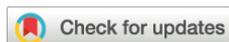


RESEARCH ARTICLE

Pollution indices of heavy metal contaminants in typical agrarian soil samples in Ihiala, South-East Nigeria

Onyenmechi Johnson Afonne^{1*} Jane Ugochi Chukwuka¹ Emeka Chinedu Ifediba¹ Ejeatuluchukwu Obi¹¹ Toxicology Unit, Department of Pharmacology, Faculty of Medicine, Nnamdi Azikiwe University, Nnewi Campus, Nnewi, Nigeria

Correspondence to: Onyenmechi Johnson Afonne, Toxicology Unit, Department of Pharmacology, Faculty of Medicine, Nnamdi Azikiwe University, Nnewi Campus, PMB 5001, Nnewi, Anambra State, Nigeria; E-mail: oj.afonne@unizik.edu.ng

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Abstract: The contamination of soil with toxic metals poses serious threats to the survival of living organisms including humans. We determined the contamination levels of cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb) and nickel (Ni) in soil samples from a typical agrarian soil in Nigeria, using various single and complex geochemical indices along with principal component analysis (PCA) for source determination. Ten soil samples (S1-S10) were collected from depths of 20 cm, with a clean shovel and brush from farmlands in Ihiala, South-East Nigeria. Three single pollution indices: geoaccumulation index (I_{geo}), pollution index (PI) and ecological risk index (E_r), as well as four complex indices: pollution load index (PLI), Nemerow pollution index ($PI_{Nemerow}$), average single pollution index (PI_{ave}) and Potential ecological risk (RI) were used for the geochemical analysis. The mean soil levels of Cd (1.94ppm) and Pb (60.83ppm) exceeded their corresponding world averages. The results of the single pollution indices of the soil samples revealed heavy Cd, moderate Pb and low Ni, Cr and Cu contaminations, while the PI_{avg} , $PI_{Nemerow}$ and RI graded the soil samples as moderately to seriously polluted. The correlation analysis revealed that the general contamination was mostly contributed by Cd and partly by Cr. The findings showed that Cd and Pb were the main heavy metal soil contaminants in the area. The levels of toxic metals found in the soils could pose health and ecological risks. The probable sources of these metals include pesticides use and poor waste disposal systems.

Keywords: farmland, pollutant, pollution index, soil contamination, toxic metal

1 Introduction

Soil has long been known to be a source of sustenance for food crops. Thus, soil contamination can lead to reduction in the use of land for agricultural purposes and consequently may lead to food insecurity [1]. One of the challenging problems of pollution is contamination of soil by heavy metals, considering the wide distribution and transferability of these metals to plants [2–4]. A natural source of heavy metals in soils is bedrock, while some quantities are introduced by industrial activities [5–7] and agricultural activities such as the use of some organic and inorganic fertilizers, organic manures, and heavy metal-containing pesticides and herbicides [8]. Another source of contamination of soil-crop systems is poor waste disposal systems, as reports have shown that leachates from municipal solid waste landfills do contain high levels of metals and metalloids [9].

The International Resource Panel of Working Group on the Global Metal Flows, which is a panel of the United Nations Environment Programme posited that accumulation of toxic metals in food crops and their consequent transfer to the food chain is a major environmental issue worldwide [10]. Other factors that may exacerbate the soil heavy metal contamination profile include temperature, moisture content, pH and organic matter [11, 12]. High contamination of soils by heavy metals has been reported in both industrialized and agrarian areas, indicating the diverse and diffusive tendencies possessed by metals [12]. There have been reported incidences of heavy metal contamination of agricultural soils in Nigeria [13, 14] and other countries [15–17]. Contamination of soil surrounding natural water bodies by toxic metals has been found to correlate positively with heavy metal pollution of the aquatic bodies themselves [18, 19].

