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Sources, Transport Pathways and Ecological Risks of Heavy Metals Present in Roadside Soil Environment in Urban Areas

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The study investigated sources and transport pathways of heavy metals in roadside soils in urban areas of a typical nodal city, alongside the associated eco-risks. Soil, leaf and bark samples were collected along the routes and analysed for metal concentration using standard methods. The data were analysed using ANOVA, principal component analysis and factor analysis. The results showed that Pb, Cu and Ni in the roadside soil were found to be mainly from traffic emissions, whereas Cd in the soil was primarily through municipal waste incineration. Pb, Cd and Ni in leaf samples were absorbed from the soil via foliar uptake, while root uptake was the primary pathway for Cu in the leaves. Potential ecological risk indicated a moderate eco-risk for 5 routes with Cd being the primary contributor to the risk. Therefore, it is important to control the source of Cd to reduce the eco-risks associated with metal pollution.

Keywords: potential ecological risk index, traffic emission, plant uptake, heavy metal, urbanisation.

Introduction

Heavy metals are toxic pollutants that can have adverse ecological and human health consequences (Tchounwou et al., 2012). They are contributed to the urban environment through various anthropogenic activities, such as traffic and industrial activities, urban farming and incineration of municipal wastes (Nabulo et al., 2012). Heavy metals emitted by these sources as particulates or aerosols eventually get deposited on the pervious and impervious surfaces that serve as sinks in urban environment (Yang et al., 2013). Several studies have investigated the negative impacts associated with heavy metals, deposited on urban impervious surfaces, in particular, when they are washed off to the water sources during storm events (Ziyath et al., 2016, Gunawardena et al., 2015). The knowledge generated through these studies is useful in developing control measures to mitigate the urban water pollution and protect the aquatic system. Similarly, it is critical to investigate the characteristics and ecological health risks of heavy metals deposited on pervious surfaces, particularly in soil environment, since the non-biodegradability and persistence of heavy metals can cause anomalies in the geochemistry and facilitate their relative mobility and accumulation along the food chain and ecosystem.

Consequently, several recent studies have focused on the ecological risk assessment of heavy metals present in different soil environments, for example, in gold mine soil environment (Li et al., 2014), soil impacted by oil industry activities (Fatoba et al., 2016), soil at a waste disposal site (Ihedioha et al., 2016), soil around metal recycling plant (Ogunkunle et al., 2016) and around a cement factory (Ogunkunle et al., 2013). Similarly, many studies have investigated human health risks through plant uptake of heavy metals present in soil (Ogunkunle et al., 2015, Gu et al., 2016). However, in comparison with other soil environments, the roadside soil environment in urban areas has unique characteristics in terms of heavy metal sources and transport pathways due to associated traffic and land use activities. Therefore, it is important to investigate the sources and transport pathways of heavy metals in the roadside soil (RSS) environment to minimise the potential ecological health risks and to develop mitigation strategies.

Therefore, the objectives of this study were to (1) define the source and transport pathways of heavy metals to the roadside soil ecosystem and (2) assess the health impacts of heavy metals on the roadside soil ecosystem using the ecological risk indices. Roadside native trees were used as the representative of a soil ecosystem since tree bark and leaves of higher plants have been widely used to detect the deposition, accumulation, and distribution of metal pollution in urban environments (Serbula et al., 2013).

