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Ecological Risk Assessment and Pollution Models of Trace Metal Concentrations in Road Dust in Parts of Enugu, Southeastern Nigeria

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ABSTRACT: Trace metal levels in road deposited sediments (RDS) from paved roads in Enugu metropolis, southeastern Nigeria were evaluated. The study aimed at determining the level of heavy metals in the RDS, with the objectives of identifying the sources of these pollutants in the environment. Thirty-two (32) RDS samples from 10 selected major roads in Enugu were collected for this study. The concentration of Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb was analyzed using a flame atomic absorption spectrophotometer. The levels of physicochemical properties; pH, electrical conductivity (EC), total organic matter (TOM), and total organic carbon (TOC) was also established using standard procedures. The metal concentrations (mg/kg) were in the range; Cr (0.67 - 3.67), Mn (2.33 - 7.67), Fe (408.33 - 512.33), Co (3.00 - 61.75), Ni (39.67 - 193.67), Cu (11.50 - 90.00), Zn (1.00 - 31.67), Cd (0.00 - 1.33), and Pb (0.00 - 46.67). The pH, EC, TOC and TOM of the RDS samples range between 6.75-8.39, 556 - 578.67 µS/cm, 0.43 % - 3.49 % and 1.27 % - 10.35 % respectively. The relationships between the parameters were ascertained with correlation analysis. The likely source of these metals was recognized with principal component analysis and hierarchical cluster analysis, deployed to identify the potential source of these metals in the RDS. The results suggested that Mn concentrations did not exceed the crustal level. However, elevated Pb and Cd levels were recorded in the RDS samples. The models showed good consistency, indicating average to considerable levels of contamination of most parts of the studied major roads. PCA result revealed that metals in the RDS originated from three main sources which include the crustal, vehicular, and industrial sources.

INTRODUCTION

Fine solid particles are usually deposited on surfaces such as paved surfaces of the urban environment [1 –3]. These deposited particles on surfaces are also called road deposited sediments (RDS). Road deposited sediments are usually a complex mixture of fine solid particles and pollutants due to a variety of industrial and urban events [1 – 5]. The origin of these pollutants may be due to activities of human beings in nature like wearing off of the vehicle tyre and body, brake-lining materials, and vehicle exhaust emissions. This is in addition to materials used in building and road construction,

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vegetable waste from plants as well as atmospheric depositions [1 – 5].

The potential health risks and high levels of heavy metal contamination recorded in many cities of the world have generated a lot of interest regarding heavy metal pollutants in road deposited sediments. Roadways and automobiles are presently considered the largest sources of toxic metals in urban surroundings [5 – 6]. Among the most common toxic metals released from travels and other anthropogenic activities on roadsides including wearing off of vehicle tyres, brake linings/pads, and exhaust emissions, are zinc, copper, and lead [6]. These metals account for about 90 percent of the total metal pollutants in road deposits and runoffs [7]. The accumulation of road dust on paved surfaces of urban surroundings can make enormous concentrations of heavy metals to be available in nearby streams and river bodies [8]. One of the notable characteristics of these trace metallic pollutants that make them different from other toxic contaminants is their non-biodegradability in nature [9]. The possibility of bioaccumulation and bio-magnifications is another problem associated with them as this exposes organisms within the surrounding environments to have higher concentrations of these contaminants than what is usually present in the natural environment [9-10]. Accumulation of these toxic metals in the tissues of humans and other living organisms are usually through the uptake of food from the immediate polluted environment [10].

An investigation into road deposited sediments is of great significance in this study for obvious reasons. Firstly, the dust in RDS is freely inhaled by humans and other living organisms in the vicinity of the polluted roads [10]. As the RDS becomes more polluted with toxic metals, the more people within the vicinity are exposed to the dangers inherent in having contact with the heavy metals. Secondly, RDS is one of the main pathways through which metal contaminants get into soils, surface, and underground water sources which eventually reach the living tissues of plants, animals as well as man. A lot of research efforts have been made by different researchers to assess metallic concentrations of RDS and their associated distributions in various cities worldwide, e.g., Oslo and Madrid [11], Hong Kong [12], Birmingham and Coventry [13], Naples [14], Istanbul [15], Southeast China [16], Brisbane [17] and Gela [18]. In Nigeria, several research works have also been carried out in cities like Ibadan, Suleja, Benin City, Makurdi, Owerri, Mubi, and Enugu among others [7,19 -25]. Most of these previous studies in major cities of Nigeria have been limited to metallic ion concentrations in soils or road deposited sediments. Also, the majority of these studies focused mostly on Fe, Zn, Pb, Cu, and Cd, while a few focused on Ni, Mn, Cr, and Co due to their negative effects on the environment [24].

The present study, therefore, investigated contamination levels of RDS in major roadways of Enugu Metropolis using various ecological risk assessment and pollution models. The main aim of the study was to assess the level of heavy metals and their sources in the RDS of major roads in Enugu Metropolis. The need for this study mainly because RDS is a major pathway through which metallic pollutants get into soils, surface, and underground water sources. Therefore, the concentrations of nine (9) metallic pollutants (Cd, Cr, Cu, Ni, Pb, Mn, Fe, Co, and Zn) were assessed in the RDS samples from the study area. The level of heavy metal contamination was evaluated using indices of pollution, which include the index of geo-accumulation (Igeo), enrichment factor (EF), contamination factor (and degree of contamination), and potential ecological risk index. Multivariate statistical techniques which include the principal component analysis (PCA) and hierarchical cluster analysis (HCA) were adopted to identify the source(s) of the heavy metals, while the relationships among the various pollutants concerning their sources and fate were established with the aid of the Pearson correlation coefficient analysis.

MATERIALS AND METHODS

Study area

The study was carried out in the Enugu metropolis of Enugu State, Southeastern Nigeria (Figure 1). Enugu is the capital city of Enugu State which is situated in the southeastern part of Nigeria. Enugu metropolis comprises three local government areas including Enugu East, Enugu North, and Enugu South. The population of Enugu metropolis has grown from 3,170 in 1921 to 722,664 in 2006 (FRN 2006) [26], and this must have increased tremendously since 2006. Enugu has a good network of paved roads. The area under investigation is

within the humid tropical rain forest zone of Southeastern Nigeria, with the coordinates defined by longitudes 7.27°E - 7.32°E, and latitudes 6.24°N - 6.30°N. Rainy and dry seasons are the two dominant seasons in the study area. The season of rainfall which starts from April and ends around October is usually accompanied by strong thunderstorms together with air mass moving towards the northern part of the city. The stormy runoff results in leaching and sheet erosion which most often leads to erosion gullies [27]. The average temperature across the study area ranges from 20.30ºC to 32.16ºC in the wet season and dry seasons respectively. The dry periods which are often associated with low visibility mostly at night and in the early morning hours due to the Harmattan dust haze are usually between November and February. The elevations of the study area above sea levels are between 182.88 - 219.45 metres [28]. Some of the soil types found within Enugu Metropolis include ferralitic soils (which are reddish to brown soils derived from sandstones and shales), litho-soils, and hydromorphic soils.

