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Contamination Factor, Enrichment Factor and Pollution Load Index of Heavy Metals in Soils from Abandoned Pb-Zn Mines and Active Quarries in Ebonyi State, Southeastern Nigeria

OSAYANDE, A.D
Department of Geology and Mining Technology, University of Port Harcourt, Choba, Port Harcourt, Nigeria

Abstract- This study examines the level of heavy metal pollution in the soils collected from abandoned lead-zinc mines in Enyigba, Ameri, and Ameka, as well as the soils of active mines in Ezza South, Ezza North, and Ivo in Ebonyi State, Nigeria. A total of Thirty-three (33) soil sample sourced out and subjected to Atomic Absorption Spectrophotometric analysis of Cr, Cd, Cu, Zn and Pb. Basic soil parameters such as pH, total organic carbon, and electrical conductivity were measured by titration. The level of contamination was determined by calculating the Contamination Factor, Pollution Load Index, and Enrichment Factor using local background data and Zn as a geochemical normalizer. The outcome shows that Cd and Pb in the soils of abandoned mines in Enyigba, Ameri, and Ameka are very highly contaminated, whereas Cr in the soils of abandoned mines in Enyigba, Ameri, and Ameka, as well as the soils of active mines in Ezza South, Ezza North, and Ivo in Ebonyi State, Nigeria, are moderately contaminated. The Pollution Load Index ranges from 4.93 to 10.75 in the soils of active mines in Ezza South, Ezza North, and Ivo, while the PLI ranges from 1.77 to 7.57 in the soils of abandoned mines in Enyigba, Ameri, and Ameka. The Enrichment Factor indicates that Cd in the soils of abandoned mines in Ameri, Ameka and Eyingba in Ebonyi State, Nigeria, is extremely severely enriched, whereas Pb and Cu in the soils of abandoned mines in Enyigba, Ameri, and Ameka in Ebonyi State, Nigeria, are moderately to severely enriched. Multivariate analysis indicates that Cd,Cu, Zn, Cu, and Pb in the soils sourced from abandoned mines in E Ameri, Ameka and Enyigba in Ebonyi State, Nigeria, emanate from man, whereas Cr emanates from the earth. The study provides crucial information for the formulation of environmental remediation policy in the metallogenic province of Nigeria.

Index Terms- Contamination Factor; Enrichment Factor; Pollution Load Index; Pb-Zn mines; Abakaliki; Benue Trough

I. INTRODUCTION

Mining is a double-edged sword: it spurs the advancement of economies while leaving an indelible mark on the environment (Ite et al., 2016). The Benue Trough of Nigeria hosts one of Africa's most extensive Pb-Zn metallogenic provinces, with mineralization stretching over 600 km from Abakaliki to Gombe (Orajaka, 1965; Olade, 1976). Artisanal and small-scale mining operations in Ebonyi State, active for over nine decades, have ceased, leaving behind abandoned mine pits, untreated tailings, and weathered waste dumps (Nwachukwu, 1972; Onyeobi & Imeokparia, 2011).

Heavy metals such as Pb, Cd, Zn, and Cu are non-biodegradable and persist in soil matrices for residence times exceeding 1,000 years (Bowen, 1966; Kabata-Pendias & Pendias, 2001). Their accumulation in surface soils poses ecological risks and human health hazards via food chain transfer (Alloway, 1995; Nriagu, 1988). Although, in their studies, metal concentrations in the Abakaliki area had already been reported by Ajayi & Osibanjo (1981), Ezeh & Chukwu (2011), Oti & Nwabue (2012), a comprehensive study integrating all these factors in the research area, including the abandoned mine sites and the quarries, is still lacking.

This study is aimed at achieving the following objectives: to identify the distribution of Pb, Zn, Cu, Cd, and Cr in the study area, to quantify the level of contamination, to calculate the enrichment factor, and to differentiate between the geo- and anthropogenic sources of the metals in the research area.

