JCARM
Journal of Contemporary Academic Research and Methodologies

An Online Peer Reviewed / Refereed Journal
Volume 1 | Issue 6 | July 2026
ISSN: 3139-7247 (Online)
Website: www.journal.jcarm.in

Original Research Article

ASSESSMENT OF SELECTED HEAVY METAL CONTAMINATION, MODIFIED DEGREE OF CONTAMINATION AROUND LEAD ZINC MINE IN THE ENYIGBA, SOUTHEASTERN NIGERIA.

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Article No: 008 | **Accepted**: 21 July 2026 | **Published**:10 August 2026

Abstract: This paper focuses on heavy metal (Pb, Zn, Cu, Cd and As) contamination, modified degree of contamination in the soils of Enyigba, southeastern Nigeria. Pollution by these metals was examined using several calculation methods: modified degree of contamination and geoaccumulation index. The proposed modified calculation method of degree of contamination, which involves the determination of the arithmetic mean of contamination factors by individual studied metals, revealed moderate contamination to very high contamination by metals. And the modified degree of contamination was moderate to ultra-high. These results were further supported by values obtained from indices of geoaccummulation method of pollution assessment. The spatial distributions of the three main controlling processes- rock weathering/water-rock interaction, mining activities factor and salinity factor across the study area were revealed. This evaluation would serve to create a consistent calculation method as a contribution towards building a consensus on methodologies applicable to the study of polluted soils.

Keywords: Contamination Factor, Modified Degree Of Contamination, Spatial Distributions, Geoaccumulation Index

 https://doi.org/10.5281/zenodo.21858858

Introduction

The investigation of heavy metal deposition and accumulation has attracted growing attention because of the increasing recognition that heavy metals present in soils pose significant risks to both human health and the environment (Egbueri & Enyigwe, 2022; Abrahams, 2002). The behavior, mobility, chemical speciation, and solubility of heavy metals are strongly influenced by soil characteristics. Even when certain contaminants remain relatively immobile in soil and are not readily taken up by plants, they may still threaten human health through alternative exposure pathways such as geophagy and the inhalation of contaminated dust (Sheppard, 1998). The study area is an active mining region where agricultural activities are also extensively practiced, and the combined effects of these activities have adversely affected both the environment and the livelihoods of the local population. Soil contamination by heavy metals arises from both natural and anthropogenic sources. Natural processes, including the weathering of parent rocks, contribute to background metal concentrations, whereas mining, agricultural practices, industrial operations, and improper waste disposal significantly increase heavy metal accumulation in soils. Environmental contamination studies commonly assess the extent of soil pollution by determining heavy metal concentrations and comparing them with established guideline values or standard classification systems (Abrahim et al., 2008). Over the past several decades, numerous methods have been developed to identify and evaluate the severity of heavy metal contamination in soils. Likewise, different pollution assessment indices have been widely applied by researchers to determine contamination levels in soils and sediments across various regions of the world (Wuana et al., 2011). Although extensive research has been conducted on environmental contamination by a wide range of pollutants (Sezgin et al., 2023), considerable differences remain in the methods used to quantify the degree of contamination at polluted sites. Therefore, this study aims to establish baseline concentrations of Zn, Pb, Cu, Cd, and As in soils surrounding the Enyigba Pb–Zn mining area in Nigeria; determine the extent of heavy metal contamination using these baseline values; apply and compare different contamination assessment methods; and propose a suitable approach for estimating the overall degree of enrichment of multiple heavy metal pollutants. It is anticipated that the findings of this study will contribute to the development of a more consistent and reliable methodology for assessing heavy metal contamination in polluted soils.

Materials and Methods

2.1 The Study Area And Geographic Description

The study was conducted in Enyigba, Ebonyi State, Nigeria (longitudes $8^0\ 00^1$ and $8^0 12^1$ east, latitude $6^0 00^1$ and $6^0\ 14^1$ north) with an elevation of 20-70 metres above the sea level. Covering an area of about $15km^2$. The Enyigba lead zinc mine started its full operation more than four decades ago. No environmental mitigation impact assessment or reclamation of the devastated land was carried out; hence, no mitigation programs were put in place. The soils of the Enyigba area are predominantly derived from the weathering of shales and are classified as ultisols and inceptisols. (Egbueri & Enyigwe, 2020).