Sample collection

Ten major roadways were selected in Enugu metropolis based on traffic load, population density, and anthropogenic activities. These locations have a mean traffic density of 6,000 vehicles/day. The selected roads include Chime Avenue (CH), Agbani Road (AG), Ogui Road (OG), Okpara Avenue (OK), Abakaliki Road (AI), Abakpa Nike Road (AB), Zik Avenue (ZK), Obiagu Road (OB), Onitsha Road (ON) and Kenyatta-Ugwuaji Road (KU). Sampling was done on each road at intervals of 500 m. The sediment samples were collected by sweeping an area of 1 m^2 on the road pavements and curbs. They were packed using a clean plastic dustpan and stored in polyethylene bags, carefully labeled, and taken to the laboratory for analysis. At each sampling point, 3 sub-samples (1 m apart from each other) were taken, pooled, and homogenized to obtain a composite sample (about 200 g) for each point. In all, thirty-two (32) road deposited sediment (RDS) samples were collected for analysis. All the points were geo-referenced with a portable hand-held GPS unit, Garmin *eTrex Venture HC.* To eliminate temporal variations, all the RDS samples were collected in February 2014.

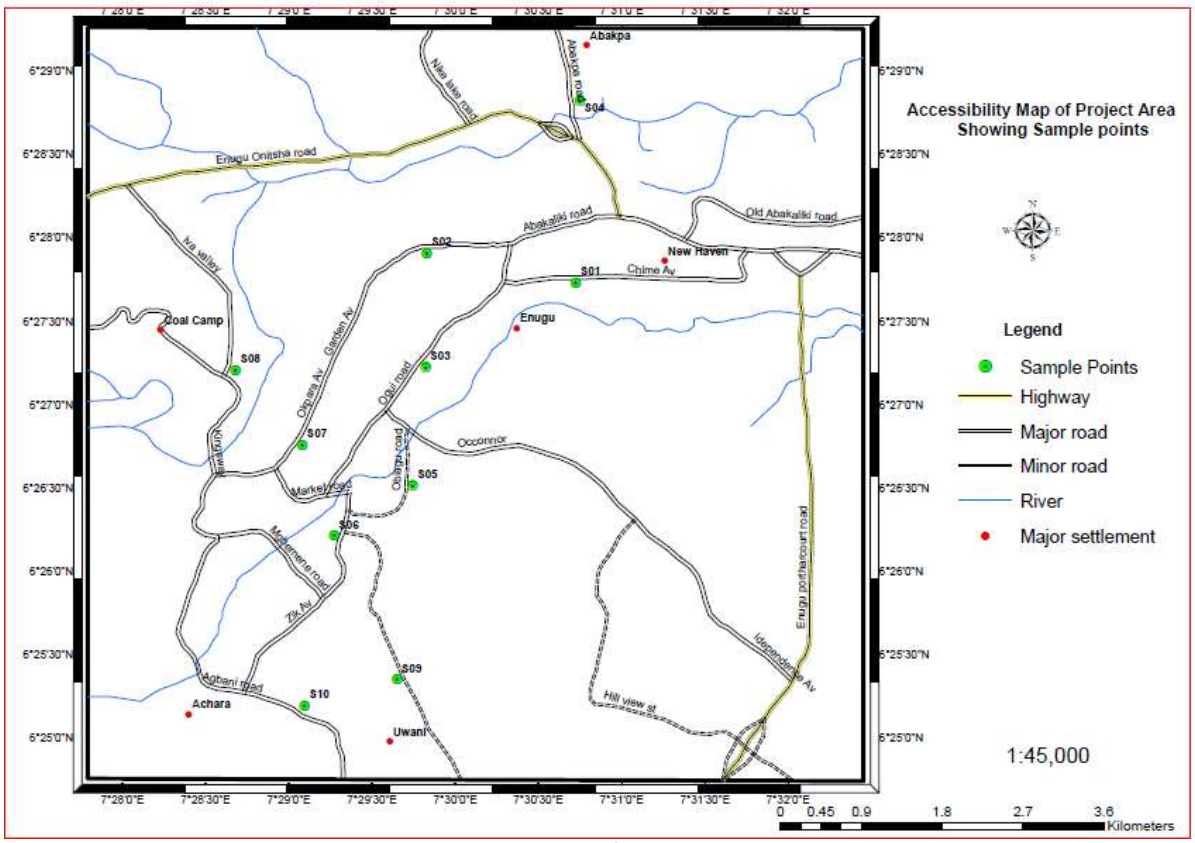

Figure 1. Map of study location

Sample pre-treatment and analysis

The collected samples were air-dried at room temperature and sieved through a 1 mm sieve to get rid of extraneous materials. The dried samples were further passed through a 230-mesh size (63 μm aperture) plastic sieve to get fine particles. A high precision analytical balance (Ohaus EB series, China) was used to accurately weigh 1g of each sample. The weighed samples were subsequently transferred into a 50 cm^3 glass beaker and digested at 105℃ on a hot plate for 2 hours in a fume cupboard. 20 cm^3 of a mixture of concentrated HNO_3, HF, and $HClO_4$ in the ratio of 2:2:1 by volume were used for the digestion. The digested samples were cooled and filtered with Whatman No. 1 filter paper. The filtrates were made up to 100 cm^3 with distilled deionized water produced with Eco-Still Mark, BSIC/ECO-4 (Bhanu Scientific Instruments Company, India) in a volumetric flask. Concentrations of metals in the digested samples were established using Buck 210 VGP atomic absorption spectrophotometer. The blanks and duplicates were similarly determined. Samples were analyzed for pH and electrical conductivity using pH meter (LIDA Instruments) and EC meter (Sanxin, SX 723) respectively, in a 1:2 (w/v) sample/water suspension. Walkley-Black wet acid oxidation method was used to analyze the total organic matter and total organic carbon of the road deposited sediments [29].

Quality control

Standard procedures were maintained following quality laboratory assurance to ensure the reliability of the analytical results. Double distilled deionized water using Eco-Still Mark, BSIC/ECO-4 (Bhanu Scientific Instruments Company, India), was used throughout the sample preparation and determinations. High-quality analytical grade standard reagents, metal standards, and chemicals were purchased from Finlab Owerri, Imo State, and were used without further purification. All the containers and glassware used for the analysis were properly washed with detergent and rinsed with distilled deionized water and dried in an oven using DHG – 9023A (B. Brans Scientific and Instrument Company, England). Precision and accuracy were ensured during the assay with the instruments through triplicate analysis of the sample alongside the reference standards and blanks. A flame atomic absorption spectrophotometer (Buck scientific VGP 210 model) was used for the heavy metal analysis. The instrument has a wavelength range of 190 to 900 nm, an accuracy of ±0.2 nm, a precision of ± 0.1nm, and reproducibility of <+5% RSD. The detection limits of the elements investigated and other instruments operating conditions of Buck's scientific VGP 210 model are stated in Table 1.