Ihiala is an agrarian suburb in the South-East region of Nigeria, and our recent findings showed that natural water sources in the area were polluted by heavy metals, especially lead [20]. There is need, therefore, to determine the heavy metal pollution of soils from farmlands surrounding water sources, with a view to predicting the plausible quantity of toxic metals that could be transferred to farm produces. Soil pollution indices are useful tools for toxicological evaluation of the extent of contamination, and thus are useful in the assessment of soil quality

and prediction of future ecosystem sustainability, particularly farmlands [21]. They help to determine whether the heavy metals accumulation was due to natural processes or as a result of anthropogenic activities. Heavy metal contamination of soils in the South-East Nigeria has been found not to undermine the use of lands in the region for agricultural activities [22]. This may, however, not be the case with most other regions having poor waste disposal systems. The aim of this study was to determine the contamination levels of some heavy metals in samples of soils close to natural water sources from a typical agrarian area in Nigeria, using various indices based on single and complex geochemical indices along with principal component analysis (PCA) for source determination.

2 Material and methods

2.1 Study area

The study was carried out in Ihiala and some surrounding communities. Ihiala is a semi-urban area located in the Southern part of Anambra State, South-East Nigeria. The area lies approximately between latitudes 5.83°N to 5.85°N and longitudes 6.82°E to 6.85°E. It measures a distance of about 40 km from Onitsha, the commercial hub of the State, and lies along the Onitsha-Owerri highway. It is bounded by the following communities: to the North, Okija; to the South, Uli and Egbu; to the East, Azia and Mbosi; and to the West, Uliasi River (Figure 1). Ihiala has a projected 2018 population estimate of 83, 265 persons and occupies a land mass of 310 km². The town with its surrounding neighbours are situated in undulating low lands, characterized by deep valleys and rolling hills, and endowed with natural water sources like streams, rivers and springs. The major occupations of the people of this area are farming and trading. There are no major industries within the localities.