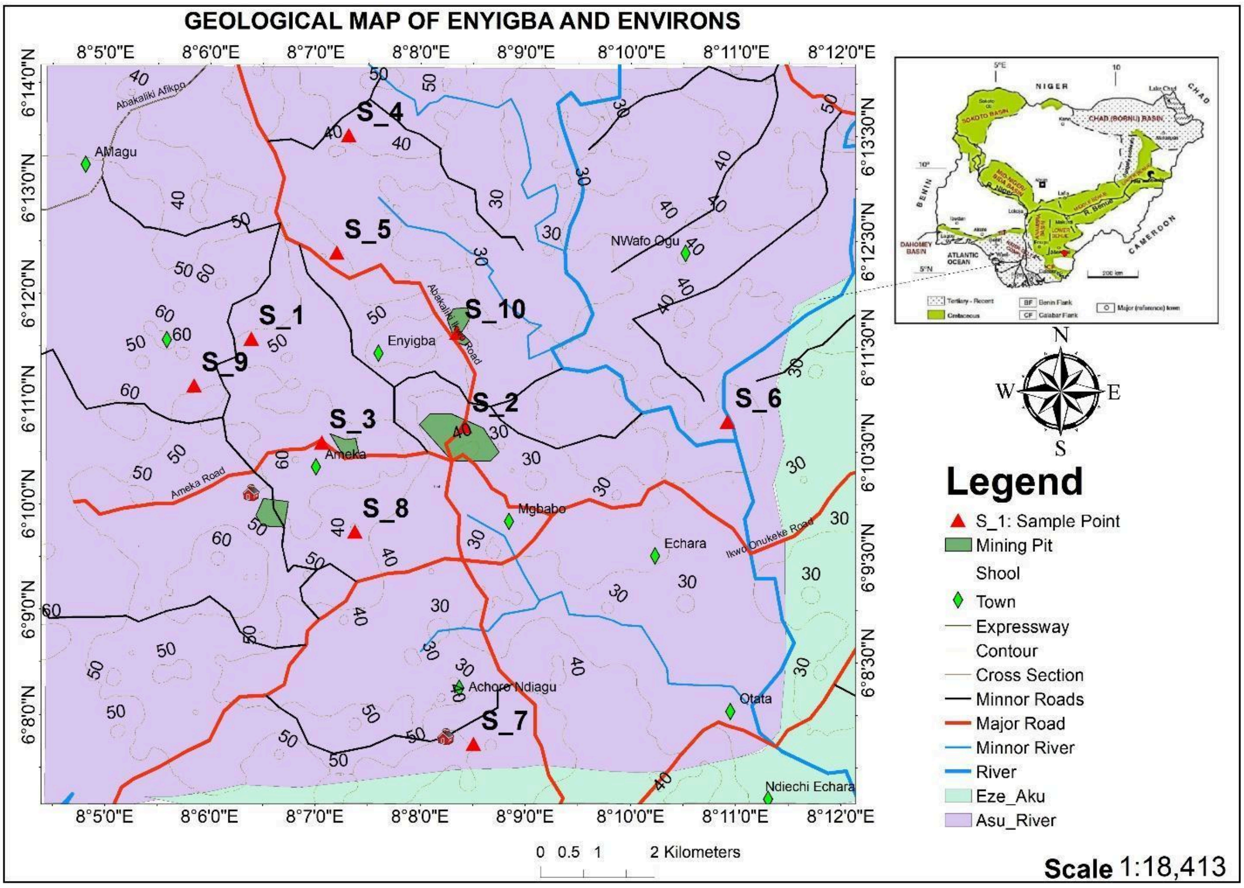

Figure 1: Geologic map of the study area showing sampling locations

The area experiences a tropical climate characterized by distinct wet (April to October) and dry (November to March) seasons. Annual rainfall averages within a range of 1,750mm to 2,000mm, while mean annual temperature ranges 27^0C to 36^0C with relatively high humidity all through the year. These climatic conditions influence soil development, weathering processes and the environmental behavior of heavy metals in the area. The area is drained by the perennial Ebonyi River and its ephemeral tributaries and is characterized by the dendritic drainage system, which is a function of the lithology.

2.2 Sampling Technique And Preparation

Surface and subsurface soils of up to 30cm deep were collected using a hand-driven stainless steel auger, following a random and judgmental sampling technique. Exact locations for all sampled sites were determined using a global positioning system and entered into a geographic information system for data processing. In the laboratory, the collected soil samples were sieved through a 500µm stainless steel mesh wire. Samples of 0.5g were digested in 20ml freshly prepared aqua regia (1:3$HN0_3$; HCI) on a hot plate for 4 hours, then evaporated and analyzed for metal concentration. Standard reference materials were prepared using stock solutions from SAARCHM and MERCH and used to check on the accuracy of the results. The total concentration of Pb, Zn, Cd, Cu and As in filtrates was then determined using a flame atomic absorption spectrometer (Varian Spectra AA 220FS) using an air-acetylene flame.

2.3 Data Analysis

In order to evaluate and assess heavy metal contamination and the degree of contamination in soils, analytical data were subjected to pollution calculation methods with the aim of bringing realistic estimates for the amount of contamination that has impacted soil around Enyigba, southeastern Nigeria.

2.3.1 Modified Degree Of Contamination, Mcd

The degree of contamination in soil by metals was calculated based on the method presented by (Abrahim, G.M.S. 2005)

$$\text{mCd} = \frac{1}{N} \sum_{1=1}^{N} CFi \text{ .... (1)}$$

Where N is the number of elements analyzed, and CF is the contamination factor calculated as shown in (2)

$$\text{CF} = \frac{Cm\ sample}{Cm\ background} \text{ .... (2)}$$

Where Cm sample and median Cm background in (2) are as described before, it is important to note that equation 1 is a modified and generalized form of the degree of contamination (Cd) formula, in equation (3) proposed by Hakanson L (1980).

$$\text{Cd} = \frac{1}{N} \sum_{1=1}^{N} CFi \text{ .... (3)}$$

The aim of calculating the contamination factor is to actually provide a measure of the degree of overall contamination of surface layers in a sampled site.