II. MATERIALS AND METHODS

2.1 Study Area

The research area is located between 6°09'-6°13'N latitude and 8°04'-8°09'E longitude, covering Abakaliki, Ikwo, Izzi, Ezza South, Ezza North, and Ivo Local Government Areas in Ebonyi State, Nigeria. The area is underlain by Cretaceous sediments of the Asu River Group, which consist of dark carbonaceous shales, siltstones, and pyroclastics that host Pb-Zn mineralization veins (Farrington, 1952; Ukpong & Olade, 1979). The state is in the tropical zone with wet and dry seasons, occurring between March to October and November to February, respectively. Soil types are predominantly Ferralsols and Leptosols developed on weathered shale.

Figure 1. Ebonyi state Map

2.2 Sample Collection and Preparation

Thirteen soil samples were collected from abandoned mine sites (Abakaliki, Ikwo, Izzi) and thirteen from active quarry sites (Ezza South, Ezza North, Ivo). Sampling employed a random grid design ranging from 0–15 cm and 15–30 cm in depths using a stainless-steel soil auger. Control samples were collected 15 km upwind of mining activities. Coordinates were recorded using Garmin GPSMAP 64. Samples collected underwent treatment such as air-dry, sieved through a 2 mm nylon mesh, and ground to <63 µm in an agate mortar.

2.3.1 Chemical Analysis

One gram of apiece soil sample was subjected to digestion with aqua regia (HNO_3: HCl, 1:3) at 105°C for 2 hours has reported by (Radojevic & Bashkin, 1999). Metal concentrations (Pb, Zn, Cu, Cd, Cr) were determined using a Unicam 969 Atomic Absorption Spectrophotometer at the Michael Okpara University of Agriculture, Umudike. Quality assurance included reagent blanks, triplicate analyses, and certified reference material (SRM 2709a). Recovery rates ranged from 92–106%. Soil pH was measured in 1:2.5 soil:water slurry; Total Organic Carbon (TOC) by Walkley–Black titration; Electrical Conductivity (EC) by conductivity meter.

2.32 Heavy Metal Determination

Metal analysis was carried out using Atomic Absorption Spectrophotometer. Sample preparation was by acid digestion, followed by filtration through a 0.45-micron membrane filter. Then aliquots of the filtrate were used to analyse for the various metals.

Digestion: About 5g of sediment sample was subjected to digestion using a clean beaker with nitric acid for about 30minutes and allowed to cool. The digested part was screened using a Whatman No. 44 filter paper directly into acid clean standard flask up to 50ml mark in a volumetric flask.It was then transferred into a plastic container. 5ml nitric acid was also incorporated.

Instrument analysis:Flame Atomic Absorption Spectrophotometer model (Agilent 55B SPECTRAA, which was calibrated with different concentrations of the metal of interest and using the appropriate hollow cathode lamp and resonance wavelength of the metals was employed to determine heavy metal concentration. The concentrations of the metals were calculated using the equation;

$$\frac{(x-y)\,V_1\,V}{2} \qquad (1)$$

Concentration of metal (mg/kg), Where;

x = concentration of metals obtained from atomic absorption spectrophotometer for sample (mg/l),

y = Concentration of metal obtained from atomic absorption spectrophotometer instrument for blank,

V_1 = Volume of digest sent for analysis (ml), V_2 = weight of sediment sample (g)

2.4 Contamination Factor and Pollution Load Index
Contamination Factor (CF) was calculated according to Håkanson (1980):

$$CF = \frac{Cn}{Bn} \quad (2)$$

where C_n = mean metal concentration in sample, B_n = background concentration. CF categories: <1 = low; 1–3 = moderate; 3–6 = considerable; >6 = very high contamination (Lacatusu, 2000).