Table 1. Instrument operating conditions for heavy metal determination in RDS by FAAS

Metal	Wavelength (nm)	Slit width (nm)	Detection limit	Lamp current (mA)	Flame type
Cd	228.8	0.5	0.01	4	Air-acetylene
Co	240.7	0.2	0.05	7	Air-acetylene
Cr	357.9	0.2	0.04	7	Air-acetylene
Cu	324.7	0.5	0.005	4	Air-acetylene
Fe	248.3	0.2	0.05	6	Air-acetylene
Mn	279.5	0.2	0.07	5	Air-acetylene
Ni	232.0	0.2	0.05	4	Air-acetylene
Pb	217.0	1.0	0.04	5	Air-acetylene
Zn	213.9	1.0	0.005	5	Air-acetylene

Data analysis

The data obtained from the RDS were analyzed to determine the geo-accumulation index, enrichment factor, contamination factor (degree of contamination), and potential ecological risk index of the metals in the environment. Correlation co-efficient analysis and multivariate analysis were carried out the IBM™ SPSS

20.0 Software. Correlation co-efficient analysis measured the strength of the linear relationship between the variables while multivariate statistical analysis was used to ascertain the source(s) of the heavy metals in the roads. The multivariate analytical tools used include Principal Component Analysis (PCA) and Hierarchical Cluster Analysis (HCA). The PCA used varimax rotation and retention of principal components with eigenvalues >1 as suggested by the Kaiser criterion [30].

Geo-accumulation index (I_{geo})

The Geo-accumulation Index (I_{geo}) has been previously used to measure the level of heavy metal pollution in urban road deposited sediments [31-32]. I_{geo} is mathematically expressed as shown in equation (1).

$$I_{geo} = log_2 \left(\frac{C_{RDS}}{1.5Bs}\right) \quad (1)$$

Where C_{RDS} is the concentration of the elements in the sediment or road deposited sediment, B_s is the geochemical background value in the soil. In this study, the background geochemical concentrations of the metals (crustal average), were chosen as the background values for calculating the I_{geo} values [34]. The value of 1.5 is a constant which accounts for the natural variations of the heavy metals in the environment and accommodates a very small influence due to human origin [31]. The geo-accumulation index (I_{geo}) scale is made up of seven pollution grades of the geo-accumulation index of metals, ranging from unpolluted (0) to extremely polluted (6) [35].

Enrichment factor (EF)

The enrichment factor (EF) of a metal ion in the road deposited sediments is used to determine the extent of anthropogenic influence on a reference element. A reference element is often the one characterized by low occurrence fluctuation. It helps in distinguishing heavy metals that originated from anthropogenic activities and those from geogenic or natural sources. This is determined by the relationship in equation 2:

$$EF_y = [X_i/E_{i(ref)}] \div [Y_w/E_{i(ref)}] \quad (2)$$

where EF_y is the enrichment factor of the element Y, Y_i is the concentration of the element in the sample, $E_{i(ref)}$ which is the concentration of the reference element in the sample (Iron (Fe) was used as the reference element in this case), Y_w is the concentration of the element in the crust and $E_{w(ref)}$ is the concentration of the reference element used for normalization in the crust [34]. Five contamination categories are have been previously documented based on the enrichment factor according to Kartal et al. [36].

Contamination factor and degree of contamination

To evaluate the degree of heavy metal pollution in the RDS, contamination factor, and degree of contamination was employed [37]. C_f^i is the single element indicator which is estimated using equation 3:

$$C_f^i = C_{0-1}^i \div C_n^i \quad (3)$$

where C_f^i is the contamination factor of the element of concern, C_{o-1}^i is the value of the element in the road deposited sediment, C_n^i is the value of the background concentration. Continental crustal averages were used as the background concentration in the present study [34]. C_f^i is classified using four categories as follows: $C_f < 1$ indicates low contamination, $1 \leq Cf < 3$ implies moderate contamination, $3 \leq Cf < 6$ is an indication of significant contamination, while $6 \leq Cf$ stands for excessive contamination. There are also four descriptions of the degree of the contamination which include; $C_{deg} < 8$ = low degree of contamination, $8 \leq C_{deg} < 16$ = Reasonable degree of contamination, $16 \leq C_{deg} < 32$ = Substantial degree of contamination, and $32 \leq C_{deg}$ which means an elevated degree of contamination. The sum of the contamination factors of all the elements in the sample gives the extent of pollution as indicated in equation (4),

$$C_{deg} = \sum C_f^i \quad (4)$$

The potential ecological risk index

The concept of potential ecological risk index was initiated by Hakanson, as a way of estimating the heavy metal pollution from the viewpoint of nature and formation of sedimentary rocks [38]. It not only considers heavy metal levels in the soil but also links ecological and environmental effects with toxicology and

assesses pollution using a comparable and equivalent property index grading method. The potential ecological risk index is associated with the individual pollution coefficient, the toxic–response factor of the heavy metal, and the potential ecological risk factor of the individual metals. The relationship is given in equations (5), (6), and (7).[38]

$$E_r^i = T_r^i \quad (5)$$

$$RI = \sum_{i=1}^{n} E \quad (6)$$

$$C_f^i = C_s^i \div C_n^i \quad (7)$$

where $\boldsymbol{E_r^i}$ implies the potential ecological risk index of heavy metal $\boldsymbol{I}$, $\boldsymbol{RI}$ indicates the potential ecological risk factor of multiple metals, and $\boldsymbol{T_r^i}$ is the "toxic-response" factor of heavy metal $\boldsymbol{i}$. Hakanson proposed that $\boldsymbol{T_r^i}$ of the elements are in the order Zn = 1 < Cr = 2 < Cu = Ni = Pb = 5 < Cd = 30 [38]. $\boldsymbol{C_f^i}$ is the pollution co-efficient of the heavy metal $\boldsymbol{I}$, $\boldsymbol{C_s^i}$ is the measured concentration of heavy metal *i*, in the soil (mg/kg) while $\boldsymbol{C_n^i}$ denotes the background value of the heavy metal *i* in the unpolluted soil [34]. In this study, mean values of $\boldsymbol{E_r^i}$ and $\boldsymbol{RI}$ for Cd, Cr, Cu, Ni, Pb, and Zn were estimated for each metal contaminant. There are four categories of $\boldsymbol{RI}$ and five categories of E_r^i as described by Hakanson as shown in Table 2 [38].

Table 2. Potential Ecological Risk Categories based on E_r^i and *RI* values

E_r^i Value	Single-potential ecological risk (E_r^i)	RI value	Comprehensive-potential ecological risk
$E_r^i < 40$	Low risk	RI<150	Low risk
$40 \leq E_r^i < 80$	Moderate risk	150≤ RI<300	Moderate risk
$80 \leq E_r^i < 160$	Considerable risk	300≤RI< 600	Considerable risk
$160 \leq E_r^i < 320$	High risk	600≤RI	Very high risk
$320 \leq E_r^i$	Very high risk		

RESULTS AND DISCUSSION

Heavy metal concentrations

Table 3 presents the mean concentrations (mg/kg dry weight) of the heavy metals in the RDS samples. The concentrations of Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd and Pb in the road deposited sediments varied from 0.67 - 3.67, 2.33 - 7.67, 408.33 - 512.33, 3.00 - 61.75, 39.67 - 193.67, 11.50 - 90.00, 1.00 - 31.67, 0 - 1.33 and 0 - 46.67 mg/kg, respectively. The mean concentrations were 1.97, 4.49, 466.06, 16.63, 101.69, 59.28, 7.56, 0.81, and 16.25 $mgkg^{-1}$ for Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb respectively.