2.3.2 Geoaccumulation Index

The index of geoaccumulation really enables the assessment of contamination by comparing the current levels of metal concentrations and the original pre-industrial concentrations in the soils. Effects of mining activities and farming were assessed following the geoaccumulation index approach proposed by Muller (1969), also given in equation (5)

$$\text{Igeo} = \log_2 \left[\frac{Cm\ sample}{1.5\ x\ Cm\ background}\right] \text{ .... (5)}$$

The factor 1.5 is introduced in equation 5 to minimize the effect of possible variations in the background values, Cm background, which may be attributed to lithologic differences.

2.3.3 Statistical Analysis

Correlation of different elements in soil samples was calculated, and principal component (PC) analysis was performed to establish possible factors that contributed towards the metal concentrations and source apportionment.

2.3.4 Principal Component Analysis

Table 1 presents detailed results of the principal component analysis carried out on the data from the present study. The three extracted components account for the mean, skewness, sample variance and eigenvalues of each principal component, with a factor loading ≥ 0.45 being considered for each component.

Table 1: Principal Component Scores Of The Tested Data Set.

Sample Location	*Component 1*	*Component 2*	*Component 3*
S1	-0.069728	0.225546	**-0.509546**
S2	**0.929614**	**-0.83442**	0.0342237
S3	0.170133	**0.82188**	**-0.582812**
S4	**1.54934**	**4.28417**	-0.232066
S5	**-1.20263**	**-1.10096**	**-0.77208**
S6	**0.454439**	**-1.66634**	**1.49273**
S7	**0.521385**	0.086855	0.342747
S8	**-0.971249**	**0.964105**	-0.068948
S9	**-1.20263**	0.231408	0.305981
S10	**-1.04733**	**0.520416**	**0.597287**
Mean	0.01194	0.502969	0.27956
Variance	0.97081	2.62442	1.186818
Eigen value	4.79557	3.72863	2.0601

The spatial distributions of the three main controlling processes – rock weathering/water - rock interactions, mining activities factor and salinity factor across the study area are presented in Figure 2

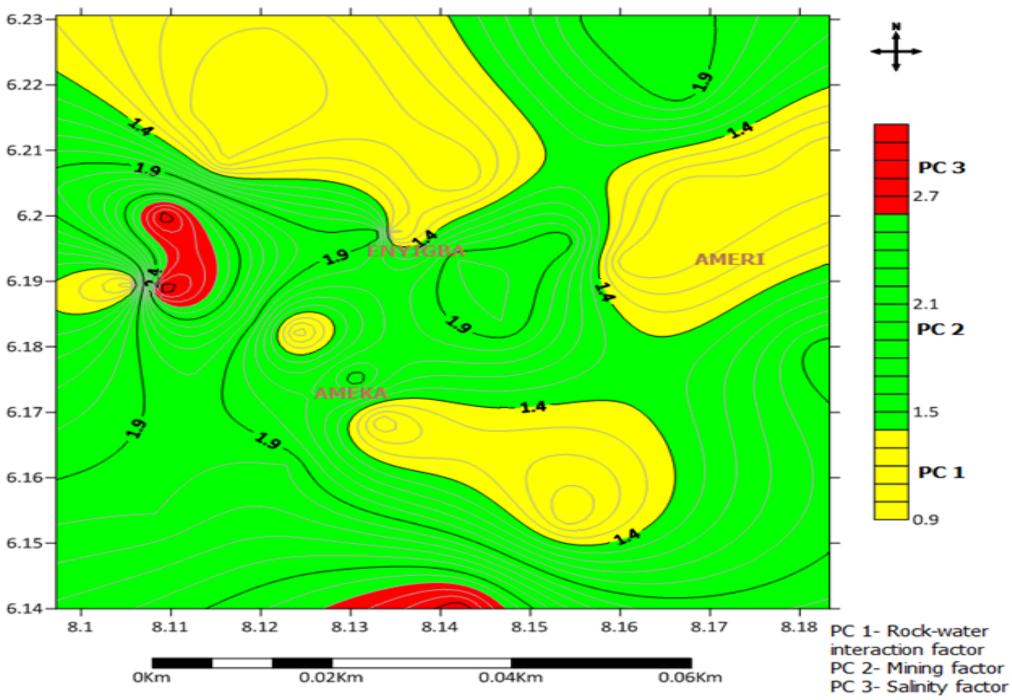

Figure 2: Spatial distribution of the major controlling processes

3.3.5 Correlation Analysis

Spearman's rho correlation analysis was performed between all variables for both sites. The level of significance ($P \leq 0.05$ and $P \leq 0.01$) of multi-element correlation for soil samples was determined, and the results are given in Tables 2 and 3. The tables showed a linear relationship between pairs of variables using numbers from -1 to +1. Positive numbers are perfect correlation while negative numbers are perfect negative correlation. However, 1 is perfectly correlated with itself. Intermetallic correlations in soil were Pb-Zn, Cu-Pb and Zn-Cd. The association of Pb with Zn suggests that the mineral deposition is geogenic. Lack of association of As, Cd with Zn and their significant correlation suggests anthropogenic origin presumably from mining activities, indiscriminate waste dumps and salinity.