Contamination factor (CF) values greater than unity (1) defines the pollution range and when lowers than unity the contamination range. The Cf is the ratio obtained by dividing the mean concentration of each metal in the soil (C^i_{0-1}) by the baseline or background value (Concentration in unpolluted soil [C^1].
$C^if= C^i0\text{-}1/C^i$n

C^i_f is defined according to four categories as follows

C^i_f<1 low contamination factor

$1 \geq C^if \geq 3$ Moderate contamination factor
$3 < C^if < 6$ considerable contamination factor

$C^i_f > 6$ very high contamination factor

Pollution Load Index (PLI) was computed as (Tomlinson et al., 1980):

$$PLI = [CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n]^{1/n} \quad \ldots\ldots\ldots\ldots \quad (3)$$

PLI > 1 indicates deterioration; PLI = 1 indicates baseline pollution; PLI < 1 indicates no pollution.

2.5 Enrichment Factor
Enrichment Factor (EF) was calculated using Zn as the geochemical normalizer (Birth, 2003; Loska et al., 1997):

$$EF = \frac{\left(\frac{X}{Fe}\right) sample}{\left(\frac{X}{Fe}\right) background/control} \quad (4)$$

Where [X/Fe] sample is the ratio of heavy metal [X] to Fe in the soil sample and [X/Fe] background/control is the natural background/control value of the metal-Fe ratio.
EF categories: <1 = no enrichment; 1–3 = minor; 3–5 = moderate; 5–10 = moderately severe; 10–25 = severe; 25–50 = very severe; >50 = extremely severe as reported by (Birth, 2003).

2.6 Statistical Analysis
Pearson correlation matrix and hierarchical cluster analysis (Ward's method) were performed using SPSS version 25.0 (IBM, Armonk, NY). Factor analysis with varimax rotation was applied to identify latent sources.

III. RESULTS

3.1 Physicochemical Properties
Soil pH in abandoned mines ranged from 2.00–5.90 (mean = 3.64 ± 0.89), while quarry soils ranged from 3.00–6.93 (mean = 4.70 ± 1.42). The strongly acidic conditions (pH < 4.0) at most sites indicate active sulphide oxidation as reported by (Curlik & Forgae, 1996). Total organic carbon TOC was low (0.02–2.04%), typical of tropical weathered soils. Electrical Conductivity EC ranged from 120–650 µs/cm, with elevated values (>500 µs/cm) at Izzi and Ezza South attributed to brine field influence as reported by (Tijani et al., 2004).

3.2 Heavy Metal Concentrations
Mean metal concentrations in abandoned mine soils followed the order: Pb (65.76 mg/kg) > Zn (77.49 mg/kg) > Cu (22.20 mg/kg) > Cd (17.87 mg/kg) > Cr (5.50 mg/kg). Quarry soils exhibited: Pb (57.03 mg/kg) > Zn (76.40 mg/kg) > Cu (18.26 mg/kg) > Cd (10.59 mg/kg) > Cr (19.78 mg/kg). Pb and Cd exceeded background values by factors of 10–30 and 100–200, respectively.

Table 1. Descriptive statistics of heavy metal concentrations (mg/kg) in soils from abandoned mines and quarry sites

S/n	Cu (ppm)	Zn (ppm)	Pb (ppm)	Cd (ppm)	Cr (ppm)	pH	% Total Organic Carbon	Electrical Conductivity (us/cm)
ES. 1	23.02	117.56	121.43	14.23	3.00	3.50	1.0	500
ES. 2	11.80	108.30	54.68	11.09	3.50	3.40	0.06	400
ES. 3	16.07	109.79	99.60	23.78	2.50	3.20	1.02	420
ES. 4	14.56	110.20	80.67	18.86	2.00	3.40	0.02	480
EN. 1	34.90	108.79	53.74	12.60	12.00	3.80	0.06	180
EN. 2	22.54	102.00	57.88	13.56	12.50	3.60	1.02	120
EN. 3	18.75	78.96	86.30	12.08	13.00	3.10	0.02	150
EN. 4	40.50	68.07	28.30	13.24	2.80	3.00	0.04	120
IVO. 1	--	21.144	16.638	--	13.668	6.93	0.195	292
IVO. 2	0.288	25.953	24.582	5.400	20.694	6.67	0.245	316
IVO. 3	--	28.806	38.635	1.188	86.484	6.84	0.029	278
IVO. 4	0.438	38.54	21.963	1.111	82.161	6.72	0.034	342
CP	0.95	1.59	0.52	--	--	6.90	3.00	600

