Spatial Variation of heavy metal Pollution on road surfaces from Traffic sources in University of Nigeria, Enugu campus

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Article details:
Received: 17th Jan, 2019
Revision: 25th Jan, 2019
Accepted: 10th Feb, 2019
Published: 28th Feb, 2019

ABSTRACT

This study aims to analyze heavy metal concentrations on road deposited sediment (RDS) using the road network within Enugu Campus of University of Nigeria, (UNEC) as a study site. RDS samples were collected at two transverse positions from different sites within a week in order to describe the influence of traffic on heavy metal emissions. The heavy metal concentrations of the RDS were determined by strong tri-acid digestion and atomic absorption spectrometry. The mean concentrations for Co, Mn, Zn, Ni and Fe were found to be 11.53, 99.92, 221, 44.23 and 216 mg kg^{-1} from samples near the curb and 12.37, 92.25, 248.87, 43.62 and 197.5 mg kg^{-1} from samples 1 m from the curb respectively. The mean electrical conductivity (EC), pH and total organic matter (TOM) were found to be 304µS/cm, 7.13, and 6.9% from samples near the curb and 408µS/cm, 7.33 and 5.57% 1 m from the curb, respectively.

The results of the geochemical models: contamination factor (degree of contamination and enrichment factor agree to a very high level of contamination and enrichment of Zn in all the sampled sites. These results suggest a high anthropogenic influence on the heavy metals levels.

Keywords: *University of Nigeria, Enugu campus, heavy metals, AAS, road deposited sediments*

1. INTRODUCTION

Roads are known as the second largest non-point source of creating pollution in urban environment.[1] Road and highway surfaces are impervious, and serve as temporary sinks for various types of pollutants that are washed off during rainfall to the surrounding environment.[2-4] Road surfaces receive varying amount of heavy metals by the process of atmospheric deposition, sedimentation, impaction and interception.[5] In urban environments, the top soils and road deposited sediments (RDS) or road dust are indicators of heavy metal contamination from atmospheric deposition. Industries, traffic, mining activities, smelters and construction are some of the main anthropogenic sources of heavy metal pollution. The traffic source includes vehicles (tyrewear, brake linings, fuel combustion, etc.) and road infrastructure (pavement wear, corrosion of galvanized steel crash barriers, etc.).[6] It has been reported that the pollutants such as As, Cd, Cr, Cu, Ni, Pb and Zn due to heavy traffic are at high concentrations on road surfaces, roadside soils and particulate matter which affect the environmental airquality.[7]It has been estimated that the annual amount of dust emitted into the air by Nigerian motor vehicles was 612,000 tonnes and 187,000 tonnes for unpaved and paved roads, respectively.[8]

Road deposited sediments (RDS) play a key role in road traffic pollution assessment and comprise a complex mix of particulates and contaminants derived from the road traffic environment which are of serious concern for environmental health supervisors.

Most of the previous RDS studies have been based on samples collected within 1 m from the curb along roads [1,9,10] or from road side soils.[11] Only a limited number of studies have looked at carriageway positions further from the curb and these found that a larger proportion of particles below 150 µm are found towards center of the road compared to near the curb where larger particles are dominant. Similarly, heavy metal concentrations in RDS were

higher in the centre of the road and decreased towards the road gutter - confirming their higher association with the smaller particle size fraction.[12, 13]

Although, some previous studies have focused on some major cities in Nigeria, there is scanty information on pollution build-up and heavy metal concentrations around Enugu.

The objectives of this study therefore are: (1) to characterize heavy metal emissions on road surfaces for both positions of near the curb and 1 m from the curb towards the center and (2) to analyze heavy metal concentrations in RDS in order to describe emission patterns in the road traffic environment. (3) To conduct a contamination assessment using geochemical models.

2. MATERIALS AND METHODS

2.1. Study Area

This study is based on the paved road network within Enugu Campus of University of Nigeria, (UNEC) located inside Enugu town, behind Independence Layout. The Enugu Campus has just four faculties, namely: Business Administration, Environmental Studies, Law and Medical Sciences. It is a smaller campus, compared with Nsukka campus, which happens to be the main campus of University of Nigeria. Its land mass is 200 hectares. Its co-ordinates are 6°25'40"N and 7°30'29"E. The sampling sites were 1 and 2 lanes of traffic with asphalt road surfaces, with low traffic densities. The surrounding land area is devoted to campus buildings, some farm land, foot paths, pavements and staff residential quarters.