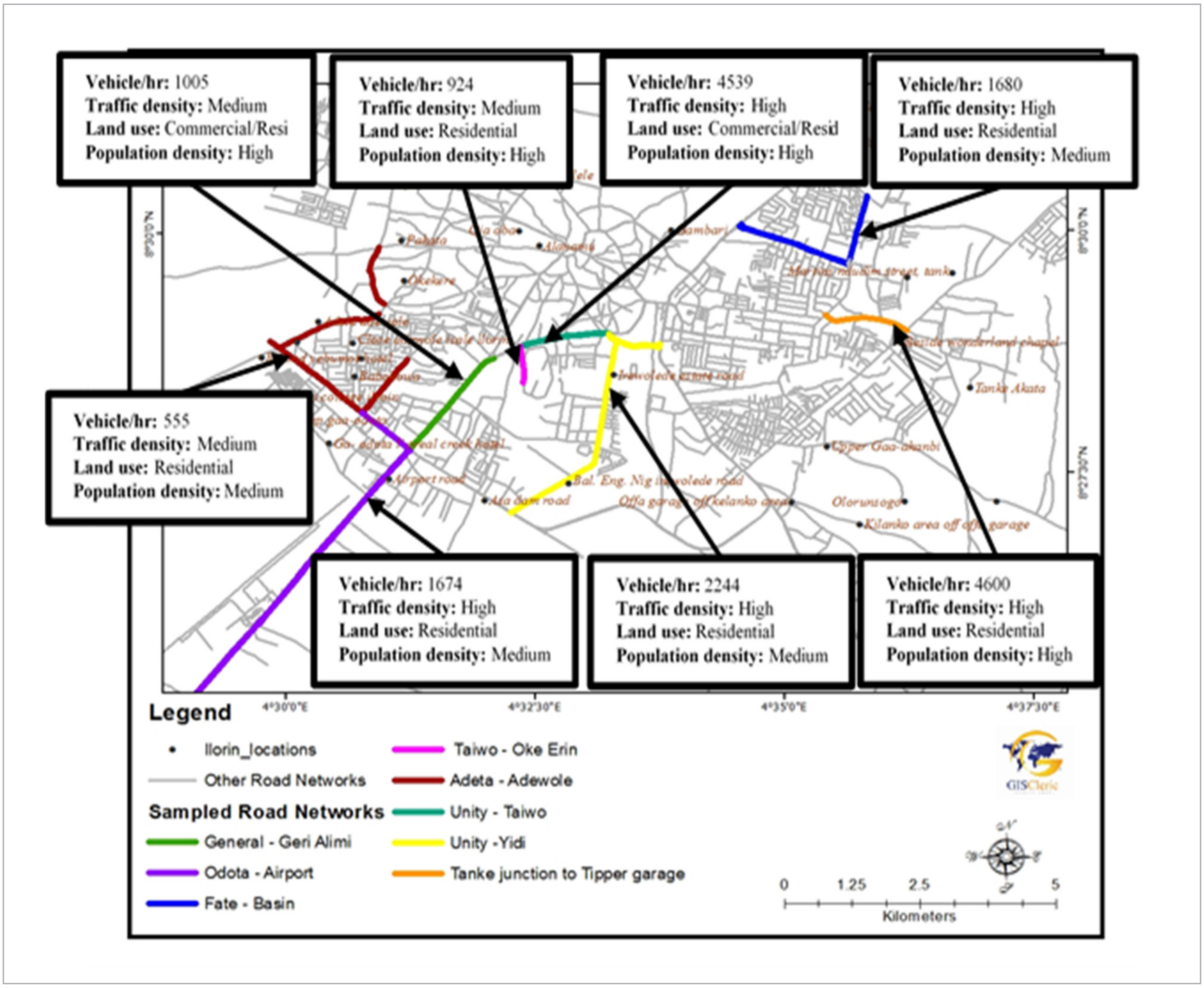
Materials and methods

Sample collection

The study sites were located in Ilorin, which is a Nigerian urban nodal city that interconnects the southwest, southeast and northern parts through air, road and rail paths (Fig. 1). Eight routes from Ilorin road network were chosen to represent different traffic, land use and population characteristics (Fig. 1). For each route, 10 sampling points, which were separated by either 200 m or 100 m depending on whether the road is over or under 2 km, respectively, were selected. For each sampling point, a composite and representative roadside soil (RSS) sample was prepared from 5 sub-samples collected using a plastic spatula at the depth of 0–15 cm from the centre and 4 corners of a 5 m^2 quadrant located within 10 m from the road pavements. Thus, a total of 80 representative samples were collected for the study, while 5 soil samples that represented the background/control were collected from the Botanical garden of the University of Ilorin (a distance of 25 km to the city). Similarly, tree species growing along the 3 main routes were identified to the species level, and samples of their leaves and barks were collected. On a standing tree, 3 to 4 matured leaves were sampled and combined to make a composite sample, while a composite sample for barks was prepared from sub-samples collected at 4 different breast height points around the tree bole using a stainless metal scalpel.

Fig. 1

Study area showing various road routes used in the study



Note:

GG: General – Geri Alimi, FB: Fate – Basin, AA: Adeta – Adewole, UY: Unity – Yidi,

OA: Odota – Airport, OT: Taiwo – Oke Erin, UT: Unity – Taiwo, TT: Tanke junction – Tipper garage

Laboratory analysis

Soil and plant samples were air-dried to a constant weight and ground into powders. Approximately 1 g of powdered sample was digested in 10 mL of HNO_3+$HClO_3$ (4:1) and tested using the atomic absorption spectrophotometry (Perkin Elmer A Analyst 200) to determine the concentrations of common heavy metals, namely Pb, Cd, Cu and Ni, present in soil and plant. Heavy metal analysis was done in duplicate, and the accuracy of the analysis was evaluated using two certified reference materials for plant and soil (IAEA 359- cabbage leaves and IAEA SL-1-lake sediment). Recovery rate of heavy metals ranged from 80% to 110% for soil and 85%-95% for plant. Reagent blanks were also included in the digestion as a part of the quality control procedure.