Table 3. Mean Concentrations (mg/kg dry weight) of heavy metals in RDS Samples

Roads	Metal Concentrations								
	Cr	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
CH	2.67 ±0.04	4.33 ±0.01	483.00 ±0.04	8.33 ±0.00	116.00 ±0.60	66.67 ±0.53	19.67 ±0.01	1.00 ±0.00	26.67 ±0.05
AG	2.25 ±0.01	5.25 ±0.05	463.50 ±0.52	3.00 ±0.00	40.50 ±0.10	77.50 ±0.45	9.50 ±0.10	0.00 ±0.00	0.00 ±0.00
OK	2.00 ±0.06	4.67 ±0.02	431.33 ±0.09	4.00 ±0.05	112.00 ±0.20	46.67 ±0.05	1.67 ±0.18	1.33 ±0.04	6.67 ±0.03
OG	2.25 ±0.01	4.00 ±0.06	455.25 ±0.38	61.75 ±0.25	87.75 ±0.03	11.50 ±0.10	1.50 ±0.10	1.00 ±0.00	15.00 ±0.10
AI	3.67 ±0.01	4.33 ±0.03	501.67 ±0.06	11.33 ±0.01	39.67 ±0.01	53.67 ±0.02	4.00 ±0.05	1.33 ±0.03	16.67 ±0.01
AB	2.33 ±0.03	7.67 ±0.01	495.67 ±0.01	9.00 ±0.03	193.67 ±0.34	56.67 ±0.01	5.67 ±0.02	0.67 ±0.01	13.33 ±0.03
ZK	1.33 ±0.01	3.67 ±0.02	512.33 ±0.07	10.67 ±0.05	134.33 ±0.01	90.00 ±0.00	31.67 ±0.01	1.00 ±0.00	16.67 ±0.03
OB	0.67 ±0.01	6.67 ±0.04	470.67 ±0.15	35.00 ±0.01	177.00 ±0.05	73.33 ±0.01	1.33 ±0.01	1.33 ±0.02	3.33 ±0.04
ON	1.00 ±0.00	2.33 ±0.01	443.33 ±0.06	9.67 ±0.01	85.33 ±0.02	56.67 ±0.03	1.00 ±0.01	0.67 ±0.01	46.67 ±0.12
KU	1.33 ±0.05	3.00 ±0.04	408.33 ±0.01	3.00 ±0.10	55.67 ±0.03	70.00 ±0.10	1.00 ±0.10	0.00 ±0.00	23.33 ±0.05
Range	0.67-3.67	2.33-7.67	408.33 -512.33	3.00 -61.75	39.67-193.67	11.50 -90.00	1.00 -31.67	0.00-1.33	0.00-46.67
Mean	1.97	4.59	466.06	16.63	101.69	59.28	7.56	0.81	16.25
SD	± 0.89	± 1.61	± 32.95	± 18.66	± 53.35	± 21.40	± 10.24	± 0.50	± 13.39
Background Values in Literature *	100	950	56300	25	76	55	70	0.2	12.5
Nigerian Soil Quality Standards**	100	NA	NA	50	70	100	421	3	164

***Taylor [34] **National Environmental Standards and Regulations Enforcement Agency [39], NA (Not available), SD (standard deviation)

Physicochemical properties

The result of pH, electrical conductivity (EC), total organic matter (TOM), and total organic carbon (TOC) of the RDS samples are shown in Table 4. The pH and EC levels indicate the nature of the chemical components of the road deposited sediments. The observed pH values in all the sampled sites were within the neutral-alkaline range: 7.31 (CH, OG) to 7.93 (KU). pH levels of RDS have a significant influence on the dynamics of metallic components as it regulates certain mechanisms like precipitation responsible for their retention in the soil [40]. The movement of metallic pollutants decreases as the soil pH increases (8 and above) [41]. The pH values of the samples therefore suggested that the movement and solubility of metallic pollutants in the soil samples is on the decrease. The EC is an indication of the presence of dissolved ions in a sample. The mean EC values measured ranged from 556 µS/cm (AB) to 578.67µS/cm (KU). EC is an indication that there is an appreciable level of ionizable materials in the samples which include cations and anions [42]. There was a slight variation in the EC values in the dust from all the sampled sites. The EC of soils is generally classified as non-saline < 2, moderately saline 2 – 8, very saline 8 – 16, and extremely saline > 16 [43]. The result of the present study indicates that the EC level is extremely saline based on the classification for EC. The mean values of TOM ranged from 4.38% (ON) to 7.09% (ZK), while the mean values of TOC ranged from 1.48% (ON) to 2.39% (ZK). The presence of organic matter significantly influences the binding of metallic elements in the soil. Also, some researchers have tested whether organic carbon (OC) compounds influence metal leaching [44]. Similarly, the influence of organic carbon on the solubility characteristics of Cd, Ni, and Zn has also been reported [44]. The high level of TOM and TOC suggested that apart from their natural sources, their content may have been enriched from other sources like hydrocarbons from vehicle oil or gasoline and organic materials from anthropogenic waste [45].

Table 4. Physicochemical parameters of road deposited sediments in Enugu Metropolis.

Roads	pH		EC (µS/cm)		TOM (%)		TOC (%)	
	Mean	Standard Deviation	Mean	Standard Deviation	Mean	Standard Deviation	Mean	Standard Deviation
CH	7.31	±0.54	567.67	± 26.58	7.00	± 1.01	2.36	± 0.35
AG	7.60	± 0.13	574.50	± 7.42	4.89	± 2.46	1.65	± 0.83
OK	7.75	± 0.44	573.67	± 16.17	5.30	± 2.07	1.78	± 0.70
OG	7.31	± 0.22	556.50	± 23.74	6.45	± 85	2.17	± 0.29
AI	7.66	± 0.62	573.00	± 16.82	5.51	± 2.23	1.86	± 0.76
AB	7.76	± 0.32	556.00	± 26.21	5.65	± 4.40	1.91	± 1.48
ZK	7.37	± 0.23	572.33	± 6.81	7.09	± 1.06	2.39	± 0.36
OB	7.54	± 0.35	572.00	± 24.02	6.67	± 2.77	2.25	± 0.93
ON	7.50	± 0.21	558.00	± 23.64	4.38	± 0.70	1.48	± 0.24
KU	7.93	± 0.41	578.67	± 16.29	6.73	±1.46	2.27	± 0.49

Correlation analysis of the parameters

The Pearson correlation matrix of the physicochemical properties and the heavy metals determined in the RDS samples are presented in Table 5. The result of the analysis revealed that Cu significantly correlated with Ni and EC at a 5 % (0.05) significance level suggesting that these metallic pollutants are from similar sources like industrial activities, vehicular emissions, etc. This could also mean that the presence of these heavy metals was enhanced by the electrical conductivity (EC) of the samples. Also, there is a significant correlation at 0.05 significant levels between Zn and Mn, Pb and pH, TOC, and TOM which indicates that apart from these metals having common origins such as traffic emissions, their retention in the samples was affected by the TOM and TOC while their mobility and solubility are probably affected by the pH of the samples.