2.2 Site Description

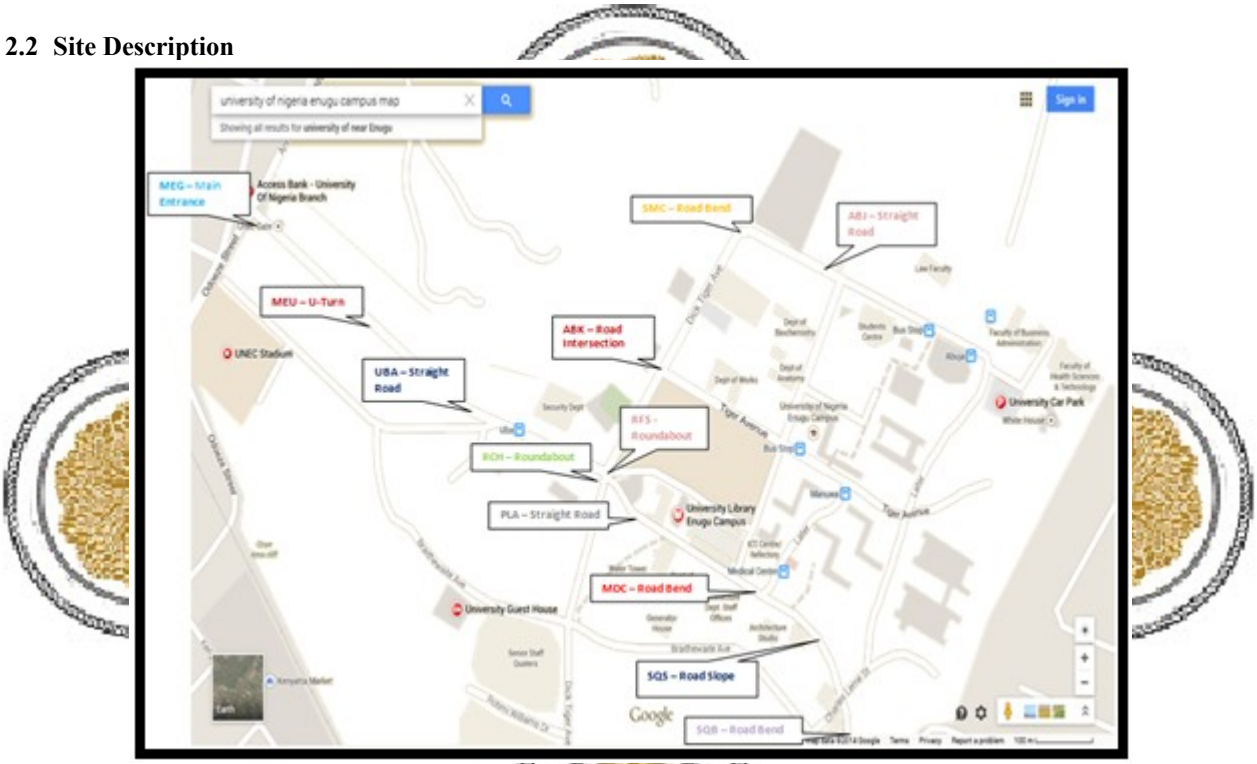

Figure 1: Map showing the University of Nigeria, Enugu Campus and sampling sites

Table 1: Description of sampling sites

Site No	Sampling Sites	Site code	Description of sampling sites
1	Main Entrance Gate	MEG	2 lane asphalt road, all vehicles slow down here while exiting and entering the University
2	Main Entrance, U-turn	MEU	A U-turn along the main entrance road
3	UBA	UBA	Location of a bank and a construction site
4	Roundabout, FS	RFS	Roundabout
5	Roundabout, Chapel	RCH	Roundabout
6	Parking lot, Auditorium	PLA	Parking lot of the main auditorium
7	Medical Centre	MDC	Road bend to the University medical centre

8	Staff Quarters	SQS	Slopy road leading to the staff quarters
9	Staff Quarters	SQB	Road bend within the staff quarters
10	Staff Quarters	SQI	Road intersection leading out of the quarters
11	Abuja Road	ABJ	Long stretch of road leading to classrooms, business centres, hostels and offices.
12	St. Mulumba Church	SMC	Road bend by a Roman Catholic Church
13	Access Bank	ABK	Road Intersection by a bank and construction site.