Ecological risk analysis

Geo-accumulation (Igeo) (Müller 1981) and potential ecological risk (PERI) (Hakanson, 1980) indices were used in this study to assess the impact of heavy metals on the ecosystem. The Igeo values were calculated using the following expression:

$$Igeo = \log_2\left(\frac{C_n}{1.5 \times B_n}\right)$$

where C_n is the concentration of a specific metal in soil and B_n is the background concentration of investigated metals in a relatively uncontaminated soil samples collected from the botanical garden at the University of Ilorin. The constant 1.5 was used to allow for the natural variation of heavy metals between the study sites (Ji et al., 2008).

The PERI integrates the factors of ecological risk potentials of individual metals and associates their ecological and environmental effects with their toxicology (Ogunkunle and Fatoba, 2013). The PERI was calculated as follows:

$$PERI = \sum_{r=i}^{r=n} E_r\,;\; E_r^i = T_r^i \times C_f^I;\; C_f^I = \frac{C_{n,i}}{B_n}$$

where C_f^I is the pollution factor for each metal in the roadside soil, $C_{n,i}$ is the concentration of a heavy metal in a soil sample, E_r^i is the potential ecological risk individual coefficient to define the single potential ecological risk of each metal, and T_r^i is the toxic response coefficient developed by Hakanson (1980). The toxic response coefficients are 30, 5, 5 and 5 for Cd, Cu, Pb and Ni, respectively.

2.4 Statistical analysis

The data analyses were conducted using the R Studio (Version 0.99.896), and the relevant codes used in this study are given in the Appendix. The outliers were identified using the box plots, while the normality test was undertaken using the Shapiro-Wilk test. Analysis of variance (ANOVA) was used to investigate the significance of variations in heavy metal concentrations, along with Tukey's honest significance difference (THSD) test as the post-hoc analysis. Multivariate principal component analysis (PCA) and factor analysis were used to investigate the source and transport pathways of heavy metals to RSS and native trees.

Results and discussion

The soil data matrix used in this study consisted of the concentrations of heavy metals present in RSS samples collected from 8 routes, while the plant data matrix was composed of heavy metal concentrations in barks and leaves of the native trees along the three main routes.