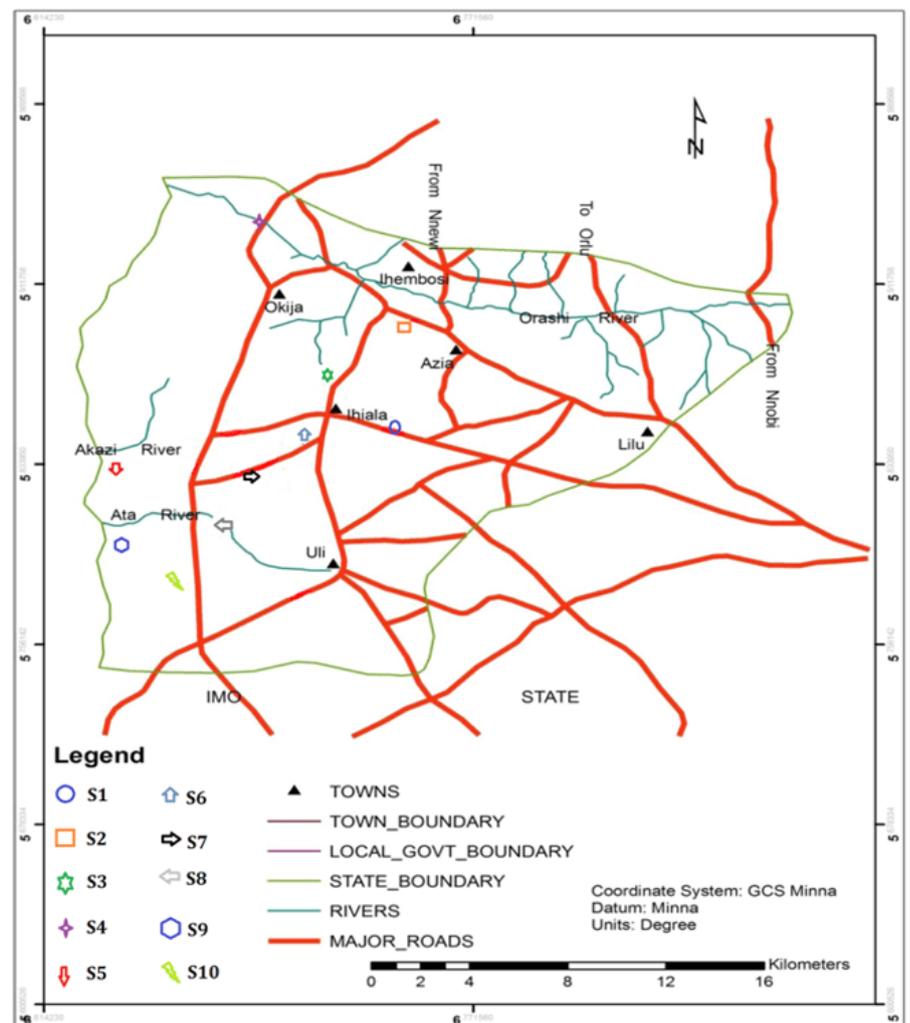


Figure 1 Map of Ihiala and neighbouring towns showing the sampling sites

2.2 Sample collection

Ten soil samples (labeled S1 to S10) were collected from a 20cm depth, with a clean polymethyl methacrylate shovel and small brush from farmlands at different villages in Ihiala. The samples were collected under the same conditions in airtight sterile plastic containers on the same day, during the farming period. These were taken to the National Veterinary Research Institute (NVRI), Vom, Nigeria, and National Chemical Research Institute (NCRI), Zaria, Nigeria, for processing and analysis. The plastic containers were sterilized by the manufacturers with ethylene oxide gas.

2.3 Sample analysis

The pH of the soil samples was measured in the ratio of 2:5 (sample: distilled water) with a pH glass electrode. For the determination of heavy metals, the samples were air dried at room temperature and sieved through a 2 mm nylon sieve to remove big coarse debris and then digested according to the Association of Official Analytical Chemistry (AOAC) method [23]. Briefly, 5 ml of HNO₃ was added to the sample already containing 1 ml of perchloric acid and 0.5 ml of conc. H₂SO₄. The mixture was then placed on a Khedjal heater and brought to boil until it was colourless under a ductless fume cabinet. The digested samples in the flask were allowed to cool and filtered, using a filter paper, into a 100 ml volumetric flask. The filtrate was made up to mark with deionized water. The solution was put into clean metal-free sample bottles in duplicates and properly labelled for the analysis of Cd, Pb, Ni, Cu and Cr using a flame atomic absorption spectrophotometer (Shimadzu AAS 6800 B model Japan), according to the specifications of the manufacturer.

2.4 Heavy metal pollution indices

Three single and four complex pollution indices were used in this study. The single pollution indices were: geoaccumulation index (I_{geo}), pollution index (PI) and single ecological risk index (E_r), while the complex pollution indices included: pollution load index (PLI), Nemerow pollution index ($PI_{Nemerow}$), average single pollution index (PI_{ave}) and Potential ecological risk (RI). These indices were calculated with five metals (Cd, Ni, Cr, Cu and Pb) to evaluate the geochemical pollution and quality of the agricultural soil samples. Due to the lack of information on the official standards for pollution levels in agricultural soils in Nigeria, calculations of pollution indices were done using reference heavy metal composition from the upper continental crust (UCC) proposed by Rudnick and Gao as geochemical background values [24]. Recently, the UCC reference values have been said to provide a more universal character [21]. These values (in mg/kg) for the elements studied are Cd = 0.09; Ni = 47.0; Cu = 28.0; Cr = 92.0 and Pb = 17.0.

2.5 Single pollution indices

2.5.1 Geoaccumulation index (I_{geo})

The I_{geo} is a quantitative measure of metal pollution that allows for the assessment of soil contamination with heavy metal based on its contents in O or A level horizons soil layers, referenced to a specified geochemical background (GB). It was first defined by Muller [25] to determine and define metal contamination in sediments, by comparing current concentrations with pre-industrial levels. It is used to calculate the enrichment of trace element concentration above background values. I_{geo} values are estimated as follows:

$$I_{geo} = \text{Log}_2\left(\frac{C_n}{1.5 \times B_n}\right)$$

where C_n is the measured concentration of heavy metals in soil and B_n is the geochemical background value of each metal. The constant 1.5 is used for the possible variations of the background data due to the lithogenic effects. Generally, the I_{geo} has been distinguished by Muller into 7 classes in the range of $5 < I_{geo} < 0$ (Table 1) [25].