Table 2: Correlation Matrix Between The Selected Heavy Metals

	Zn	*Cu*	*Pb*	*Cd*	*As*
Zn	1				
Cu	0.150796	1			
Pb	0.554821	0.495164	1		
Cd	0.021487	0.36114	0.36331	1	
As	0.281377	0.885363	0.548635	0.483087	1

Table 3: Correlation Matrix Between The Sampling Points

	S1	*S2*	*S3*	*S4*	*S5*	*S6*	*S7*	*S8*	*S9*	*S10*
S1	1									
S2	-0.06973	1								
S3	0.929614	0.086855	1							
S4	0.170133	0.964105	0.342747	1						
S5	0.224324	0.231408	-0.06895	0.176031	1					
S6	-0.11544	0.993845	0.061504	0.948871	0.174041	1				
S7	0.454439	0.520416	0.305981	0.536989	0.830477	0.491751	1			
S8	0.521385	0.81147	0.597287	0.912008	0.378836	0.781838	0.757886	1		
S9	0.144726	0.79437	0.069041	0.759837	0.747916	0.739003	0.803149	0.781509	1	
S10	-0.0244	0.818427	-0.08937	0.732588	0.737717	0.786231	0.807393	0.715341	0.967356	1

3. Results and Discussion

Concentrations of Zn, Pb, Cu, Cd and As in soils of Enyigba from surface (0-15cm) to the sub-surface (15-30cm) are presented in Table 4. The distribution of heavy metal concentration within the study area exhibits substantial spatial and depth-dependent variability. The high metal concentrations are consistent with two main sources. First, geogenic enrichment from mineralization of the area, second, anthropogenic dispersion from mining, ore processing and waste dumping concentration of zinc varies at different locations and at different depths of the soil.

Table 4 Concentrations (in mg/kg) of chemical constituents in natural soil samples at different locations and depths

Sample ID	Parameter	Latitude (N)	Longitude (E)	0-15 cm	15-30 cm
S1	Zn	6.1922	8.1107	25.57	18.38
	Cu			120.23	58.75
	Pb			56.27	29.17
	Cd			6.24	4.54
	As			24.37	11.24
S2	Zn	6.1789	8.1408	137.49	110.29
	Cu			23.04	12.34
	Pb			56.62	41.25
	Cd			0.17	2.07
	As			1.07	0.37
S3	Zn	6.168	8.1178	70.12	75.03
	Cu			208.03	210.84
	Pb			56.08	68.17
	Cd			4.57	6.03
	As			37.35	41.22
S4	Zn	6.2234	8.1224	6.56	12.24
	Cu			0.58	5.8
	Pb			2.5	4.51
	Cd			0.71	0.52
	As			0.91	0.23
S5	Zn	6.2056	8.1432	4.02	8.51
	Cu			3.21	5.35
	Pb			21	27.35
	Cd			1.67	3.2
	As			1.07	2.34
S6	Zn	6.1792	8.1823	115.12	132.37
	Cu			25.26	18.47
	Pb			36.01	42.13
	Cd			6.01	5.12
	As			22.01	12.03
S7	Zn	6.1401	8.1423	25.19	22.03
	Cu			17.52	19.23
	Pb			32.01	37.25
	Cd			2.2	3.01
	As			13.2	12.37
S8	Zn	6.1619	8.1231	50.37	44.34
	Cu			34.43	35.56
	Pb			37.01	32.03
	Cd			1.7	0.34
	As			7.24	6.01
S9	Zn	6.2044	8.0972	15.17	17.51
	Cu			6.45	8.42
	Pb			17.13	21.11
	Cd			4.24	6.34
	As			2.03	2.01
	Zn			56.01	57.31

S10	Cu	6.1931	8.1605	2.01	3.02
	Pb			58.02	58.01
	Cd			3.41	3.45
	As			3.01	2.02

Table 5: Post Hoc Test For Zinc

		Subset		
Depth	N	1	2	3
15-30	30	49.7860		
0-15	30		50.5467	
30-45	30			56.0373
Sig.		1.000	1.000	1.000