Table 5. Pearson Correlation Matrix

		pH	EC	TOM%	TOC%	Cr	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
PH		1												
EC	Pearson Correlation	-.025	1											
	Sig. (2-tailed)	.893												
TOM%	Pearson Correlation	-.308	.005	1										
	Sig. (2-tailed)	.087	.979											
TOC%	Pearson Correlation	-.308	.004	1.000**	1									
	Sig. (2-tailed)	.086	.984	.000										
Cr	Pearson Correlation	.088	-.111	-.023	-.022	1								
	Sig. (2-tailed)	.632	.546	.900	.903									
Mn	Pearson Correlation	-.135	.080	.114	.114	-.097	1							
	Sig. (2-tailed)	.461	.664	.535	.536	.597								
Fe	Pearson Correlation	-.014	-.194	-.103	-.103	-.208	-.157	1						
	Sig. (2-tailed)	.939	.286	.575	.576	.253	.392							
Co	Pearson Correlation	.072	-.285	-.109	-.110	.140	.076	-.019	1					
	Sig. (2-tailed)	.697	.114	.551	.549	.446	.680	.916						
Ni	Pearson Correlation	-.306	.138	-.042	-.041	-.192	.129	.410*	-.171	1				
	Sig. (2-tailed)	.088	.450	.821	.824	.293	.482	.020	.348					
Cu	Pearson Correlation	-.208	.366*	.210	.211	-.069	.019	.136	-.287	.415*	1			
	Sig. (2-tailed)	.254	.039	.249	.247	.706	.918	.459	.112	.018				
Zn	Pearson Correlation	-.090	-.144	.094	.096	.215	-.357*	.112	-.054	-.034	.202	1		
	Sig. (-2-tailed)	.625	.431	.610	.603	.237	.045	.540	.770	.852	.267			
Cd	Pearson Correlation	.172	-.028	.084	.084	.093	-.048	.178	-.015	.082	-.131	.124	1	
	Sig. (2-tailed)	.346	.878	.649	.649	.612	.794	.330	.936	.654	.473	.500		
Pb	Pearson Correlation	-.379*	-.104	-.036	-.035	-.094	.086	-.154	-.113	.284	.222	-.045	-.202	1
	Sig. (2-tailed)	.032	.570	.846	.848	.608	.640	.400	.540	.116	.223	.807	.267	

Enrichment Factor (EF) analysis

Enrichment factors of the metals were determined according to Eq. 2 as shown in Table 6 for the RDS samples using the continental crust averages with Fe as the reference metal for normalization. The observed EF according to Eq.2 and Table 6 revealed that Mn (0.3 - 0.97) and Cr (0.79 - 4.37) obtained in the sampled RDS showed minimum to moderate enrichment. The EF values of the remaining metals showed minimal to extremely high enrichment: Co (16.09 – 331.23), Ni (65.47 – 319.65), Cu 24.67 – 193.11), Zn (1.79 – 56.62), Cd (0.00 – 953.62), and Pb (0.00 – 500.65). Their mean values were 83.55, 171.97, 129.31, 13.77, 596.01 and180.59 respectively, and showed significant to extremely high enrichments. EF values of elements above 10 are regarded as having originated from anthropogenic activities [46]. Hence, EF can also be assumed to be an indicator of natural and anthropogenic sources [47]. The maximum EF obtained for Cd was 953.62 at OK and OB. There are high levels of anthropogenic activities on these roads which include high population density, traffic volume, and commercial activities. The fact that the background value of Cd in the soil is 0.2 mg/kg indicates that its enrichment is from human activities. The mean EF's of the metals decreased in the order Cd >Pb > Ni > Cu > Co > Zn > Cr > Mn. Low average EF values were recorded in the case of Mn which suggested that natural sources must have been responsible for the observed level. Combustion of fossil fuel and biomass, vehicular emissions, and industrial activities are the main sources of heavy metal contamination of the environment [48-49].

Table 6. Enrichment Factors of the metals.

Road	Metal concentrations								
	Cr	**Mn**	**Fe**	**Co**	**Ni**	**Cu**	**Zn**	**Cd**	**Pb**
CH	3.18	0.55	1	44.70	191.46	143.04	35.16	715.22	286.09
AG	2.68	0.67	1	16.09	66.85	166.29	16.99	0.00	0.00
OK	2.38	0.59	1	21.46	184.86	100.13	2.98	953.62	71.52
OG	2.68	0.51	1	331.23	144.83	24.67	2.68	715.22	160.92
AI	4.37	0.55	1	60.79	65.47	115.15	7.15	953.62	178.80
AB	2.78	0.97	1	48.28	319.65	121.59	10.13	476.81	143.04
ZK	1.59	0.47	1	57.22	221.72	193.11	56.62	715.22	178.80
OB	0.79	0.85	1	187.74	292.14	157.35	2.38	953.62	35.76
ON	1.19	0.30	1	51.85	140.84	121.59	1.79	476.81	500.65
KU	1.59	0.38	1	16.09	91.88	150.20	1.79	0.00	250.33
Range	0.79 -4.37	0.3- 0.97	1	16.09-331.23	65.47-319.65	24.67-193.11	1.79-56.62	0.00-953.62	0.00-500.65
Mean	2.33	0.58	1	83.55	171.97	129.31	13.77	596.01	180.59

Index of geo- accumulation

The results of the calculated I_{geo} for the metals are shown in Table 7. The ranges of the I_{geo} values of the metals are as follows: Cr (-5.38 to -7.97), Mn (-7.53 to -9.29), Fe (-7.38 to -7.70), Co (-3.64 to 0.72), Ni (-1.52 to 0.76), Cu (-2.85 to 0.12), Zn (-6.80 to -1.73), Cd (0 to 2.15) and Pb (-2.49 to1.32). Their mean I_{geo} values are 6.44, -8.37, -7.51, -1.96, -0.32, -0.61, -4.86, 1.40, -0.30 respectively. The mean I_{geo} value of Cd showed moderate contamination of the metal in the Enugu metropolis. There was moderate to heavy contamination of Cd at OK, AI, and OB. The maximum value of 2.15 was observed in these areas. These roads experience heavy vehicular and human traffic with considerable commercial activities. This indicates that this contamination is from human and industrial activities. Pb showed moderate contamination with values of 0.51, 1.32, and 0.31 for CH, ON, and KU respectively. These roads generally experience heavy human and vehicular traffic as well as commercial activities.