2.5.2 Single pollution index (PI)

This index can be used to determine which heavy metal represents the highest threat for a soil environment [21]. The PI is computed using the equation:

$$PI = \frac{C_n}{B_n}$$

Where C_n is the measured concentration of heavy metals in soil and B_n the geochemical background value of each metal [26].

2.5.3 Ecological risk factor (E_r)

The E_r of a given contaminant was suggested by Hakanson [26], with the formula:

$$E_r = Tr \times C_f$$

where C_f is contamination factor and the Tr is toxic-response factor for the given substance. The Tr accounts for both the toxic factor requirement (the St -value) and the sensitivity requirement (given by the BPI-value), and is analogous to the C_f . The Tr values for the metals studied have been established and given as follows: Ni = 5, Cd = 30, Cr = 2, Cu = 5, and Pb = 5 [26,27]. The terminologies used to describe E_r are presented in Table 1.

Although E_r was used mainly as a diagnostic tool for the purpose of controlling water pollution, it has been used to assess ecological geochemistry [27].

2.6 Combined pollution indices

2.6.1 Pollution load index (PLI)

The PLI provides an easy way to prove the deterioration of the soil conditions as a result of the accumulation of heavy metals. It is calculated as a geometric average of PI as follows:

$$PLI = \sqrt[n]{PI_1 \times PI_2 \times PI_3 \times \dots \times PI_n}$$

where n = the number of analyzed heavy metals and PI = calculated values for the single pollution index. The index was proposed by the Irish Estuarine Research Group [28], and has been used by Varol [29] to assess water sediments. Its assessment criteria are shown in Table 1.

Table 1 Pollution Indices Classification

Parameter	Value	Environmental Risk Class	References
Single Indices			
I_{geo}	$I_{geo} \leq 0$	Practically uncontaminated	Muller [25]
	$0 < I_{geo} \leq 1$	Uncontaminated to moderately contaminated	
	$1 < I_{geo} \leq 2$	Moderately contaminated	
	$2 < I_{geo} \leq 3$	Moderately to heavily contaminated	
	$3 < I_{geo} \leq 4$	Heavily contaminated	
	$4 < I_{geo} \leq 5$	Heavily to extremely contaminated	
	$I_{geo} > 5$	Extremely contaminated	
PI	$PI < 1$	Low contamination	Hakanson [26]
	$1 \leq PI < 3$	Moderate contamination	
	$3 \leq PI < 6$	Considerable contamination	
	$PI > 6$	High contamination	
Er	$Er < 40$	Low potential ecological risk	Hakanson [26]
	$40 \leq Er < 80$	Moderate potential ecological risk	
	$80 \leq Er < 160$	Considerable potential ecological risk	
	$160 \leq Er < 320$	High potential ecological risk	
	$Er \geq 320$	Very high potential ecological risk	
Complex Indices			
PLI	$PLI < 1$	Not polluted	Tomlinson et al. [28]
	$PLI = 1$	Baseline level of pollutants	
	$PLI > 1$	Polluted	
$PI_{Nemerow}$	$P_N \leq 0.7$	Safety domain	Cheng et al. [30]
	$0.7 < P_N \leq 1$	Precaution domain	
	$1 < P_N \leq 2$	Slightly polluted domain	
	$2 < P_N \leq 3$	Moderately polluted domain	
	$P_N > 3$	Seriously polluted domain	
RI	$RI < 150$	Low contamination	Hakanson [26]
	$150 \leq RI < 300$	Moderate contamination	
	$300 \leq RI < 600$	Considerable contamination	
	$RI \geq 600$	High contamination	
	Value	Environmental Risk Class	

2.6.2 Nemerow pollution index ($PI_{Nemerow}$)

The Nemerow Pollution Index ($PI_{Nemerow}$) permits the assessment of the overall degree of pollution of the soil and makes use of all the heavy metals analyzed [27]. It is calculated thus:

$$PI_{Nemerow} = \sqrt{\frac{(\frac{1}{n} \sum_{i=1}^n PI)^2 + PI_{max}^2}{n}}$$

where PI = calculated values for the PI, PI max = maximum value for the PI of all heavy metals and n = the number of heavy metals. According to the Soil Environmental Quality Standards (GB15618-1995), five grades of soil quality have been defined based on $PI_{Nemerow}$ (Table 1) [30].