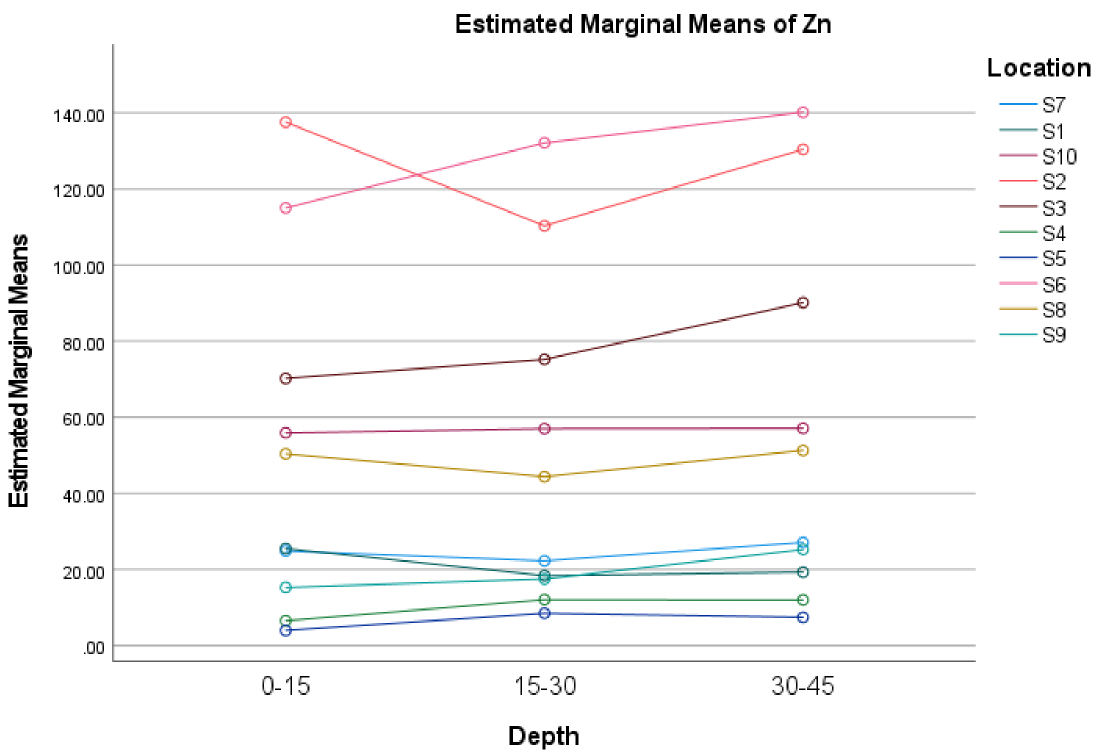

Figure 3: Estimated Marginal Means For Zinc

Table 6: Post Hoc Test For Copper

Depth	N	Subset 1	2	3
30-45	30	36.1443		
15-30	30		37.6880	
0-15	30			44.0727
Sig.		1.000	1.000	1.000

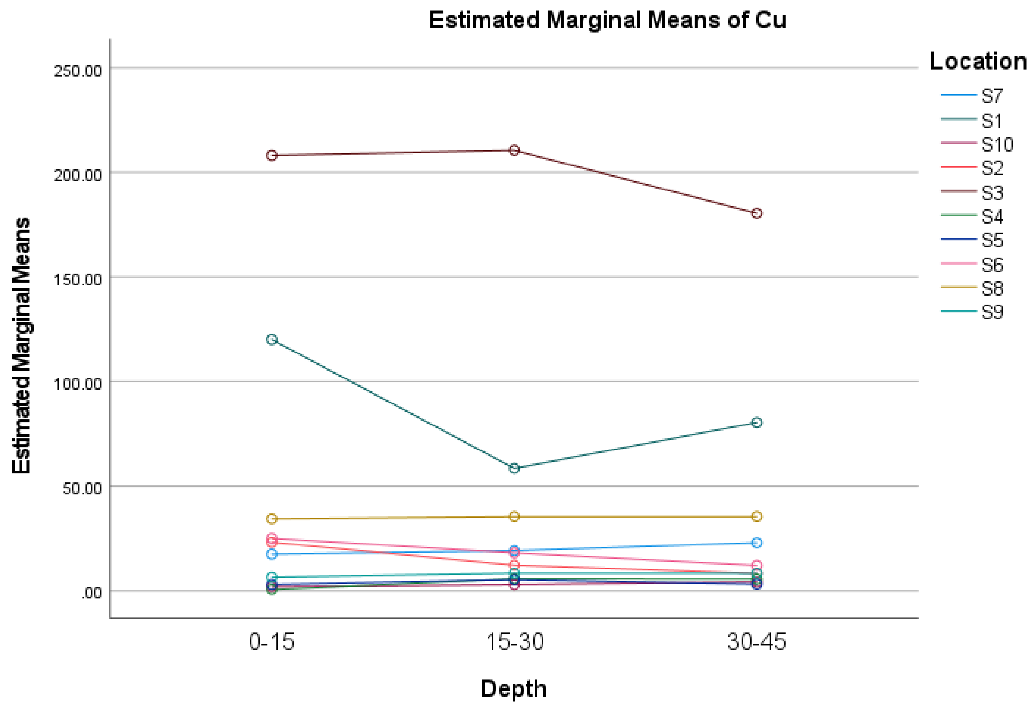

Figure 4: Estimated Marginal Means For Copper

Table 7: Post Hoc Test For Lead

Depth	N	Subset 1	2	3
15-30	30	36.1180		
0-15	30		37.3007	
30-45	30			38.7353
Sig.		1.000	1.000	1.000