Table 7. Index of geo-accumulation

Road	Metal								
	Cr	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb
CH	-5.81	-8.38	-7.45	-2.17	0.02	-0.31	-2.42	1.74	0.51
AG	-6.06	-8.12	-7.51	-3.64	-1.49	-0.09	-3.47	0	0
OK	-6.23	-8.29	-7.62	-3.24	-0.03	-0.82	-5.97	2.15	-1.49
OG	-6.06	-8.48	-7.53	0.72	-0.38	-2.85	-6.14	1.74	-0.36
AI	-5.38	-8.38	-7.41	-1.73	-1.52	-0.62	-4.72	2.15	-0.17
AB	-6.01	-7.53	-7.41	-2.06	0.76	-0.54	-4.21	1.16	-0.49
ZK	-6.83	-8.59	-7.38	-1.82	0.24	0.12	-1.73	1.74	-0.17
OB	-7.97	-7.73	-7.48	-0.10	0.63	-0.17	-6.31	2.15	-2.49
ON	-7.24	-9.29	-7.59	-1.96	-0.42	-0.54	-6.80	1.16	1.32
KU	-6.83	-8.90	-7.70	-3.64	-1.04	-0.24	-6.80	0	0.31

Contamination factor

The contamination factor of the metallic elements under investigation was estimated according to Eq. 3 as shown in Table 8. The results revealed that Cr, Mn, Zn, and Fe had contamination factor values less than 1 suggesting low contamination resulting from anthropogenic factors. Cobalt showed moderate contamination at OG and OB while Nickel had moderate contamination in all the roads except AG, AI, and KU. Cadmium had considerable to very high contamination factors in all the roads except AG and KU. The very high contamination factors observed for Cd in OK, AI, and OB agree with the results of the Index of geo-accumulation. This shows high contamination of Cd from anthropogenic sources.

The degree of contamination (C_{deg}) as calculated according to Eq. 4 is presented in Table 8. The values for the sampled roads are as follows: CH (10.52), AG (2.24), OK (9.73), OG (10.08), AI (10.06), AB (8.46), ZK (10.64), OB (12.05), ON (9.63), and KU (4.02). These results show a moderate degree of contamination in CH, OK, OG, AI, AB, ZK, OB, and ON. A low degree of contamination was recorded at AG and KU. The result revealed an elevated degree of contamination for Cd at a value of 41.67 in all the roads sampled. Pb, Cu, and Ni also showed moderate degrees of contamination in the paved roads in Enugu Metropolis.

Table 8. Contamination factors of the metals

Road	Cr	Mn	Fe	Co	Ni	Cu	Zn	Cd	Pb	Degree of Contamination
CH	0.03	0.00	0.01	0.33	1.53	1.21	0.28	5.00	2.13	10.52
AG	0.02	0.01	0.01	0.12	0.53	1.41	0.14	0.00	0.00	2.24
OK	0.02	0.00	0.01	0.16	1.47	0.85	0.02	6.67	0.53	9.73
OG	0.02	0.00	0.01	2.47	1.15	0.21	0.02	5.00	1.20	10.08
AI	0.04	0.00	0.01	0.45	0.52	0.98	0.06	6.67	1.33	10.06
AB	0.02	0.01	0.01	0.36	2.55	1.03	0.08	3.33	1.07	8.46
ZK	0.01	0.00	0.01	0.43	1.77	1.64	0.45	5.00	1.33	10.64
OB	0.01	0.01	0.01	1.40	2.33	1.33	0.02	6.67	0.27	12.05
ON	0.01	0.00	0.01	0.39	1.12	1.03	0.01	3.33	3.73	9.63
KU	0.01	0.00	0.01	0.12	0.73	1.27	0.01	0.00	1.87	4.02
Degree of Contamination	0.19	0.03	0.1	6.23	13.7	10.96	1.09	41.67	13.46	

Potential Ecological Risk Index (PERI)

The heavy metal potential ecological risk and potential ecological risk indices in Enugu metropolis roads are shown in Table 9. The calculated values for the potential ecological risk (E^i_r) of the metals in all the roads revealed a low-risk classification of the metals: Cr, Ni, Cu, Zn, and Pb. Cd had a low-risk classification in CH, OG, AB, ZK, and ON. There was however moderate risk classification of Cd in OK, AI, and OB. The results of the potential ecological risk indices revealed an index of 1250 for Cd, which implies a very high-risk classification in the RDS in Enugu roadways. These results agree with those of other mathematical models used in assessing metallic contamination in the RDS.

Table 9. Heavy metal potential ecological risk and potential ecological risk indices of the study area

Road	E_i					
	Cr	**Ni**	**Cu**	**Zn**	**Cd**	**Pb**
CH	0.05	7.63	6.06	0.28	150.00	10.67
AG	0.05	2.66	7.05	0.14	0.00	0.00
OK	0.04	7.37	4.24	0.02	200.00	2.67
OG	0.05	5.77	1.05	0.02	150.00	6.00
AI	0.07	2.61	4.88	0.06	200.00	6.67
AB	0.05	12.74	5.15	0.08	100.00	5.33
ZK	0.03	8.84	8.18	0.45	150.00	6.67
OB	0.01	11.64	6.67	0.02	200.00	1.33
ON	0.02	5.61	5.15	0.01	100.00	18.67
KU	0.03	3.66	6.36	0.01	0.00	9.33
RI	0.39	68.55	54.79	1.10	1250	67.33
Potential ecological risk index	Low risk	Low risk	Low risk	Low risk	Very high risk	Low risk

Principal component analysis (PCA)

The result of the principal component analysis of the present study is presented in Table10. PCA was carried out to detect the likely sources of heavy metal pollution of RDS in the Enugu metropolis. The component loadings as well as the eigenvalues are shown in Table 10. Three components have initial eigenvalues higher than one (>1) as indicated in Table10. The first component is dominated by Ni and Cu (with a high variation value of 0.676 and 0.788) accounting for 21.775%, while the second component is dominated by Zn and Mn (with a high variation value of 0.790 and -0.687) accounting for 18.039%. However, the third principal component is dominated by Fe and Cd with a variation value of 0.811 and 0.603 respectively, accounting for 14.715%. There is a close association between Ni and Cu in Component 1. These metals showed a high positive correlation, suggesting that the observed levels of these metallic pollutants are influenced by related anthropogenic inputs. This may have arisen from metallic works and related industrial sources. The dominance of Component 2 by Zn and Mn with a high variation value could only attest that they are not of the same source. While Zn is obtained from mining and industrial activities, Mn is a naturally occurring contaminant. It can also come from man-made sources like welding, mining, etc. The third Component which is dominated by Fe and Cd may be associated with a mixed source. There is however a contribution from natural sources of Fe while Cd is contributed from metal-related works and other industrial sources.