2.6.3 Average single pollution index (PI_{avg})

The PI_{avg} has been used by Bhattacharya *et al.* [31] to assess soil quality. It can be defined as the average of the single PIs and designated as follows:

$$PI_{avg} = \frac{1}{n} \sum_{i=1}^n PI$$

where n = the number of studied heavy metals, and PI = calculated PI values. The PI_{avg} values >1.0 indicate low soil quality due to high contamination [27].

2.6.4 Potential ecological risk index (RI)

Potential ecological risk index (RI) is an index that is applied in the assessment of the degree of ecological risk caused by heavy metal concentrations in the water, air, as well as the soil. Introduced by Hakanson [26], it is defined as the sum of the risk factors:

$$RI = \sum_{i=1}^n E_r^i$$

where n = number of heavy metals and E_r = ecological risk factor calculated. On the basis of the potential ecological risk, four classes of soil quality are known (Table 1).

3 Results

3.1 Heavy metal concentration

The soil pH values varied from 7.62 to 9.0 with a mean value of 8.40 indicating that soils of this region were alkaline. The summary of descriptive statistics of the assayed metals in all the soil samples and values of international standard limits is presented in Table 2. The ranges of heavy metal levels (in ppm) in the soil samples were Cd (0.55 – 2.94), Ni (1.72 – 10.86), Cu (0.87 – 1.92), Cr (3.56 – 21.85) and Pb (19.90 – 122.22). The order of soil heavy metal levels is Pb > Cr > Ni > Cd > Cu. However, the mean, first quartile (Q_1) and third quartile (Q_3) concentrations of Cd exceeded world average value while the mean and Q_3 of soil Pb levels exceed its world average (Table 2). The spatial distribution of the pH and metals in the sites (Figure 2) indicates that the changes in the distribution parameters across the sites were not consistent.

Table 2 Summary statistics of pH and heavy metals

Parameter	pH	Metal level (ppm)				
		Cd	Ni	Cu	Cr	Pb
Mean ± SD	8.40 ± 0.51	1.94 ± 0.80	5.08 ± 2.98	1.14 ± 0.30	13.21 ± 7.02	60.83 ± 41.77
Median	8.55	2.18	5.235	1.08	13.72	52.59
Min – Max	7.62 – 9.00	0.55 – 2.94	1.72 – 10.86	0.87 – 1.92	3.56 – 21.85	19.90 – 122.22
Q_1	7.99	1.45	2.82	0.97	8.14	22.74
Q_3	8.75	2.37	6.23	1.15	18.67	88.83
WA	–	0.41	29	38.9	59.5	27

Note: Q_1 = First quartile; Q_3 = Third quartile; SD = Standard deviation; WA = World average (Kabata-Pendias and Mukherjee, 2007).

3.2 Metal pollution indexing

The result of the geochemical index (Table 3) indicated that the soils were heavily contaminated by Cd (2.03 – 4.44) but moderately contaminated by Pb (-0.36 – 2.26). The contamination by Ni, Cr and Cu were however classified as low by the indices. The result of the PI (Table 4) also showed that samples were highly contaminated with Cd (6.11 – 32.67) while the pollution by Pb ranged from moderately to highly contaminated (1.17 – 7.19). The result of the single ecological risk (Table 5) showed that Cd contamination poses a high to very high potential ecological risk (183.33-980.00) while none of the meals poses potential ecological risk. Results

