikanth and Naga Raju, 2019).

Geo-accumulation Index (I_{geo}) in Soil Samples from Abuni and Adudu Mines

The geo-accumulation index (I_{geo}) values are presented in Table 4 and classified into seven categories (Nikolaidis *et al.,* 2012) where $I_{geo} < 0$ indicates No contamination; $0 < I_{geo} \leq 1$ indicates light to moderate contamination ¸ $1 < I_{geo} \leq 2$ indicates moderate contamination; $2 < I_{geo} \leq 3$ indicates moderate to strong contamination; $3 < I_{geo} \leq 4$ indicates strong contamination; $4 < I_{geo} \leq 5$ indicates strong to extreme contamination; $5 < I_{geo}$ 10 indicates extreme serious contamination.

Table 5: Geo-accumulation Index (Igeo) in Soil Samples from Abuni and Adudu Mines

Elements	Abuni mine	Adudu mine
Al	-3.70	-3.30
Cd	5.00	4.80
Cr	-2.70	-2.70
Cu	-1.60	-1.30
Fe	1.00	0.80
Mn	2.80	3.30
Zn	8.10	11.30

As shown in Table 5, variation occurs in the geo-accumulation index values. Al, Cu, and Cr reveal numerical values of no contamination or unpolluted (Igeo < o). The soils were light to moderately polluted by Fe in both mines (I_{geo} = 0.8 and I_{geo} = 1), moderately to strongly polluted by Mn in Abuni (Igeo = 2.80), and strongly polluted by Mn in Adudu (Igeo = 3.30), Strong to extremely polluted by Cd in both mines (Igeo = 5.0 and Igeo = 4.80). Again, Zn confirms its abundance in commercial quantities in both mines.

Pollution Index (PI) in Soil Samples from Abuni and Adudu Mines

To evaluate the magnitude of contamination with heavy metals in the soil, a pollution index (PI) was introduced for each metal. The pollution indices were calculated by comparing the values of the elements in the contaminated soil samples with their standard permissible limits (for Nigerian soils) as presented in Table 6 (Asubiojo *et al.*, 2012). The results show that Abuni and Adudu mines were not polluted by Al, Cu, and Cr (PI < 1), but moderately polluted (2 < PI ≤ 3) by Cd and Fe in both mines and very highly polluted (PI > 6) by Mn in both mines. Once again, the extremely high values for Zn only confirm its abundance in commercial quantities, as both sites are lead and zinc mines.

Table 6: Pollution Index (PI) in Soil Samples from Abuni and Adudu Mines

Elements	Abuni mine	Adudu mine
Al	0.03	0.04
Cd	1.33	1.26
Cr	0.22	0.21
Cu	0.15	0.18
Fe	2.36	2.08
Mn	23.52	31.29
Zn	65.65	594.54

CONCLUSION

Instrumental Neutron Activation Analysis (INAA) has been employed to identify the elemental concentrations of harmful heavy metals in soil samples taken from the Abuni and Adudu lead and zinc mines in Southern Nasarawa State, Nigeria. Significant amounts of Al, Cd, Cr, Cu, Fe, Mn, and Zn were discovered, according to this investigation. All of the heavy metals were above the NESREA-recommended acceptable limits for Nigerian soils, except Al, Cr, and Cu. The levels of Cd, Fe, Mn, and Zn in Abuni and Adudu mines exceeded the allowable limits of 3 mg/kg, 50,000 mg/kg, 200 mg/kg, and 421 mg/kg, respectively, set by the Food and Agricultural Organization of the United Nations (FAO) and the Nigerian National Environmental Standards and Regulations Enforcement Agency (NESREA) for Nigerian soils. Long-term exposure to cadmium can lead to cardiovascular problems, renal damage, bone diseases, and an increased risk of some cancers, especially prostate and lung cancer. Long-term exposure to iron can cause substantial health problems and damage to organs, including the liver, heart, and brain. Long-term exposure to Mn can cause manganism, a persistent neurological condition that resembles Parkinson's disease and manifests as tremors, walking difficulties, and facial muscle spasms. Long-term zinc exposure can cause anemia, nausea, and vomiting. It may also interfere with the absorption of iron and copper. The contamination factor causes variations in the contamination factor values. According to the Fe numerical values, the soil is moderately contaminated. On the other hand, the soil is highly contaminated with Cd and Mn. Depending on the enrichment factor, differences arise in the values of the enrichment factor. Numerical values for Cd and Mn indicate that these metals have moderate to severe enrichment in the soil. According to variations in the geo-accumulation index, the Abuni mine is moderately to severely polluted by Mn, the Adudu mine is strongly polluted by Mn, and both mines are strongly to extremely polluted by Cd. According to the pollution index values, the soil in both mines has very high levels of Mn pollution and moderate levels of Cd and Fe contamination. The results of the pollution index and enrichment factor show the crustal origin and the anthropogenic input, which may be partially caused by mining and weathering materials. It is worth noting that the extremely noticeable values for Zn in all the evaluated indices simply confirm its existence in commercial quantities, as both sites are mining zinc metal.

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