Characteristics of heavy metals present in soil

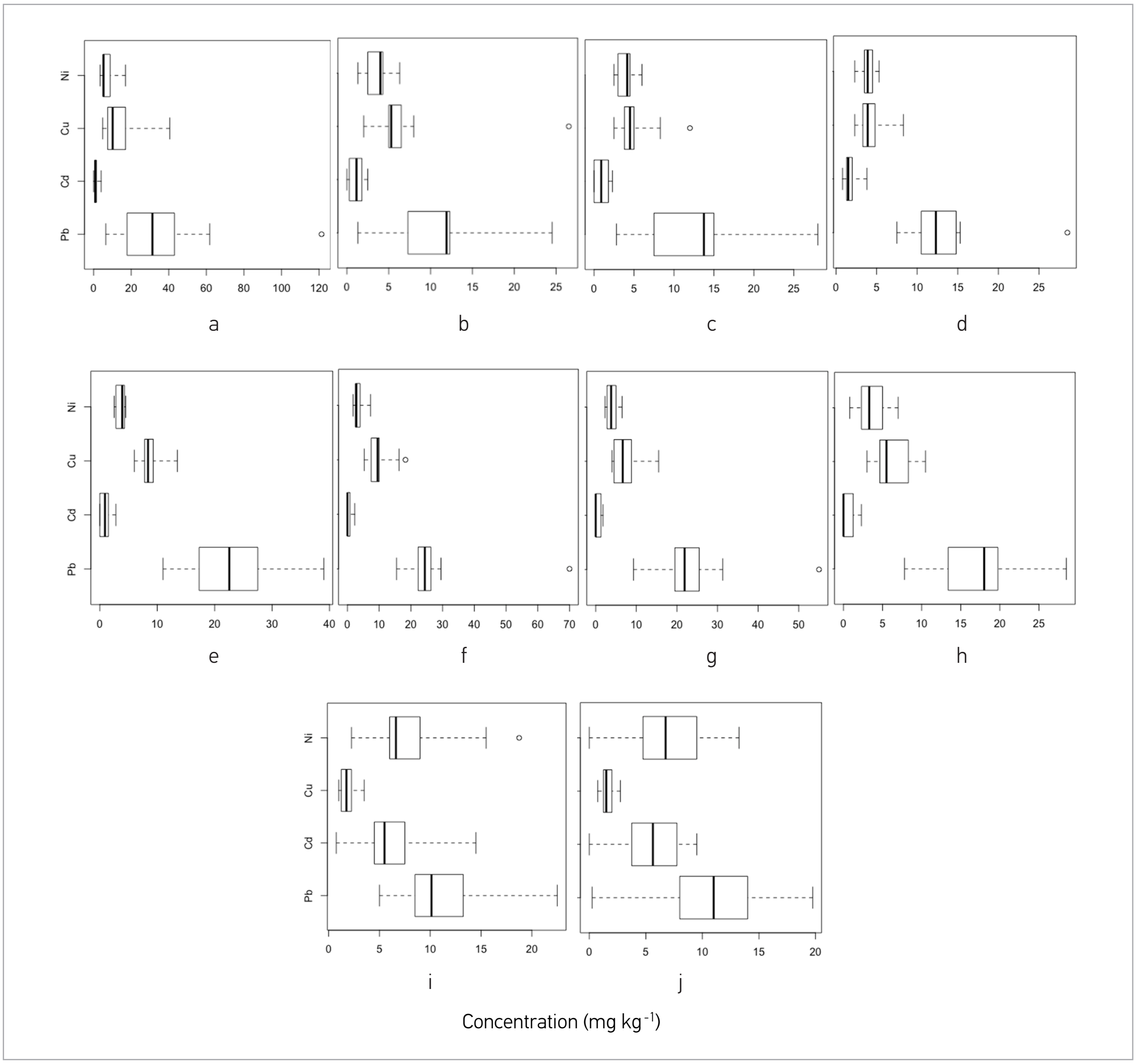
The box plots (Fig. 2 a–h) suggest that heavy metal concentration in soil followed a similar pattern regardless of the study routes. That is, Pb concentration was generally found to be the highest, while Cd concentration was the lowest. Cu and Ni concentrations were found to be approximately the same. Since the study sites are located in a nodal city, which experiences constant flow of traffic, Pb might have been contributed to the RSS by the fuel combustion in the past and accumulated in soil over the years, because leaded fuel was phased out in Nigeria more than a decade ago (UNEP, 2004).

The primary source of Cd could have been the incineration of the wastes in the households, which is a common practice in this city (Abdulrazaq et al., 2015). However, waste incineration is not intensive in comparison with traffic activities. Thus, the Cd concentration in soil was relatively lower. A relatively moderate amount of Cu and Ni in RSS can be attributed to the contribution from vehicular activities including the wear of vehicular components, which is generally a major source of these heavy metals in an urban environment (Škrbić et al., 2012). The variation of heavy metal concentration in soil across the study routes was also investigated. The outliers in the data matrices were first identified using the box plot and removed. The outlier free data matrices were subjected to the Shapiro-Wilk test to assess the normality of the data. The test suggested that the data matrices can be approximated to normality at a 5% significance level (Table S2 in Appendix). Therefore, the study employed parametric statistical techniques such as ANOVA followed by the Tukey's HSD for further analyses.

The results of the ANOVA test for the soil samples across 8 study sites suggested that there was a highly significant difference in the concentrations of Pb [$F(7) = 6.5$, $p < 0.01$], Cu [$F(7) = 6.8$, $p < 0.01$] and Ni [$F(7) = 3.5$, $p < 0.01$] across the study routes. According to the pairwise comparison results using the Tukey's HSD (Fig. S1 in Appendix), there was a statistically significant difference at the 5% level between Pb concentrations in soil from sites OA and AA, OA and FB, OA and GG, AA and TT, and AA and UY. Similarly, Cu concentrations between OA and AA, OA and FB, OA and GG, OA and OT, OA and UT, TT and AA, TT and GG, UY and GG, UT and TT, and UY and UT were significantly different at the 5% level. Furthermore, Ni concentrations in OA were different at the 5% significance