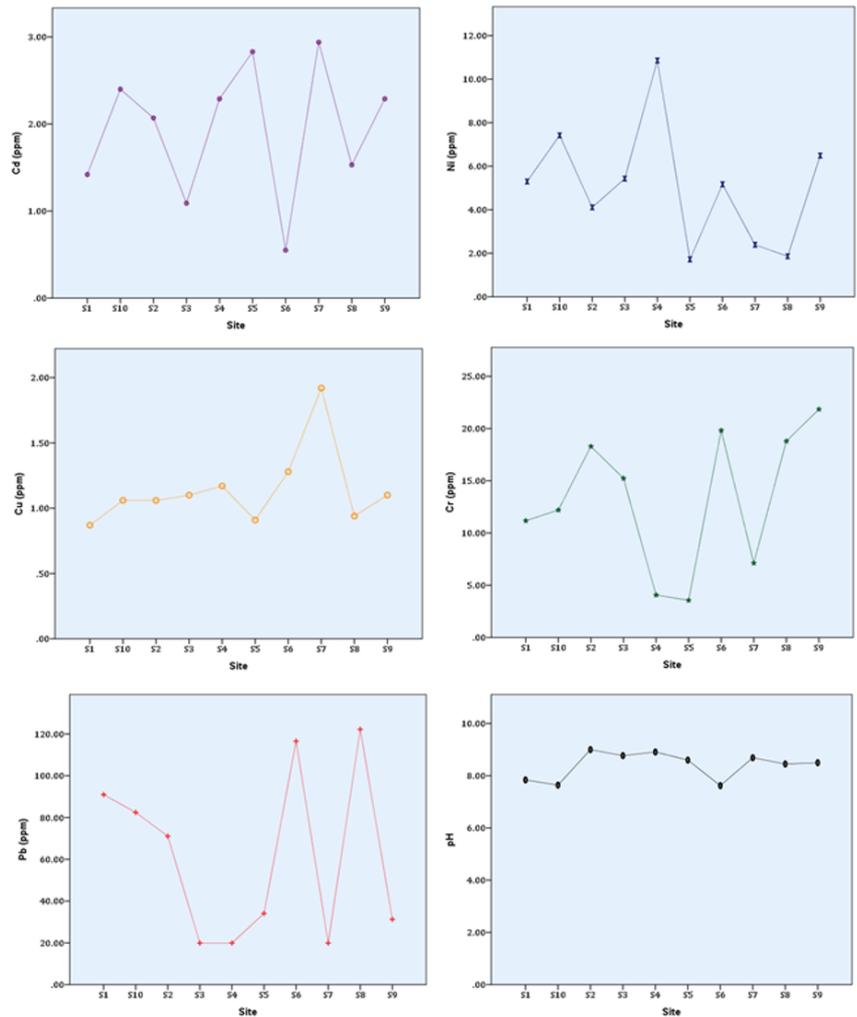


Figure 2 Spatial distribution of metals and pH

of complex indices (Table 6) indicated that PLI classified all the soil samples as unpolluted while the PI_{avg} grades all the samples as of low quality. The domain of the $PI_{Nemerow}$ (2.81 – 15.05) and RI (189.602 – 1017.92) showed that HM soil contamination ranged from moderately polluted to seriously polluted.

Table 3 Geoaccumulation index

Parameter	Metal				
	Cd	Ni	Cu	Cr	Pb
Mean ± S.D.	3.85 ± 2.56	-3.80 ± 4.47	-5.20 ± 7.12	-3.38 ± 4.30	1.25 ± 0.71
Median	4.01	-3.75	-5.28	-3.33	1.04
Min-Max	2.03 – 4.44	-5.36 – 2.70	-5.59 – -4.45	-5.28 – -2.66	-0.36 – 2.26
Q ₁	3.42	-4.64	-5.44	-4.08	-0.17
Q ₃	4.14	-3.50	-5.19	-2.89	1.80

Note: Min = minimum value; Max = maximum value; Q₁ = first quartile; Q₃ = third quartile; SD = Standard deviation.