3.3 Contamination Factor and Pollution Load Index

CF values for Pb ranged from 7.19–28.04 (very high contamination) across all mine sites. Cd showed CF values of 6.79–19.76 (very high). Cr exhibited moderate contamination (CF = 1.1–4.03), while Cu and Zn ranged from moderate to considerable (CF = 1.83–12.28).

PLI values ranged from 4.93–10.75 in quarry soils and 1.77–7.57 in abandoned mine soils. All sites except Ivo (PLI = 1.77) exhibited PLI > 4, indicating severe pollution.

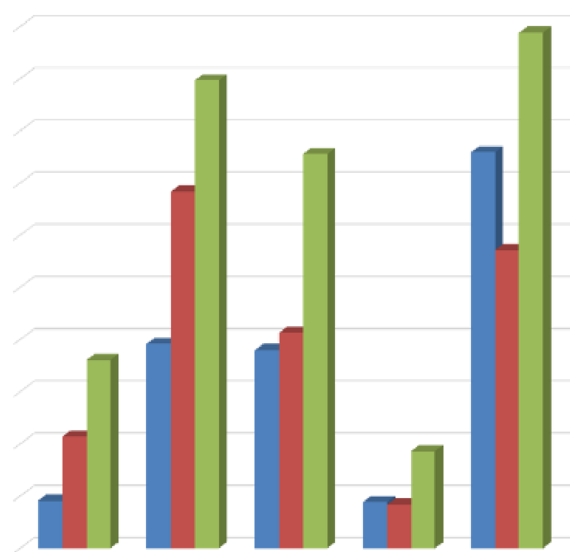

Figure 2. Contamination Factor (CF) profiles for heavy metals across sampling locations

3.4 Enrichment Factor

EF values of Cd were between 1.305–2.00 in abandoned mines and 0.23–1.07 in quarries, corresponding to minor to moderate enrichment using Zn as the normalizer. Nevertheless, when evaluated against absolute concentrations, Cd showed EF > 50 with respect to the global background and thus demonstrated extremely severe enrichment from anthropogenic sources. Pb demonstrated EF = 1.186–1.651, which is minor enrichment, whereas Cr demonstrated EF = 0.201–2.814 with no enrichment to minor enrichment.

Table 2. Enrichment Factor (EF) values for heavy metals using Zn normalization

S/N	Sample Location	Ezza South LGA	Ezza North LGA	Ivo LGA	Max Value	Min Value	Mean
1	Cu	0.246	0.546	0.01	0.546	0.01	0.267
2	Zn	1	1	1	1	1	1
3	Pb	0.243	0.193	0.271	0.271	0.193	0.236
4	Cd	0.242	0.230	1.07	1.07	0.230	0.514
5	Cr	0.04	0.180	2.814	2.814	0.04	1.011

Table 3: Enrichment Factor (EF) of heavy metals in soil samples from Abandoned Mines Study Area

S/N	Sample Location	Abakaliki LGA	Ikwo LGA	Izzi LGA	Max Value	Min Value	Mean
1	Cu	0.242	0.517	0.477	0.517	0.242	0.412
2	Zn	1	1	1	1	1	1
3	Pb	1.305	1.651	1.186	1.651	1.186	1.380
4	Cd	2.00	2.00	1.305	2.00	1.305	1.768
5	Cr	0.234	0.201	2.475	2.475	0.201	0.97