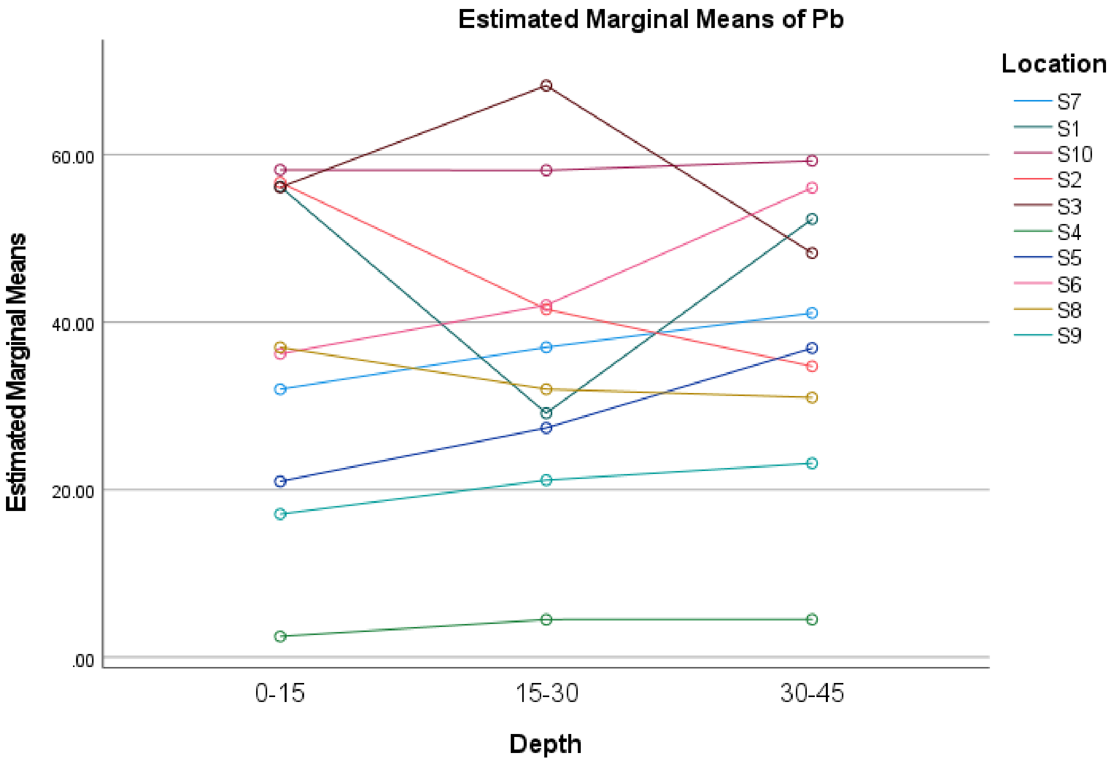

Figure 5: Estimated Marginal Means For Lead

Table 8: Post Hoc Test For Cadmium

Depth	N	Subset 1	2	3
15-30	30	49.7860		
0-15	30		50.5467	
30-45	30			56.0373
Sig.		1.000	1.000	1.000

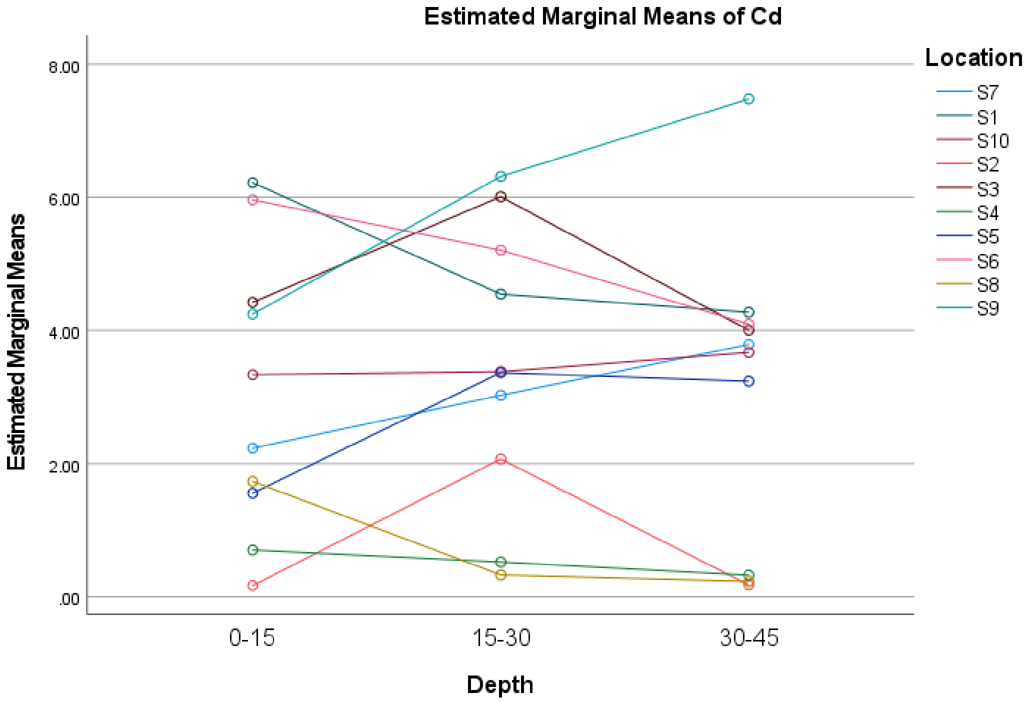

Figure 6: Estimated Marginal Means For Cadmium

Table 9: Post Hoc Test For Arsenic

		Subset		
Depth	N	1	2	3
30-45	30	8.077610		
15-30	30		8.996333	
0-15	30			11.239333
Sig.		1.000	1.000	1.000