Table 4 Single pollution index

Parameter	Metal				
	Cd	Ni	Cu	Cr	Pb
Mean ± SD	21.57 ± 8.83	0.11 ± 0.06	0.04 ± 0.01	0.14 ± 0.08	3.58 ± 2.46
Median	24.22	0.11	0.04	0.15	3.09
Min-Max	6.11 – 32.67	0.04 – 0.23	0.03 – 0.07	0.04 – 0.24	1.17 – 7.19
Q ₁	16.08	0.06	0.03	0.09	1.34
Q ₃	26.36	0.13	0.04	0.20	5.23

Note: Min = minimum value; Max = maximum value; Q₁ = first quartile; Q₃ = third quartile; SD = Standard deviation.

Table 5 Single ecological risk index

Parameter	Metal				
	Cd	Ni	Cu	Cr	Pb
Mean ± SD	647.00 ± 265.02	0.54 ± 0.32	0.20 ± 0.05	0.29 ± 0.15	17.89 ± 12.28
Median	726.67	0.56	0.19	0.30	15.47
Min-Max	183.33 – 980.00	0.18 – 1.16	0.16 – 0.34	0.08 – 0.48	5.85 – 35.95
Q ₁	482.50	0.30	0.17	0.18	6.69
Q ₃	790.83	0.66	0.21	0.41	26.13

Note: Min = minimum value; Max = maximum value; Q₁ = first quartile; Q₃ = third quartile; SD = Standard deviation.

Table 6 Complex indices

Parameter	PLI	PI _{avg}	PI _{Nemerow}	RI
Mean ± SD	0.55 ± 0.26	5.09 ± 2.29	9.91 ± 4.08	665.92 ± 277.83
Median	0.54	5.52	11.11	743.18
Range	0.20 – 0.98	1.48 – 8.08	2.81 – 15.05	189.602 – 1017.92
Q ₁	0.33	3.52	7.36	489.84
Q ₃	0.69	6.39	12.13	818.23

Note: PLI = Pollution Load Index; PI_{avg} = average Pollution Index; PI_{Nemerow} = Nemerow Pollution Index; RI = Potential Ecological Risk; Q₁ = first quartile; Q₃ = third quartile.

3.3 Correlation/Factor analysis of metals and indices

The results of the Pearson correlation analysis (Table 7) showed that none of the metals were correlated with each other. However, Cd was found to be significantly correlated with PI_{avg} (r = 0.965, p < 0.01), PI_{Nemerow} (r = 1.00, p < 0.01) and RI (r = 0.999, p < 0.01). Cr was also significantly correlated (r = 0.690, p < 0.01) with PLI; PI_{avg} with PI_{Nemerow} (r = 0.968, p < 0.01) and RI (r = 0.974, p < 0.01). From the principal component analysis (PCA) (Table 8), two components emerged with the Eigen value all > 1, which explained more than 68 % of cumulative variance. The PC-1 is loaded with Cd, Cu, Pb and Cr with 45.71 % of the total variance, while the PC-2 loads only Ni with 22.32 % of the variance.

The rotated component matrix (Figure 3) showed that component one loads Cd, Cu, Cr and Pb, while the second component correlated with Ni.

Table 7 Correlation matrix between metals and complex indices

Cd	Ni	Cu	Cr	Pb	PLI	PI _{avg}	PIN	RI	
Cd	1.00								
Ni	-0.07	1.00							
Cu	0.28	-0.10	1.00						
Cr	-0.58	-0.10	-0.18	1.00					
Pb	-0.57	-0.23	-0.36	0.50	1.00				
PLI	-0.26	0.28	-0.19	0.690*	0.58	1.00			
PI _{avg}	0.965**	-0.15	0.21	-0.50	-0.33	-0.11	1.00		
PIN	1.000**	-0.08	0.29	-0.58	-0.56	-0.25	0.968**	1.00	
RI	0.999**	-0.09	0.27	-0.57	-0.54	-0.24	0.974**	0.999**	1.00

Note: PLI = Pollution load index; PI_{avg} = Average pollution index; PIN = Nemerow pollution index; RI = Potential ecological risk.