Table 10. The component matrix and initial Eigenvalues of the 9 heavy metals in the RDS

	Component Matrix			Initial Eigenvalues		
	1	**2**	**3**	**Total**	**% of Variance**	**Cumulative %**
Cr	-0.427	0.279	-0.397	1.960	21.775	21.775
Mn	0.081	-0.656	0.262	1.628	18.093	39.868
Fe	0.452	0.469	0.546	1.324	14.715	54.583
Co	-0.472	-0.088	0.255	0.980	10.892	65.475
Ni	0.802	0.026	0.219	0.912	10.130	75.605
Cu	0.705	0.102	-0.355	0.763	8.480	84.085
Zn	0.043	0.712	-0.419	0.615	6.836	90.922
Cd	-0.070	0.473	0.445	0.469	5.213	96.135
Pb	0.445	-0.386	-0.434	0.348	3.865	100.000

Cluster analysis

The result of hierarchical cluster analysis (HCA) is presented as a dendrogram in Figure 2. Hierarchical cluster analysis was carried out to segregate the components of different sources and to organize them into several classes. The HCA was accomplished according to Ward's Method and the distance apart is a reflection of the extent of the relationship among the metals. The dendrogram indicates that there are two main clusters or groups. Cluster one has Ni and Fe in one subgroup with Zn and Cu in another subgroup, while in cluster two there also two subgroups in which the first subgroup involves Cr, Cd, Mn, and Pb with Co alone in a separate subgroup. Cobalt seems to be an outlier since it has no relationship with other metallic pollutants. This implies that factors that influenced the presence of this metal did not affect the other metals that formed coherent clusters.

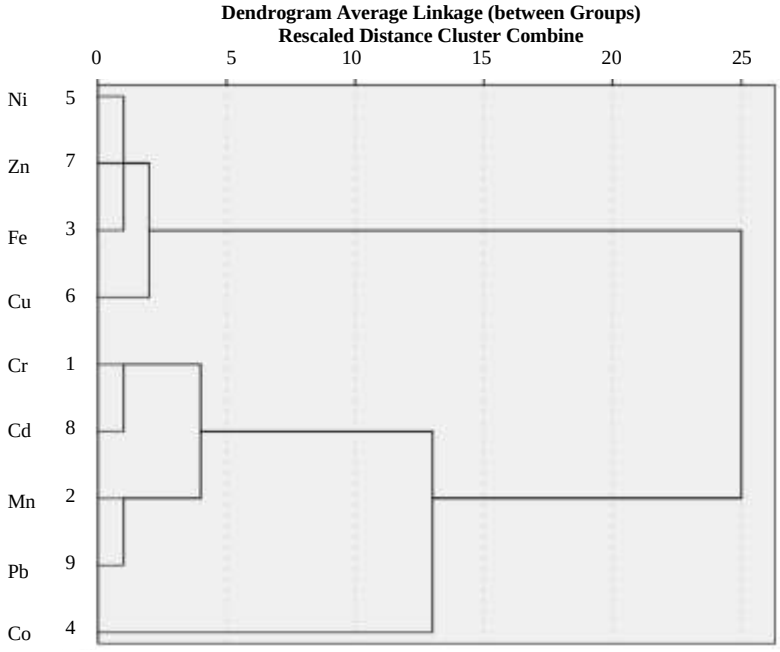

Figure 2. Dendrogram of heavy metal pollutants in the study area

Spatial representation of heavy metal concentrations in Enugu and environs

The concentrations of the individual analyzed metallic contaminants were used to generate the various geospatial maps using ArcGIS 10.3. The spatial variability of the contaminants is shown in Figures 3 - 6. Low cadmium concentration was noticed mainly within the southwestern half of the map with concentrations ranging from -0.05375 – 0.622 mg/kg. Areas with low

concentrations include Zk, AI, and AB areas (Figure 3). The map also revealed that the highest concentrations of cadmium 2.6485-2.9836 mg/kg were observed around OG. High chromium concentration was noticed within the northeastern part of the study area with concentrations of 2.733 – 3.0 mg/kg recorded around New Haven, Ogbete, Independence Layout, etc (Figure 4). Similarly, the highest lead concentration 23.33 – 46.67 mg/kg was recorded around KU and ON while low concentrations were observed at other areas including New Haven, Ogbete, and AB areas (Figure 5). Finally, the highest concentration of copper which ranges from 76.853-82.634mg/kg was obtained around Z A and Achara Layout areas with very low values noticed around the northwestern half of the map especially around OG and Asata areas (Figure 6).