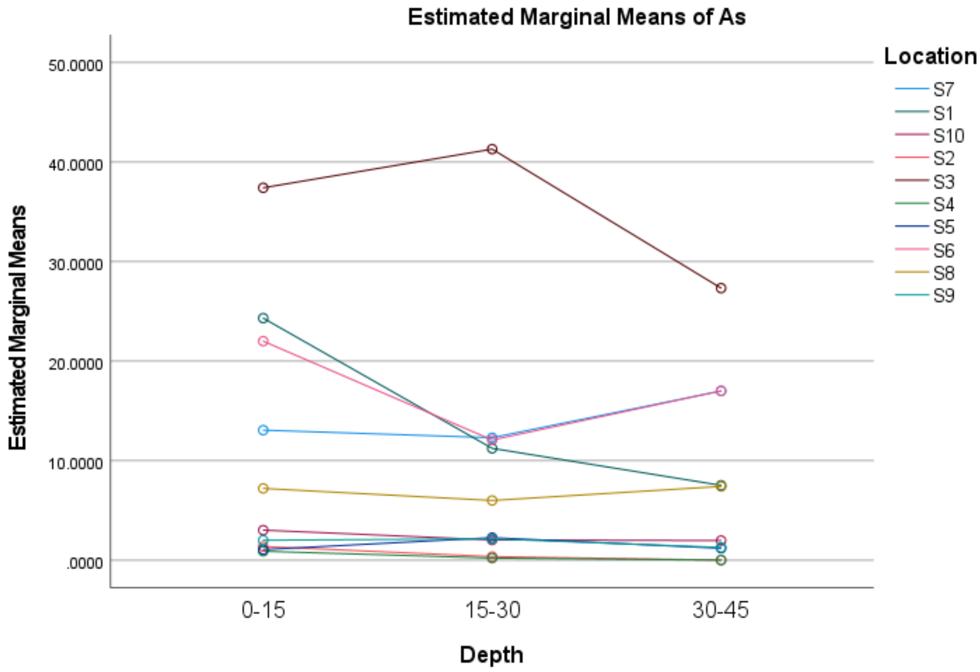

Figure 7: Estimated Marginal Means For Arsenic

Post hoc test reveals the degree of concentration, the estimated marginal means for zinc across different locations and depth depict how and in what manner the mineral increases and also decreases at different locations with depth (Table 5 and Figure 3). Copper concentrations vary at different locations and depths with a significant difference of less than 0.05 (Table 6 and Figure 4). Lead as a dependent variable varies at different layers within the soil and distribution of its concentrations across different locations (Table 7 and Figure 5).

Cadmium concentration varies at different locations and depths within the study area and shows spatial distribution across different locations (Table 8 and Figure 6). Arsenic shows estimated marginal means across different locations and depths, and varies at different layers of the soil (Table 9 and Figure 7). The assessment was carried out using the contamination factor and the degree of contamination. The modified degree of contamination shows that the study area is extremely contaminated, apart from sample location 4, which is low; all other locations within the study area are moderate to ultra-high (Figure 8)

Table 10: Contamination Factor And Modified Degree Of Contamination

ID	Cfi_Zn	Cfi_Cu	Cfi_Pb	Cfi_Cd	Cfi_As	mCd
S1	0.297	3.461	2.702	51.054	9.587	13.4201
S2	1.778	0.582	2.594	8.231	0.322	2.70124
S3	1.105	7.986	3.38	49.694	23.536	17.1399
S4	0.145	0.160	0.227	5.239	0.256	1.2050

S5	0.094	0.157	1.678	25.782	1.027	5.7476
S6	1.821	0.748	2.634	51.497	11.351	13.6099
S7	0.350	0.801	2.168	30.374	9.462	8.6312
S8	0.686	1.41	1.963	7.755	4.6	3.2828
S9	0.273	0.309	1.201	61.633	1.1356	12.9102
S10	0.801	0.124	3.438	35.952	1.564	8.3761