Fig. 2

Box plots showing variations in metal concentrations: (a) Soil - OA, (b) Soil -AA, (c) Soil -UT, (d) Soil -GG, (e) Soil -UY, (f) Soil -TT, (g) Soil -OT, (h) Soil -FB, (i) leaves and (j) barks



level in comparison with that in AA, FB, OT, TT and UY. The differences in Pb, Cu and Ni concentrations between other routes were not significant. In contrast, Cd concentrations in RSS did not have any statistically different variation among study routes [F(7) = 2.0, p > 0.05]. A general trend of increased metal concentrations in Site OA can be attributed to the airport in the study route, resulting in an increased deposition of metals from air and road traffic flow. Though the study site characteristics vary in terms of traffic flow, traffic density, population density and land use, a clear pattern in metal concentration variations in soil between study routes could not be observed suggesting that these influential factors had a compound effect on soil metal concentrations.

Characteristics of heavy metals present in plant leaves and barks

Fig. 2 (i–j) shows the variation in heavy metal concentrations in leaves and barks of roadside native trees. Both leaves and barks exhibit a similar trend for the variation in metal concentrations. Pb concentration in leaves and barks is the highest, while the Cd concentration is the lowest. Ni and Cd concentrations in leaves and barks are moderate. The ANOVA test showed that there is no difference between concentrations of Pb [F (1) =0.039], Cd [F (1) = 0.405], and Ni [F (1) =0.346) in leaves and barks at a significance level of 5% except in Cu [F (1) = 4.1] suggesting that absorbed heavy metal ions are distributed between barks and leaves in a similar proportion. However, the concentration of Cu in leaves was higher than that in barks according to Tukey's HSD. Cu is a micronutrient that is essential for the growth. Hence, a higher amount of Cu could have been allocated to the relatively active leaves in comparison with barks, which are not actively involved in metabolism.

Sources and transport pathways of heavy metals in soil and plants

The PCA and the factor analysis were used to investigate the sources and transport pathways of heavy metals in soil and plants. The data matrix for the analysis consisted of heavy metal concentrations in RSS, leaves and barks. The PCA biplot and factor analysis results are given in Fig. 3 and Table 1, respectively.

Fig. 3

PCA biplot for heavy metal concentrations in soil (S), leaves (L) and barks (B)

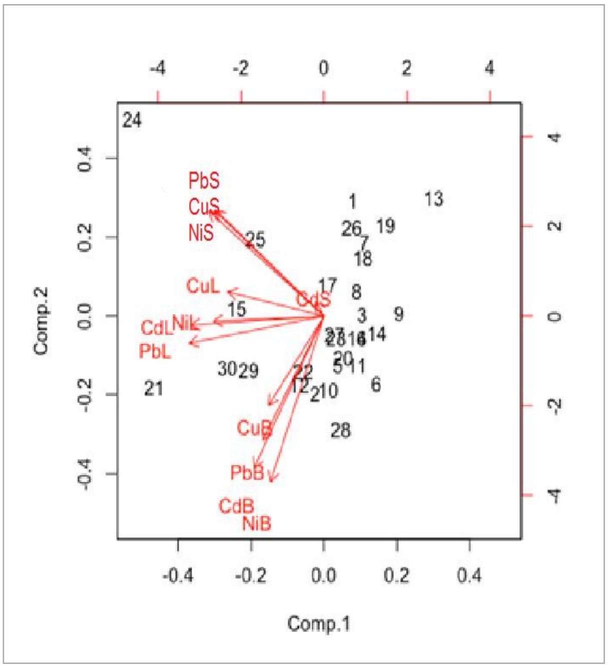


Table 1

Factor analysis results (L – Leaves, B – Barks and S – Soil)

Metal	F1	F2	F3	F4	Communalities
1	2	3	4	5	6
PbL		0.8			0.73
CdL		0.9			0.95
CuL				0.8	0.69
NiL		0.9			0.90
PbB			0.7		0.53
CdB			0.9		0.93
CuB			0.5		0.42
NiB			0.9		0.88
PbS	0.9				0.83
CdS					0.08
CuS	0.9				0.87
NiS	0.9				0.94

Note: Factor loadings ≥ 0.5 are only shown.

The first two principal components explained 51% of the data variance. Hence, factor analysis was used to complement and improve the reliability of the PCA outcomes. The communalities for variables were generally over 0.6, with a mean communality of 0.7 (Table 1). As such, the outcomes of the factor analysis are reliable (Egodawatta et al., 2013). The strong positive correlation between vectors corresponding to Pb, Cu and Ni concentrations in RSS along with their similar size, and their association with factor F1 further confirms that these heavy metals have similar sources. Though Cd is positively correlated with other metals in the PCA biplot, its smaller vector size in the biplot compounded by no association with F1 confirms that Cd comes from a different source. As discussed in Section "Characteristics of heavy metals present in soil", localised garbage incineration was found to be the predominant source of Cd to RSS, while traffic activities were the primary sources of the rest of heavy metals to soil.