2.3 Sample Collection

RDS samples were collected from thirteen (13) sites on the campus roads and for two transverse positions (near the curb; and from 1 m further away from the curb towards the center of the road) to characterize the influence of road-traffic on heavy metal deposition. A close inspection of the sites showed that the RDS particles originate primarily from the vehicles, road surface, atmospheric deposition and the surrounding land. The sites comprise a road bend, a roundabout, a U turn, a road intersection, a slope and a typical road section to represent a typical road layout. Samples were collected within a week in February 2014 to avoid temporal variation. Sampling plots comprising a 1 m^2 road surface area for both positions were initially cleaned by repeated sweeping. Then RDS samples were collected in a clean plastic dustpan after sweeping according to literature[1, 4, 14]. The sweeping technique was also kept consistent to avoid sample variability. During sampling, care was taken to minimize sweeping pressure so that (artificial) detachment of road material particles could be avoided. Samples were then transported back to the laboratory in sealed and well labeled plastic bags to avoid contamination.

2.4 Sample Pretreatment and Analysis

After collection, samples were air-dried at room temperature and weighed prior to further analysis. Samples were dry-sieved using a 1 mm aperture metal sieve to remove large objects, litter, plants and leaves. The dried samples were further passed through a 230 mesh size (63 µm aperture) sieve to separate them. This size has some importance for metal concentrations as suggested by various researchers.[15] 1 g of the RDS was weighed with an electronic balance. They were placed on a hot plate at 105°C in 50 cm^3 beakers and digested for 1hr 30 minutes with 20cm^3 acid mixture of concentrated HNO_3, HF and $HClO_4$ (4:4:1) by volume, in a fume cupboard. The digested samples were cooled and filtered with Whatman No. 1 filter paper. The filtrates were made up to 100 cm^3 with distilled water in a volumetric flask. A Buck Model 210 VGP atomic absorption spectrophotometer was used to analyze for Zn, Cu, Cd, Cr, Ni, Pb, and Fe. The sample pH in water was carried out using a glass electrode (LIDA Instruments), while electrical conductivity (EC) was determined using Sanxin, SX 723 model electrical conductivity meter, both in a 1:2 (w/v) sample/water suspension. Walkley-Black chromic acid oxidation method was used to determine the total organic matter (TOM) of the samples.

2.5 Geochemical Models

Contamination factor (and Degree of Contamination)

To assess the extent of contamination of heavy metals in RDS, contamination factor and degree of contamination have been used.[16] The C_f^i is the single element index which is determined by the relationship:

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

where C_f^i is the contamination factor of the element of interest, $C_{o\text{-}1}^i$ is the concentration of the element in the sample, C_n^i is the background concentration. In this study, the continental crustal averages have been used.[17] C_f^i is defined according to four categories as shown in Table 2. The sum of the contamination factors of all the elements in the sample gives the degree of contamination as indicated in equation 2:

$$C_{deg} = \Sigma C_f^i \quad (2)$$

Table 3 presents the description for the various regimes of Degree of Contamination

Table 2: Description of Contamination Factor (C_f)

Value	RDS Quality
C_f¿ 1	Low contamination factor indicating low contamination
1 $\leq Cf < 3$	Moderate contamination factor
3 $\leq Cf < 6$	Considerable contamination factor
$6 \leq Cf$	Very high contamination factor

Table 3: Description of Degree of Contamination (C_{deg})

Value	RDS Quality
C_{deg}¿ 8	Low degree of contamination
8 ≤C_{deg}¿ 16	Moderate degree of contamination
16 ≤C_{deg}¿ 32	Considerable degree of contamination
32 ≤ C_{deg}	Very high degree of contamination