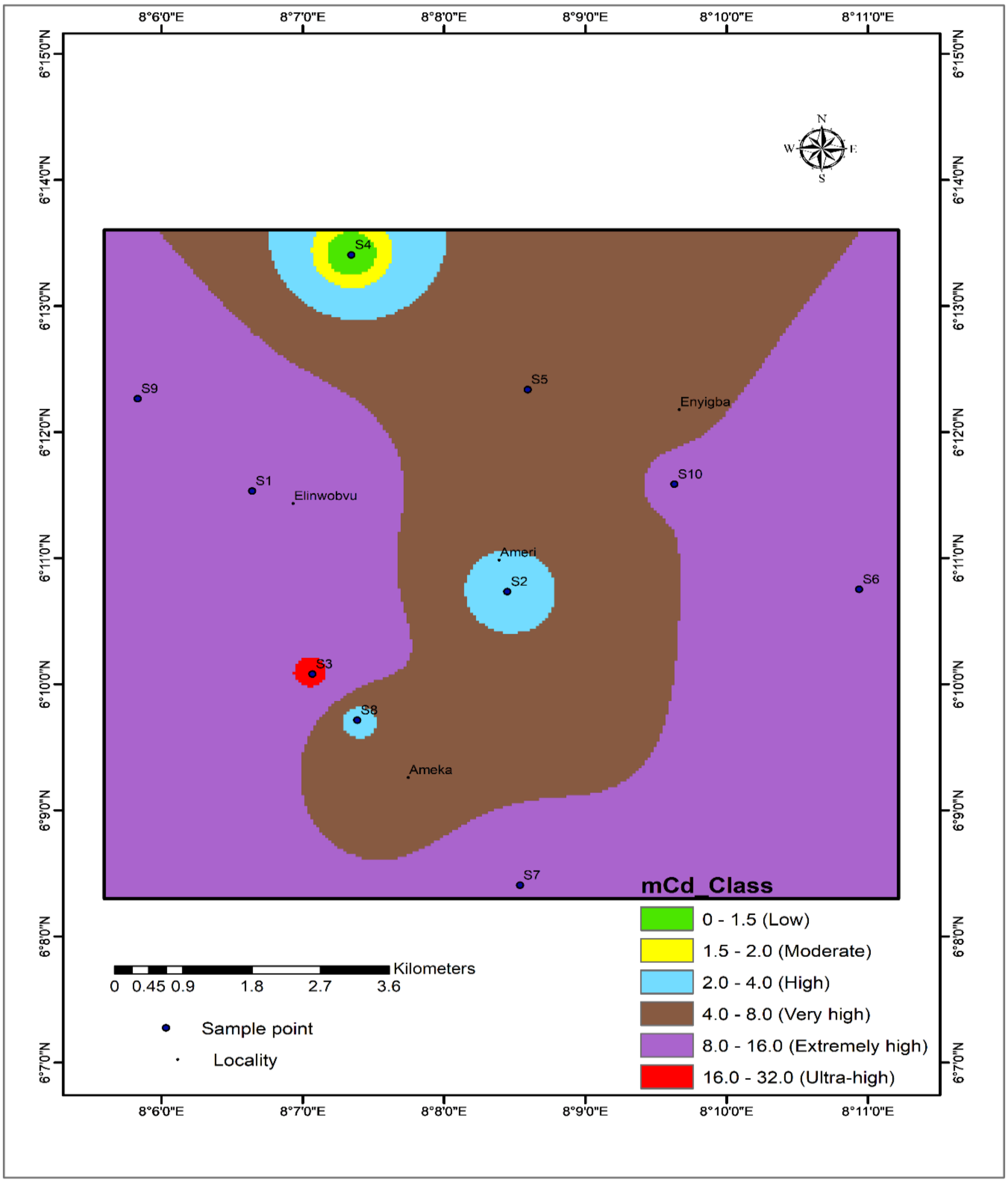

Figure 8: Modified Degree of Contamination With Its Classification

Table 11: Heavy Metal Contamination In Soils And Classes of Contamination For Geo-Accumulation Index (Igeo)

ID	**Igeo_Zn**	**Igeo_Cu**	**Igeo_Pb**	**IgeoCd**	**Igeo_As**	
S1	-2.33782	1.206199	0.848835	5.08900	2.676067	
S2	0.243982	-1.36587	0.79039	2.45616	-2.21883	
S3	-0.44156	2.412414	1.172061	5.05003	3.971808	
S4	-3.37435	-3.23002	-2.72507	1.80408	-2.55325	
S5	-3.99774	-3.25429	0.161814	4.10335	-0.54699	
S6	0.279348	-1.00462	0.81225	5.10144	2.919799	
S7	-2.09934	-0.90473	0.531559	4.33981	2.657217	
S8	-1.12807	-0.08927	0.387766	2.37018	1.616671	
S9	-2.46045	-2.27835	-0.32075	5.36066	-0.40156	
S10	-0.90469	-3.5919	1.196541	4.58305	0.060688	
Igeo < 0	**0-1**	**1-2**	**2-3**	**3-4**	**4-5**	**>5**
Unpolluted	unpolluted to moderate	moderately polluted	moderately polluted to heavily polluted	heavily polluted	Heavily polluted to extremely polluted	extremely polluted

The geoaccumulation index of cadmium in location S1 indicates that it was extremely polluted. Lead, copper, zinc and arsenic were moderately to heavily polluted. In location S2, copper, arsenic and cadmium were moderately polluted to heavily polluted. In location S3, the geo accumulation index of lead, copper, arsenic and cadmium was moderately polluted to extremely heavily polluted. In location S4, cadmium, arsenic, lead, copper and zinc were moderately to heavily polluted. In location S5, the cadmium was heavily polluted to an extreme; copper and zinc were also heavily polluted. In location S6, the geo accumulation index of cadmium was extremely polluted, and arsenic was moderately to heavily polluted. In location S7, zinc, arsenic and cadmium were moderately polluted to extremely polluted. In sample 8, zinc, arsenic, and cadmium were all moderately polluted. In sample 9, copper, zinc, and cadmium were moderately to heavily polluted. Sample 10, the geo accumulation index of lead, copper and cadmium was moderately to heavily polluted.

4.1 Conclusion

Assessment of pollution in soils by heavy metals in Enyigba, southeastern Nigeria, was evaluated using contamination factor, modified degree of contamination and geoaccunulation index. It was discovered that all the selected heavy metals (Pb, Zn, Cu, Cd and As) analyzed exceeded the standard limit and were highly contaminated. The contamination factor for all the heavy metals was moderate to high. It was also revealed that the geoaccumulation index of the heavy metals was moderately to strongly contaminated. With regard to the overall contamination of the soil in the study area, the modified degree of contamination, which determines an average value of individual contamination factor values of the studied metals, is proposed. Based on this approach, the modified degree of contamination revealed in sample 4 shows a low degree of contamination; samples 2 and 8 show a moderate degree of contamination. Samples 5, 7, and 10 reveal a high degree of contamination, while samples 1, 6 and 9 were very high, and sample 3 was extremely high. On the other hand, the geometric mean of the CF values suggests deterioration of site quality. Hence, a call for interventions to ameliorate pollution at the site is proposed. There should be a restriction on cultivating edible crops and livestock grazing in the studied area.

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