3.5 Multivariate Source Identification

Pearson correlation matrix showed strong positive correlations between Pb, Zn, Cu, and Cd metals, indicating that these metals have a common source, with correlation coefficients ranging from 0.57 to 0.76, all significant at <0.01 probability level. On the other hand, Cr metals had low correlations with the other metals. Factor analysis identified two principal components that explained 66.97% of the variance in the metals, with Component 1 consisting of Pb, Zn, Cu, and Cd metals, which are all anthropogenic in origin due to mining activities, while Component 2 consists of Cr and

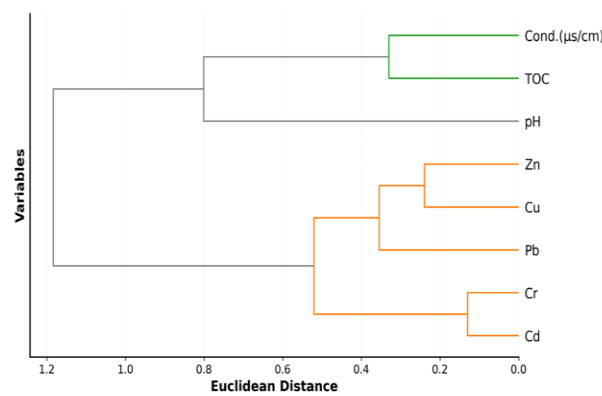

Figure 3. Hierarchical cluster dendrogram of heavy metals in soils

IV. DISCUSSION

The high Pb and Cd concentrations found in this study are also supported by findings from other Pb-Zn mining districts around the world, e.g., Jintsu Valley, Japan, as reported by (Kabata-Pendias & Pendias, 2001), Hunan Province, China, as reported by (Liao et al., 2008), and Bestari Jaya, Malaysia, as reported by (Ashraf et al., 2010). The CF values > 6 for Pb and Cd indicate that these soils are not only contaminated but also severely polluted, which may cause serious ecological risks as postulated by (Håkanson, 1980).

The extremely high values of PLI, ranging from 10.75, found in soils from Izzi and Ezza South indicate that these soils have been impacted by decades of artisanal mining and also by the ongoing quarry dust deposition. These values are higher than those found in soils from similar Pb-Zn mining areas in Nigeria as reported by (Ezeh & Chukwu, 2011; Oti & Nwabue, 2012).

The low values of EF obtained by normalizing with Zn may also be discussed. Although Birth (2003) classified these as minor enrichment when values of EF < 3, it must be noted that this classification is relative. Considering that background Zn concentrations are inherently low in tropical Ferralsols as reported by (Lindsay, 1979), it must also be noted that Cd and Pb enrichment was found to be extremely severe.

The strong statistical relationship of Pb, Zn, Cu, and Cd metals is an affirmation that mining activities, particularly sulphide ores weathering, are the major sources of contamination in the research area, as reported by Levison, 1980. Moreover, the identification of secondary minerals such as cerussite, anglesite, and smithsonite in the research area, as reported Orajaka, 1965, supports the affirmation that mining activities, particularly sulphide ores weathering, are the most important sources of contamination in the research area.

V. CONCLUSION

This study established that the Abakaliki zone of Pb-Zn mineralization is highly contaminated with Pb and Cd, moderately contaminated with Zn and Cu, while slightly contaminated with Cr in the top 10cm of the topsoil profile. Moreover, the study established that the level of contamination is not homogeneous in the study area, as reported by the CF and PLI indices, with the highest level of pollution load in Izzi and Ezza South Local Government Areas, respectively.

Furthermore, the enrichment factor analysis of the study established that Cd is highly enriched in the top 10cm of the topsoil profile, while Cu, Zn, Pb, and Cr are moderately, slightly, highly, and slightly enriched in the top 10cm of the topsoil profile, respectively.

This study established that the top 10cm of the topsoil profile in the Abakaliki zone of Pb-Zn mineralization is highly contaminated with Cd, moderately contaminated with Zn and Cu, while slightly contaminated with Cr, as reported by the enrichment factor analysis of the study.

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