Enrichment Factor (EF)

Enrichment Factor (EF) of an element in the studied samples is based on the standardization of a measured element against a reference element. A reference element is often the one characterized by low occurrence variability. It is used to differentiate heavy metals originating from human activities from those of natural sources. This is determined by the relationship:

$$EF_X = [X_S / E_{S(ref)}] \div [X_C / E_{C(ref)}] \quad (3)$$

where EF_X is the enrichment factor for the element X, X_S is the concentration of element of interest in sample, $E_{S(ref)}$ is the concentration of the reference element used for normalization in the sample, X_C is the concentration of the element in the crust and $E_{C(ref)}$ is the concentration of the reference element used for normalization in the crust.[17] Five contamination categories are recognized on the basis of the enrichment factor as shown in Table 4.[18]

Table 4: Description of Enrichment Factor (EF)

Value	RDS Quality
EF¿ 2	Deficiency to minimal enrichment
2¿ EF <5	Moderate enrichment
5¿ EF <20	Significant enrichment
20 ¿ EF <40	Very high enrichment
40 ¿ EF	Extremely high enrichment

3. RESULTS AND DISCUSSION

3.1 Heavy Metal Concentration and Physico-chemical Parameters

All the data analysis was done using IBM-SPSS 20.0. The results of metal concentrations of the RDS samples are as given in the tables that follow. In Table 5, Cr was not detected in all the sites near the curb. Pb was detected only in site 1. This is not surprising since all the vehicles slow down at this point. Pb sources include exhaust emissions and road paint.[19] Cd whose sources include exhaust emissions, brake wear and road surface wear was found in small quantities at sites 2 and 4 where vehicles slow down as their brakes are applied. The highest emission of Zn was in site 4. This is also expected at a roundabout as Zn sources include exhaust emission, tyre wear, brake wear, road surface and infrastructure wear. The maximum Ni levels occurred at road bend in the staff quarters, site 9. Since this site experiences very low traffic density, the Ni source could be from the road surface and infrastructure wear apart from brake wear.[19]

Table 5: Metal Concentrations (mg kg^{-1}) and physico-chemical parameters near the curb

Site	Co	Mn	Zn	Ni	Fe	Cr	Cd	Pb	EC(µS /cm)	pH	TOM (%)
1	16	177	187	0	177	0	0	20	308	6.56	6.25
2	13	158	259	4	198	0	2	0	261	8.42	1.79
3	45	147	325	19	132	0	0	0	381	7.20	2.95
4	0	127	465	0	242	0	3	0	323	8.05	4.39
5	0	57	206	2	212	0	0	0	233	8.20	4.92
6	0	58	128	86	267	0	0	0	238	7.46	7.63
7	18	68	186	29	196	0	0	0	241	5.00	2.43
8	0	111	194	43	160	0	0	0	288	6.34	11.74
9	24	94	196	93	316	0	0	0	296	6.05	9.95
10	8	46	155	87	244	0	0	0	412	6.78	12.84
11	3	77	178	90	294	0	0	0	254	6.60	9.60
12	0	76	165	35	179	0	0	0	288	6.09	8.33
13	23	103	238	87	202	0	0	0	432	9.20	7.69
Range	0 - 45	46-177	128- 465	0-93	132-	0	0-3	0-20	233-	5.00	1.79-

					316				432	-9.20	12.84
Mean ± SD	11.5 ±13.61	99.92 ± 41.89	221.69 ± 88.52	44.3 2±3 8.89	216.85 ± 53.35	0	0	0	304.23 ± 66.23	7.14 ± 1.12	6.96 ± 3.54

SD: Standard Deviation.

The mean value of the pH: 7.14, shows that the samples are within the neutral range. The mean value of the EC: 304.23µS/cm indicates that the samples are extremely saline.[20] Also, the mean value of the TOM of the samples at 6.96 % indicates a very high organic content. The abundance of these pollutants near the curb follow the sequence Zn>Fe>Mn>Ni>Co.