Vectors for Pb, Cd and Ni concentrations in leaves are positively correlated in the PCA biplot and associated with factor F2. In contrast, Cu is associated with factor F4. This suggests that the predominant pathway of Pb, Cd and Ni to leaves of native trees is similar, while that of Cu is different. According to the PCA biplot, there is a stronger positive association between Cu present in leaves and soil, suggesting that Cu in leaves came primarily through root uptake. In contrast, foliar uptake could have been the predominant pathway for Pb, Cd and Ni to leaves, though root uptake could also have contributed a significant amount of Pb, Cd and Ni to leaves. Similarly, heavy metals present in barks are strongly correlated with each other in the PCA biplot and associated with F3. The vectors corresponding to metal concentrations in bark are at the right angle with vectors related to metal concentration in soil, according to the biplot. Thus, the transport pathway for metals in bark could have been predominantly through lenticular uptake.

Ecological risk analysis

The pollution and the ecological risk status of the RSS due to trace metal contamination were estimated using the index of geo-accumulation (Igeo) and the potential ecological risk index (PERI). As evident in Table 2, the majority of the routes had the Igeo values for metals between 1 and 2 followed by the range between 0 and 1. An Igeo value between 2 and 3 were also found in a few sites for Pb and Cd. This indicates that the majority of the routes are moderately contaminated with heavy metals ($1 < \text{Igeo} \leq 2$), whereas some are uncontaminated to moderately contaminated ($0 < \text{Igeo} \leq 1$) or moderately to strongly contaminated ($2 < \text{Igeo} \leq 3$) (Muller, 1981). The highest Igeo value for Pb was obtained in OT, TT and OA. Though the primary source of Pb is vehicular traffic, UT and UY, which have the highest traffic density of 4,539 vehicles/hour and 2,244 vehicles/hour, respectively, had relatively lower Igeo values for Pb compared with OT, TT and OA. This further confirms that Pb present in soil is due to the accumulation of Pb emitted by past leaded fuel usage.

Only GG exhibited Igeo values for a moderately to strongly contaminated status for Cd, while AA, UY, OA and UT displayed a moderately contaminated status, and OT and FB were within an uncontaminated to moderately contaminated status. The major contributors to the high Igeo value of Cd at GG may have been the several activities of artisans such as battery repairers, metal welders, painters, and lubricating oil hawkers and various renovation/construction activities in the area, coupled with the fact that two major motor parks are situated in the area. All these activities may have increased the rate of Cd released into the surrounding soil. Five routes, namely AA, OT, TT, UY and OA, were moderately contaminated with Cu, while GG, FB and UT were found to be uncontaminated to moderately contaminated for Cu. All routes displayed ab uncontaminated to moderately contaminated index for Ni except OA (Igeo=1.8, moderately contaminated). This can be associated with the natural concentration of the soil of the area, and the vehicular tear and wear of metal alloys along the route.

The single eco-risk index (Er) indicated that Pb, Cu and Ni have a low risk (Er<40) to the ecosystem, whereas Cd posed a moderate risk (Er=62.4) to a high risk (Er=212.5). The PERI could characterise sensitivity of the local ecosystem to toxic metals and represent an ecological risk resulting from the overall contamination. The PERI values for all heavy metals combined are in the following order: OA > GG > AA > UY > UT > TT > FB > OT, with routes AA, GG, UY, UT and OA constituting a moderate risk ($150 \leq \text{PERI} < 300$), while the

Table 2

Pollution status and potential ecological risk index (PERI) of heavy metals in roadside surface soils