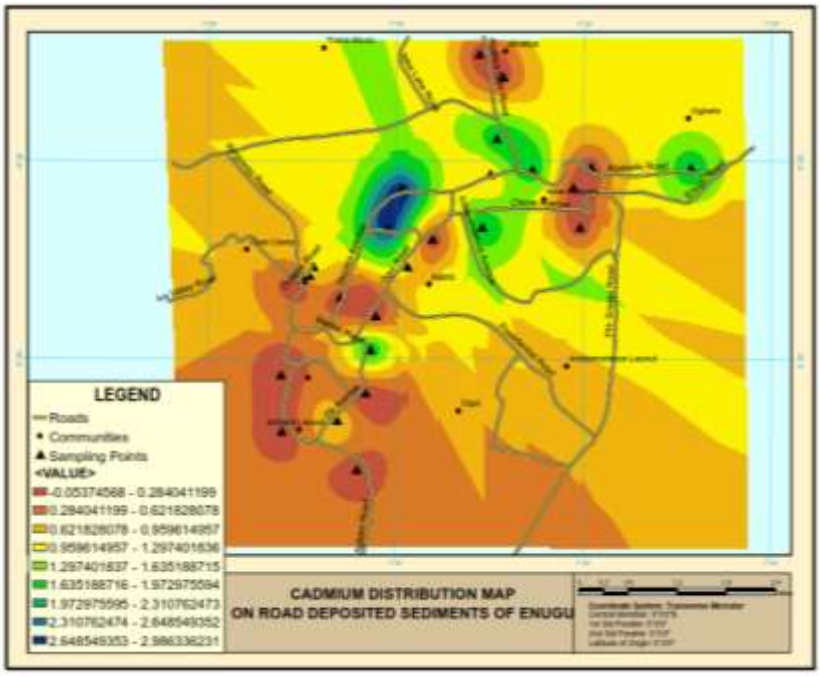

Figure 3. Spatial distribution of cadmium concentration across the study area

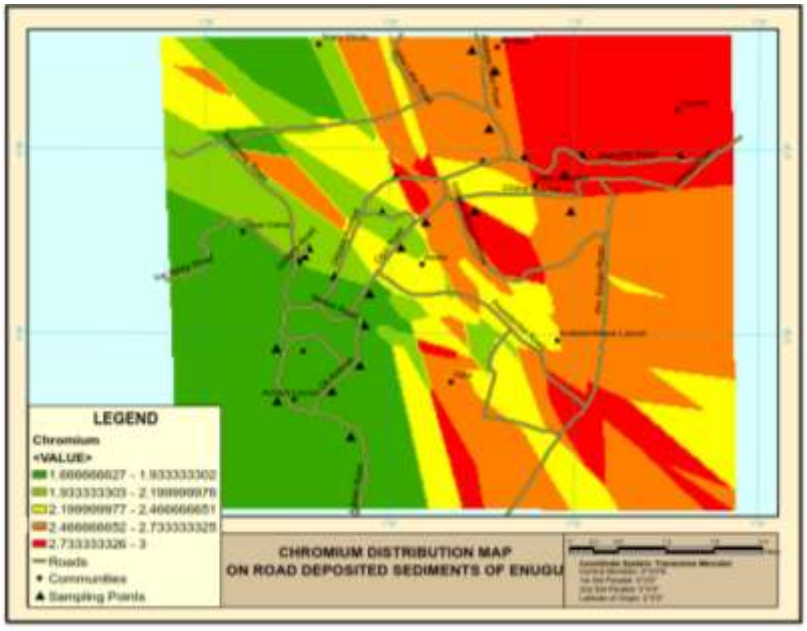

Figure 4. Spatial distribution of chromium concentration across the study area

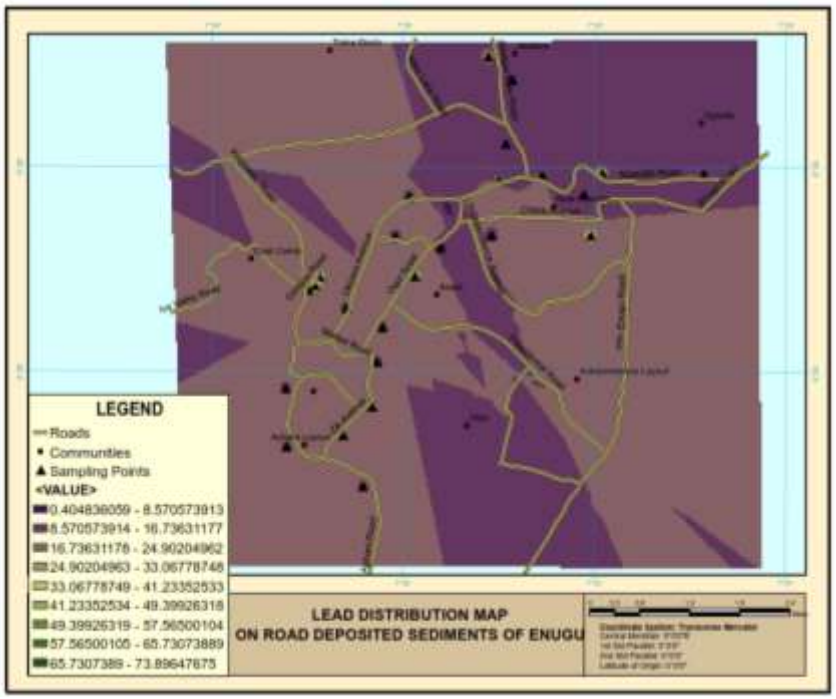

Figure 5. Spatial distribution of lead concentration across the study area

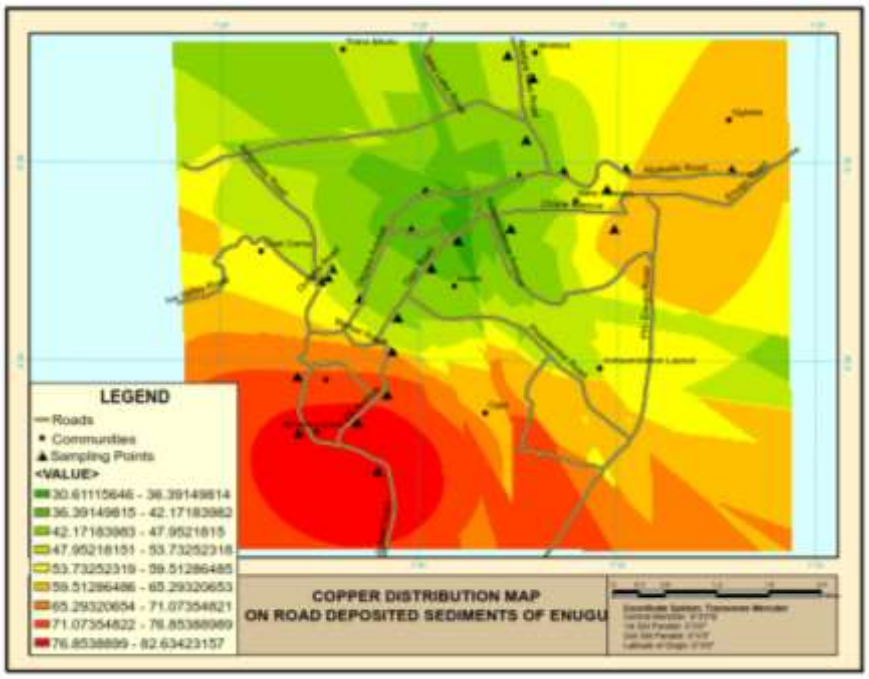

Figure 6. Spatial distribution of copper concentration across the study area

SUMMARY AND CONCLUSIONS

The levels, as well as sources of the metallic contaminants in the road deposited sediments (RDS) in some major roads in Enugu Metropolis, were investigated. The identified metallic pollutants in the RDS include Cr, Mn, Fe, Co, Ni, Cu, Zn, Cd, and Pb. The mean concentrations of Ni, Cu, Cd, and Pb were found to be greater than their background values. The concentration of Co (211 mg/kg) at the Nnamdi Azikiwe Stadium, along OG was above its background value, indicating high anthropogenic influence. The calculated enrichment factors for the metallic elements indicated that Mn had minimal enrichment; Cr and Zn showed moderate to considerable enrichment; Co, Ni, Cu, Cd, and Pb showed extremely high enrichment of the elements in the RDS samples. The estimated index of geo-accumulation showed results within moderate to

heavy pollution by Pb and Cd. This is an indication that the buildup of these heavy metals in the RDS could be of anthropogenic origin. The estimated contamination factors of the elements also disclosed that Zn, Co, Cu, Pb, Ni, and Cd yielded values ranging from low to very high contamination. Degree of contamination analysis showed that OB road is highly contaminated. The results suggested that contamination of the sampled roads are in the order; OB > ZK > CH > OG > AI > ON > AB > KU > AG. The Potential ecological risk indices calculated for the metals showed that Cd had a very high potential ecological risk in the roads. Enugu roads experienced an elevated flow of traffic and industrial activities which could contribute significantly to Cd deposition on the roads. Correlation analysis, PCA, and HCA established the association of the metallic elements under consideration. The result showed that there are three sources of heavy metal pollution in the study area. This includes the earth's crust, traffic (atmospheric depositions, corrosion, and the wearing off of vehicular parts), and industrial activities. Furthermore, the results provided explanations on the levels and origin of these metallic pollutants observed in the road deposited sediments. The study, therefore, recommends that the Government should enact laws to reduce the anthropogenic sources of these environmental pollutants. It is also advised that vehicles should be properly and regularly serviced as some of these heavy metals result from the wearing of tyres and brake parts. Besides, levels of these heavy metals should be monitored regularly in surface and groundwater sources in the area to ensure their potability. Therefore, there is a need to frequently monitor heavy metal levels in RDS, to avert the potential danger that might arise shortly due to the accumulation of these metallic pollutants in the environment.

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