Table 6: Metal concentrations (mg kg^{-1}) and physico-chemical parameters, 1 m from the curb

Site	Co	Mn	Zn	Ni	Fe	Cr	Cd	Pb	EC (µS/cm)	pH	TOM (%)
1	15	145	535	0	166	0	1	0	254	6.42	8.40
2	16	43	134	17	244	0	0	0	259	9.25	5.78
3	19	137	480	18	199	0	0	0	760	7.00	7.34
7	0	111	194	43	160	0	0	0	458	6.41	2.83
9	0	113	198	27	179	0	0	0	451	6.53	8.10
10	42	115	196	70	186	0	0	0	441	8.35	7.92
11	0	0	55	68	121	0	0	0	421	8.50	0.17
13	7	74	199	106	325	2	0	20	223	6.20	4.05
Range	0- 42	0-145	55-480	0-106	121-325	0-2	0	0-20	223-760	6.20-9.25	0.17-8.40
Mean ± SD	12.36 ± 14.29	92.25 ± 49.79	248.88 ± 167.74	43.63 ± 35.31	197.50 ± 62.27	0	0	0	408.38 ± 173.06	7.33 ± 1.18	5.57 ± 2.98

Cr, Cd and Pb were each detected in only 1 sampling site, sites 13, 1 and 13 respectively. The detection of Cr and Pb at site 13 may not be unconnected to emissions from heavy duty vehicles moving to the construction site nearby. Zn had a high occurrence in all the sites with the maximum occurring at MEG, site 1. This is expected as Zn sources include exhaust emission, tyre wear, brake wear and road surface wear. The maximum Ni levels occurred at a road intersection by a construction site, ABK. This could be attributable to the road wear imposed on the road by heavy duty vehicles conveying materials to the construction site nearby.

The mean value of the pH: 7.33, shows that the samples are within the neutral range. The mean value of the EC: 408.38µS/cm indicates that the samples are extremely saline.[20] Also, the mean value of the TOM of the samples at 6.96 % indicates a very high organic content. The abundance of these pollutants 1m away from the curb follow the sequence Zn>Fe>Mn>Ni>Co.

3.2 Correlations among the heavy metal pollutants and physico-chemical parameters

The correlation analysis was done to determine linear relationships among the metals. Pb and Cd were considered as outliers and were not considered in the correlation analysis.

Table 7: Correlation Analysis of heavy metals concentration near the curb

		Co	Mn	Zn	Ni	Fe	EC	pH	TOM
Co	Pearson Correlation	1	.433	.210	-.001	-.297	.487	-.060	-.341
	Sig. (2-tailed)		.140	.492	.997	.324	.091	.845	.254
	N	13	13	13	13	13	13	13	13
Mn	Pearson Correlation	.433	1	.506	-.557*	-.459	.173	.191	-.439
	Sig. (2-tailed)	.140		.077	.048	.115	.573	.532	.133
	N	13	13	13	13	13	13	13	13
Zn	Pearson Correlation	.210	.506	1	-.506	-.178	.270	.394	-.494
	Sig. (2-tailed)	.492	.077		.078	.560	.372	.183	.086
	N	13	13	13	13	13	13	13	13
Ni	Pearson Correlation	-.001	-.557*	-.506	1	.622*	.244	-.130	.706**
	Sig. (2-tailed)	.997	.048	.078		.023	.421	.671	.007
	N	13	13	13	13	13	13	13	13
Fe	Pearson Correlation	-.297	-.459	-.178	.622*	1	-.216	-.088	.381
	Sig. (2-tailed)	.324	.115	.560	.023		.478	.775	.199
	N	13	13	13	13	13	13	13	13

EC	Pearson Correlation	.487	.173	.270	.244	-.216	1	.314	.256
	Sig. (2-tailed)	.091	.573	.372	.421	.478		.297	.398
	N	13	13	13	13	13	13	13	13
pH	Pearson Correlation	-.060	.191	.394	-.130	-.088	.314	1	-.219
	Sig. (2-tailed)	.845	.532	.183	.671	.775	.297		.471
	N	13	13	13	13	13	13	13	13
TOM	Pearson Correlation	-.341	-.439	-.494	.706**	.381	.256	-.219	1
	Sig. (2-tailed)	.254	.133	.086	.007	.199	.398	.471	
	N	13	13	13	13	13	13	13	13

*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).