Route	Igeo				Individual Risk Index				PERI	Eco-risk status
	Pb	Cd	Cu	Ni	Er_{Pb}	Er_{Cd}	Er_{Cu}	Er_{Ni}		
1	2	3	4	5	6	7	8	9	10	11
AA	0.8	1.6	1.1	0.9	14.1	138.0	15.9	14.0	182.0	Moderate
OT	2.1	0.5	1.1	0.9	31.4	62.4	16.4	14.5	124.7	Low
GG	1.2	2.2	0.3	0.9	17.3	212.4	9.1	14.5	253.3	Moderate
TT	2.3	0.6	1.5	0.8	36.2	69.0	21.7	13.0	139.9	Low
UY	1.9	1.3	1.4	0.8	28.7	108.0	19.1	13.5	169.3	Moderate
FB	1.5	0.8	0.9	0.6	21.8	79.2	14.1	11.5	126.6	Low
OA	2.7	1.8	2.0	1.8	49.0	152.4	30.6	26.5	258.5	Moderate
UT	1.1	1.3	0.6	1.0	16.3	108.0	13.9	14.5	152.7	Moderate
Average	**1.7**	**1.3**	**1.1**	**1.0**	**26.8**	**116.2**	**17.6**	**15.2**	**Average PERI=175.8**	

Note: Background level for Pb, Cd, Cu and Ni were 3.90±0.78, 0.25±0.09, 2.27±0.41 and 3.90±0.78 mg kg^{-1}. Mean value + SD of metals (5 samples) from the control soil used to compute the Igeo and PERI.

remaining 3 routes posed a low risk (PERI ≤ 150) to the local ecosystem. The average PERI for all the routes indicated a potentially moderate risk (=175.8), and the major contributing factor for the ecosystem health risk was Cd contamination with 66.0% risk contribution. It is not surprising that Cd is the main contributor to the eco-risk, since several studies (e.g., Yisa et al., 2012, Huang et al., 2016) have also identified Cd as a major contributing factor to a high ecological risk in urban soil or roadside dusts. This is mainly due to its high toxicity coefficient (30), corresponding to its long residence time and higher toxicity (Sutherland and Tolosa, 2000).

4. Conclusion

Concentration, source, transport pathway and the eco-risk of Pb, Cd, Cu and Ni in the roadside soils of the nodal city of Ilorin, north-central Nigeria, were investigated in this study. The conclusions of this study are:

- Concentration of Pb was the highest in the roadside soil and plant samples in all the routes, though significant variation existed in the concentrations among the routes.
- Concentration of Cd was the lowest in the roadside soil and plant samples with no statistical difference in the concentrations across the routes investigated.
- Traffic activities were found to be the principal source of Pb, Cu and Ni in the roadside soils, while localised garbage incineration was considered to be the main source of Cd.
- The transport pathway of Cu to the tree leaves was through root uptake, whereas Pb, Cd and Ni were predominantly taken up in the leaves through foliar absorption and the metals present in the barks were predominantly through lenticular uptake.
- Average Igeo for all the routes indicated a moderate contamination for all metals in all the routes.
- The general ecological risk (PERI) of metals in the roadside soils suggested that the pollution level was moderate, though 37.5% of the routes exhibited a low eco-risk.

Conflicts of Interest: None.

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Appendix

Fig. S1

Pairwise comparison of heavy metal concentration variations in soil from 8 study sites