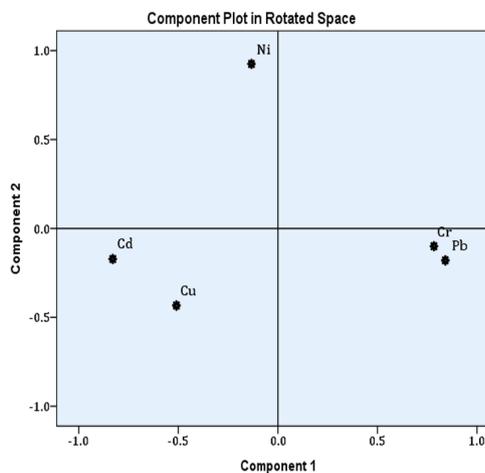


Figure 3 Component plot of metals and complex indices in rotated space

Table 8 Principal component analysis

Metal	PC-1	PC-2
Cd	-0.83	-0.16
Ni	-0.12	0.93
Cu	-0.51	-0.43
Cr	0.78	-0.11
Pb	0.84	-0.19
Eigine value	2.29	1.12
% variance	45.71	22.32
Cumulative %	45.71	68.03

4 Discussion

The presence of heavy metals in agricultural soils is of critical importance due to their accumulation in food chains and adverse effects on the entire ecosystem. The contamination of soil by heavy metals therefore, presents an important risk as regards livestock exposure to heavy metal and absorption by edible plants. In this study, of the five metals (Pb, Cu, Cd, Ni and Cr) determined, the mean levels of soil Cd and Pb were found to exceed world averages for agricultural soils [33]. Pollution indexing modeling and correlation analysis further implicated these metals as major culprits in the soil heavy metal contamination of the region. Cd, a toxic trace element is an environmental contaminant, both through natural occurrence and from industrial and agricultural sources [10]. Unfortunately, increases in soil cadmium content will result in an increase in the uptake of cadmium by plants, an important pathway of human exposure to the metal [34]. However, since Cd uptake by plants from soil is greater at low soil pH, high soil pH observed within the study area may have mitigating effect [11]. Lead is also a ubiquitous element, found naturally in the earth's crust and is used in various industrial applications and can thereby be introduced into the soil [10]. In addition to industry, it has applications in fertilizers and pesticide used for agriculture purposes, and in improving the octane rating of gasoline in vehicular traffic systems [35]. Exposure to these metals produces a wide variety of effects ranging from renal, nervous and respiratory problems, disturbances in calcium metabolism, increased in blood pressure to fertility problems [10,36]. The causes of high levels of these metals could be poor waste disposal, the use of fertilizers or other agricultural product enhancers and activities of sparsely situated fuel and service stations [22]. The variation of metal levels observed from the sites could have occurred both as a result of natural processes and of different anthropogenic sources [10]. Health and ecological risks posed by these three metals in soils have also been estimated elsewhere [16, 18], and locally [3]. The contamination of the soil by these metals should really elicit concern given the observation that they may be readily absorbed by crops [37]. Principal component analysis (PCA) was usually performed to establish possible factors that contribute towards the metal concentrations and source apportionment [38].

The rotated component matrix showed that the first component was loaded with Cd, Cu, Pb and Cr, an indication of similar pollution sources of these metals, which may probably come from road transportation, farming and wastewater [39]. The second component correlated with Ni, showing different sources of this metal in the region. These observations in this study make Cd and Pb candidates of remediation in the area given the pressure on agricultural lands for adequate food production.

5 Conclusion

Soil samples from the agrarian town of Ihiala are contaminated with heavy metals, precisely Cd and Pb. The possible sources of these contaminants include, but not limited to, anthropogenic activities, especially use of pesticides and improper waste disposal practices. We recommend that adequate measures should be put in place, by relevant authorities, to regulate and checkmate human activities in agrarian regions in order to protect farmlands from deterioration and contamination, with the attendant toxicity to man.

Conflict of interest and funding

The authors declare no conflict of interest or any financial and personal interests that could have influenced the outcome of this study.

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