The result of the correlation analysis presented in Table 7shows that Mn is significantly correlated negatively with Ni at¿0.05 significance level; this indicates that these metal pollutants do not share common sources. Ni sources include brake wear, road surface wear and from road infrastructures.[19]There are also significant correlations at¿0.05 significance level between Ni with Fe and TOM. This indicates that the retention of these metals in the RDS samples is enhanced by the TOM of the samples.

Table 8: Correlation Analysis of heavy metals and physico-chemical parameters 1 m from the curb

		Co	Mn	Zn	Ni	Fe	EC	pH	TOM
Co	Pearson Correlation	1	.338	.255	-.006	.114	.118	.384	.561
	Sig. (2-tailed)		.414	.543	.988	.788	.780	.347	.148
	N	8	8	8	8	8	8	8	8
Mn	Pearson Correlation	.338	1	.797*	-.429	-.023	.327	-.619	.769*
	Sig. (2-tailed)	.414		.018	.289	.957	.429	.102	.026
	N	8	8	8	8	8	8	8	8
Zn	Pearson Correlation	.255	.797*	1	-.572	-.026	.268	-.497	.633
	Sig. (2-tailed)	.543	.018		.139	.951	.521	.210	.092
	N	8	8	8	8	8	8	8	8
Ni	Pearson Correlation	-.006	-.429	-.572	1	.404	-.221	-.014	-.515
	Sig. (2-tailed)	.988	.289	.139		.321	.600	.973	.191
	N	8	8	8	8	8	8	8	8
Fe	Pearson Correlation	.114	-.023	-.026	.404	1	-.387	-.152	.132
	Sig. (2-tailed)	.788	.957	.951	.321		.344	.719	.755
	N	8	8	8	8	8	8	8	8
EC	Pearson Correlation	.118	.327	.268	-.221	-.387	1	-.059	.115
	Sig. (2-tailed)	.780	.429	.521	.600	.344		.890	.786
	N	8	8	8	8	8	8	8	8
pH	Pearson Correlation	.384	-.619	-.497	-.014	-.152	-.059	1	-.181
	Sig. (2-tailed)	.347	.102	.210	.973	.719	.890		.668
	N	8	8	8	8	8	8	8	8
TOM	Pearson Correlation	.561	.769*	.633	-.515	.132	.115	-.181	1
	Sig. (2-tailed)	.148	.026	.092	.191	.755	.786	.668	
	N	8	8	8	8	8	8	8	8

The result of the analysis for metals 1 m from the curb as shown in Table 8 shows that Mn is significantly correlated with Zn and TOM. Since Mn pollution is from natural sources, this result could only mean that the presence of these metals is enhanced by the TOM of the samples.

3.3. Contamination factor (and Degree of Contamination)

The result of the contamination factors of the metals near the curb in Table 9 shows moderate contamination factors for Co, Mn, Ni and Fe. Zn however showed moderate to very high contamination factors in all the sites. The result of the degree of contamination shows low degree of contamination for Co, Mn, Ni and Fe. Zn showed a very high degree of contamination.

Table 9: Contamination Factors of metals near the curb

Site	Co	Mn	Zn	Ni	Fe
1	0.64	0.19	2.67	0	0.003
2	0.52	0.17	3.70	0.05	0.004
3	1.8	0.15	4.64	0.25	0.002
4	0	0.13	6.64	0	0.004
5	0	0.06	2.94	0.03	0.004
6	0	0.06	1.83	1.15	0.005
7	0.72	0.07	2.66	0.39	0.003
8	0	0.11	2.77	0.57	0.003
9	0.96	0.10	2.80	1.24	0.006
10	0.32	0.05	2.21	1.16	0.004
11	0.12	0.08	2.54	1.2	0.005
12	0	0.08	2.36	0.47	0.003
13	0.92	0.11	3.40	1.16	0.004
Deg. Of Contamination	**6**	**1.37**	**41.17**	**7.67**	**0.05**