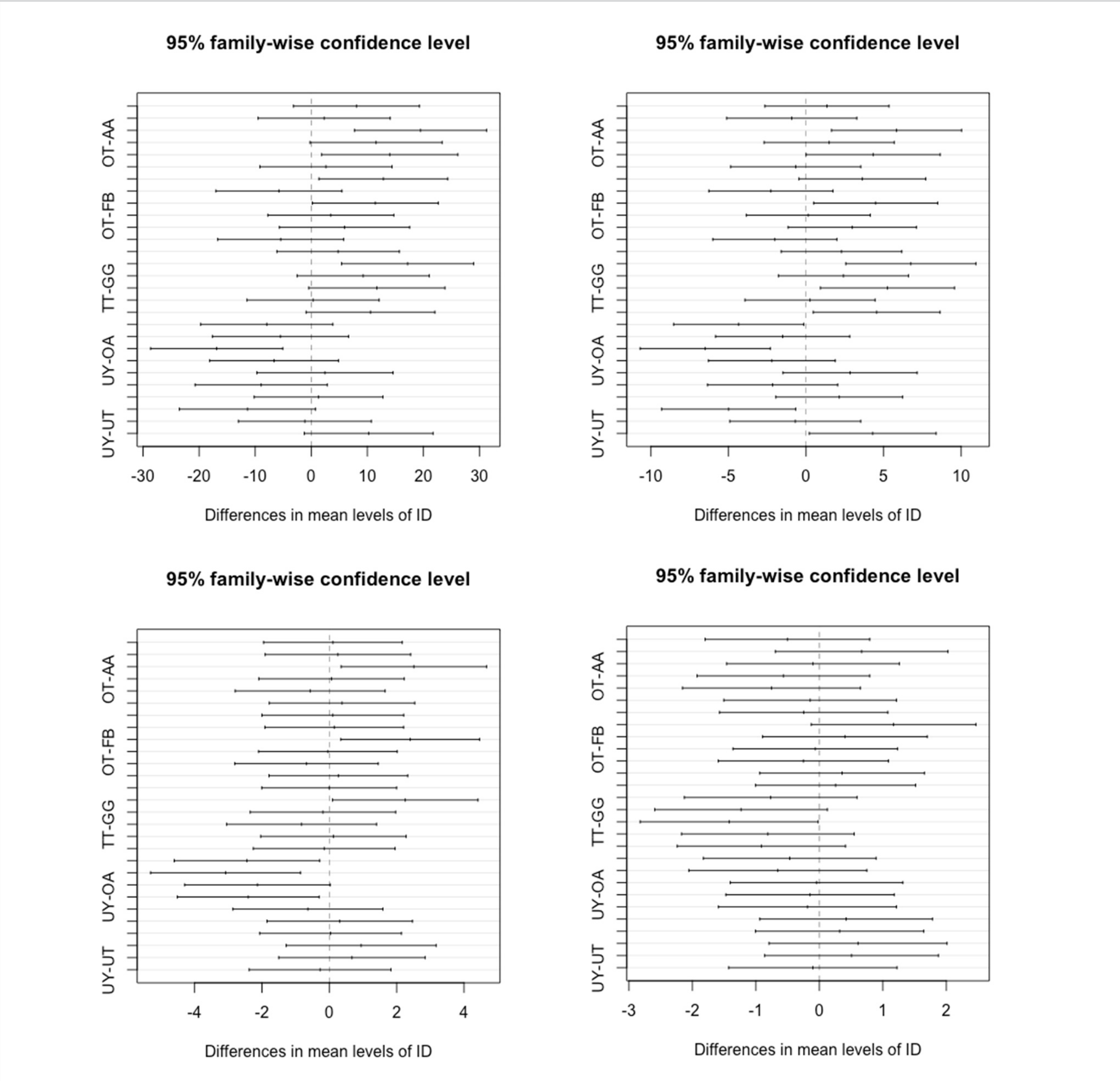

Table S1

Outcomes of the Shapiro-Wilk test

	Test statistic (* shows p values less than 0.05)			
	Pb	Cd	Cu	Ni
1	2	3	4	5
Soil samples				
OA	0.95	0.76*	0.90	0.84
AA	0.94	0.93	0.93	0.95
UT	0.95	0.87	0.83*	0.95
GG	0.97	0.89	0.84	0.98
UY	0.94	0.89	0.87	0.87
TT	0.95	0.68*	0.85	0.92
OT	0.94	0.73*	0.94	0.91
FB	0.93	0.79*	0.94	0.94
Plant samples	0.98	0.96	0.94*	0.94*

Miestų pakelėse besikaupiančių sunkiųjų metalų šaltiniai, patekimo keliai ir keliami ekologiniai pavojai

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Mokslinėje literatūroje analizuojami sunkiųjų metalų šaltiniai ir patekimo būdai, urbanistinių plotų dirvožemiuose tipinėse gyvenvietėse, kartu su susijusiais ekologiniais rizikos veiksniais. Dirvožemio, lapų ir žievės mėginiai buvo surinkti iš įvairių gyvenvietės vietų ir analizuojamos metalo koncentracijos mėginiuose naudojant standartinius metodus. Duomenys buvo analizuojami naudojant ANOVA, pagrindinių komponentų analizė ir faktorių analizė. Rezultatai parodė, kad dirvožemyje daugiausia buvo Pb, Cu ir Ni ten, kur vyksta intensyvus transporto eismas, o Cd dirvožemyje daugiausia buvo rasta po komunalinių atliekų deginimo vietose. Pb, Cd ir Ni junginiai rasti lapų mėginiuose patenka absorbcijos būdu per dirvožemį, šaknys pirminis absorbcijos kelias, kuriuo Cu patenka į lapus. Potenciali ekologinė rizika parodė, kad tiriamajame objekte yra vidutinio sunkumo ekologinė rizika, o Cd yra pagrindinis rizikos veiksnys. Todėl svarbu kontroliuoti Cd šaltinį, kad būtų sumažintas su metalų tarša susijęs ekologinis pavojus aplinkai.

Raktiniai žodžiai: ekologinės rizikos indeksas, eismas, emisijos, augalų absorbcija, sunkieji metalai, urbanizacija.