Table 10: Contamination factors of metals 1 m from the curb

Site	Co	Mn	Zn	Ni	Fe
1	0.60	0.15	7.64	0	0.003
2	0.64	0.05	1.90	0.23	0.004
3	0.76	0.14	6.86	0.24	0.004
4	0	0.12	2.77	0.57	0.003
5	0	0.12	2.83	0.36	0.003
6	1.68	0.12	2.80	0.93	0.003
7	0	0	0.79	0.91	0.002
8	0.28	0.08	2.84	1.41	0.006
Deg. Of Contamination	**3.96**	**0.78**	**28.44**	**4.65**	**0.028**

The result of the contamination factors of metals 1 m from the curb in Table 10 shows low to moderate contamination factors for Co, Mn, Ni and Fe. Zn, on the other hand showed low to very high contamination factors in all the sites. The degree of contamination result shows that Co, Mn, Ni and Fe have low degrees of contamination while Zn had a considerable degree of contamination.

3.4 Enrichment Factor (EF)

Table 11: Enrichment factors of metals near the curb

Site	Co	Mn	Zn	Ni	Fe
1	184	48	719	0	1
2	150	43	995	14	1
3	519	40	1249	67	1
4	0	34	1787	0	1
5	0	15	792	7	1
6	0	16	492	305	1
7	208	18	715	103	1
8	0	30	746	153	1
9	277	25	753	330	1
10	92	12	596	309	1
11	35	21	684	319	1
12	0	21	634	124	1
13	265	28	915	309	1
Range	0 - 519	12 - 48	492-1787	0-330	1
Mean	**150**	**28**	**890**	**158**	**1**

The results of the EF calculations in Table 11 reveal the enrichment of some of the metals in the RDS samples. The enrichment factors of Cr (0 - 519) and Ni (0 - 330) obtained in all the sites near the curb showed minimal to extremely high enrichment. The EF values of Mn showed significant to extremely high enrichment: Mn (12 - 48). Zn however showed extremely high enrichments in all the sites. Their mean values were Co (150), Mn (28), Zn (890) and Ni (158). The elements with the EF values higher than 10, are considered to be mainly of anthropogenic sources. Hence, EF can also be assumed to be an indicator of natural and anthropogenic sources of heavy metals.[21] The mean EFs of the metals decreased in the order Zn¿ Ni¿ Co ¿ Mn.

Table 12: Enrichment factors of metals 1 m from the curb

Site	Co	Mn	Zn	Ni	Fe
1	190	43	2257	0	1
2	203	13	565	66	1

3	241	41	2025	70	1
4	0	33	819	167	1
5	0	34	835	105	1
6	532	34	827	273	1
7	0	0	232	265	1
8	89	22	840	413	1
Range	0-532	0-43	232-2257	0-413	1
Mean	**178**	**26**	**1089**	**177**	**1**

The result of the enrichment factors of metals 1m from the curb in Table 12 shows minimal to extremely high enrichment of the following metals Co, Mn, and Ni. Zn showed extremely high enrichment in all the sampling sites. Their mean values which are all greater than 10 suggest that the enrichment of these metals is from anthropogenic sources.[21]Fossil fuel combustion, traffic emissions and industrial processes are considered as the major pollution sources of heavy metal pollutants in the atmosphere.[22]

4. CONCLUSION

The concentrations and emission pattern of heavy metals in the road network within the University of Nigeria Enugu campus have been studied. Cr, Pb and Cd were considered as outliers. Their emission was limited to 1 or 2 sites. This means that there is no pollution on the roads in the campus by these metals. There was a higher occurrence of Zn and Co away from the curb than near the curb. Fe and Mn are from natural sources. They showed higher concentrations near the curb than 1 m away from the curb. These may be as a result of the manual sweeping of the roads towards the curb by the sanitation officers. The contamination factor and degree of contamination results showed that Zn has very high value in all the sites and position. The metals also showed higher enrichment 1 m from the curb than near the curb suggesting enrichment from anthropogenic source such as traffic emissions which include: exhaust emissions, brake wear, car body wear, and road surface wear among others.

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