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Research Paper

Analysis of Selected Heavy Metals Concentrations in Roadside Soils: Adama, Oromia region, Ethiopia

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Abstract: Heavy metals, such as Cu, Zn, Cd and Pb are common pollutants in urban environments mainly due to high traffic emissions. The present work was aimed to determine the level of Cu, Zn, Cd and Pb concentrations in high traffic density roadside soils of Adama city, Oromia region, Ethiopia. The soil samples were collected from three roadside soils (both sides) of the selected sites symmetrically at distances in the range of 0-50, 50-100 and 100-300 meters from the main road of the city. All the samples were analyzed for the concentrations of Cu, Zn, Cd and Pb using flame atomic absorption spectrometric (FAAS) method. The levels of Cu, Zn and Pb in the roadside soils were found to vary from 20.49 to 45.01, 34.22 to 46.71 and 47.84 to 69.28 mg kg⁻¹, respectively. The level of Cd was found below the detection limit. The levels of heavy metals in both sides of the roadside soils exhibited a significant decrease with the increasing distance from the road and a positive correlation was found between the metal concentrations. The highest level of Pb, Cu, and Zn in the soil samples was found close to the main road and observed positive correlation among the metal concentrations. The indicated pollution due to vehicles emission will be a potential threat for the city and expect strict measures to reduce harmful emissions.

Keywords: Heavy metals, Urban soil, Vehicle emission, Traffic load, Flame atomic absorption.

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Introduction

Pollution of environment by heavy metals released during different operations involving industries, domestic, commercial and vehicles is a worldwide environmental and public health concern¹⁻². Vehicle emissions were the major sources of heavy metals such as Cu, Cd, Pb and Zn in urban environment and the increment of traffic activities substantially contributes to the accumulations of these metals in roadside environments. As a result, they have been the focus of urban pollution and mitigation studies in recent years³⁻⁶.

Metals are non-biodegradable and therefore, persist for long periods in aquatic as well as terrestrial environments and caused the function of the human body impaired and causing disease⁷⁻¹⁰. The toxic effects of these heavy metals have been well-studied¹¹⁻¹⁷. These contaminants can easily impact people residing within the vicinity of the roads via suspended

dust or direct contact. Moreover, if there are farmlands within the scope that the contaminant can reach, they may enter the food chain as a result of their uptake by edible plant, thus causes series health risks¹⁸. High dosage of Cu can cause health problems such as anemia, liver and kidney damage and stomach and intestinal irritation. A very high level of Zn can damage the pancreas and disturb the protein metabolism. On the other hand, Cd and Pb, even extremely low doses cause serious diseases in human beings. For example, Cd exposure may give rise to renal, pulmonary, hepatic and reproductive effects, as well as cancer. Chronic and acute exposure of Pb is particularly toxic to the brain, kidneys, reproductive system and cardiovascular system and increase of inflammatory responses¹⁹.

Roadside soils are the major reservoirs of traffic-related heavy metals and determining their levels in

roadside soils of urban areas are considered as pointers of heavy metal pollution emanated from increasing vehicular traffic²⁰⁻²². Heavy metal emissions released from vehicles are mainly related to fuels combustion, lubricating oil consumption, tire wear, break wear, road abrasion etc. Cd emission is mainly from lubricating oil and tire wear, Zn comes from tire wear and galvanized parts such as fuel tanks^{23,24}. Break wear is the most important sources for Cu and Pb^{25,26}. Moreover, Pb is also detected in exhaust gas and worn metal alloys in the engine²⁷⁻²⁹.

Like any other city, Adama city developed over the years in a progress of industrialization, increasing population and traffic density. According to Central Statistical Agency C.S.A (2008) current population of Adama city is estimated to be 402,000 with a growth rate of 4.8% per annum. Moreover, the average annual flow of tourists to the city over the last four years is 836,822. Additionally, the city is situated along the road that connects Addis Ababa with Djibouti. A large number of trucks use this same route to travel to and from the seaports of Djibouti. This makes Adama to have busy traffic activities. Moreover, the city has strategic location and geographical proximity to various agro-processing industries. However, there was no previous study carried out to understand the levels of heavy metal on the roadside soils in Adama city related to high vehicles densities.

The levels of heavy metals in road side soils is unique for every region and vary from time to time and depends on the distance from the main road, traffic volume and pattern, road and roadside terrain, and wind direction³⁰⁻³¹. The earlier study³⁰ also reported that concentrations of heavy metals decrease with increasing roadside distance. Generally, for most heavy metals influential roadside distance is 10-50 m from either side of road edges. The previous studies³²⁻³⁴ reported that elevated concentrations of trace metals specifically Pb even as far as 320 m from the roadside.

However, other research results showed that accumulated heavy metals can be transported to different environmental components and it can also be transported into the roadside soils by atmospheric precipitation or road runoff, and origins are difficult to distinguish so that the spatial distribution patterns of heavy metals in roadside soils were not always significantly correlated with the roadside distance³⁵⁻³⁸. Therefore, the focus of this study was to investigate the effect of vehicular emission on the level of selected heavy metals (Pb, Cu, Zn and Cd) in relation with the distance from the road side topsoil of Adama city, Ethiopia.

Material and Methods

Description of the study area

Adama is a city in central Ethiopia and Special Zone of Oromia. It is located at [8.54°N 39.27°E](#) at an elevation of 1712 meters, 99 km southeast of Addis Ababa. The city sits between the base of an escarpment to the west, and the Great Rift Valley to the east. According to the data from Ethiopian Road Authority middle traffic volume of per day 18,000-20,000 vehicles can transfer through Adam town. The vehicles are composed of 35% motorcycles and bagages, 50% cars, 10% trucks and 5% others.

Chemicals and Reagents

All chemicals of high purity analytical grade reagents were employed; HNO₃ (spectrosol BDH, England) and HCl (37%, sigma Aldrich, Germany) were used for both extraction and acid digestion procedures. sulphuric acid, and buffer were used to determine soil PH. Standard Cu, Cd, Zn and Pb solutions (1000 ppm) used for instrumental calibration was obtained from (EC- EMB45053 BDH, Belgium).

Instrumentation

Digital analytical balance (Model E11140, Switzerland) was used for all measurements of samples and chemicals weight. Atomic Absorption Spectrophotometer (model ZEEnit 700p, analytik Jena, Germany) was used to determine the levels of Cu, Zn, Cd and Pb. A potentiometric digital pH meter (JANWAY 3345 pH meter, Germany) was used to determine the pH of soil samples.

Sample collection and treatment

The roadside soils were collected from three selected sites along the main roads in the Adama city (Figure 1). The division of the city main road was only for convenience of handling the large area of the city. Each site was divided in to 3 sampling distance and defined as the roadside distance perpendicular to the road edge. The sampling distances to the road edge are designed as 0-50 m, 50-100 m and 100-300 m, considering previous report that most metals influential roadside distances are 10-100 m. The samples were collected from both sides of road.

A total of 12 composite soil samples (6 samples from each side) were collected from each site in 2 weeks of interval on mid-November, 2016. Composite samples of 1 kg of soil were taken from 0-5cm depth using a hand driven stainless steel shovel without removing the surface soil. The samples were taken from each hole by 'zigzag- shape' which was then pooled together to give a composite sample. Finally, the samples were packed in polyethylene bags and brought to the laboratory. In the laboratory, all samples were oven dried at 105°C to constant weight for 6 hours. The samples were then

ground using mortar and pestle and sieved through 2.0 mm mesh and kept packed until further analysis³⁹.

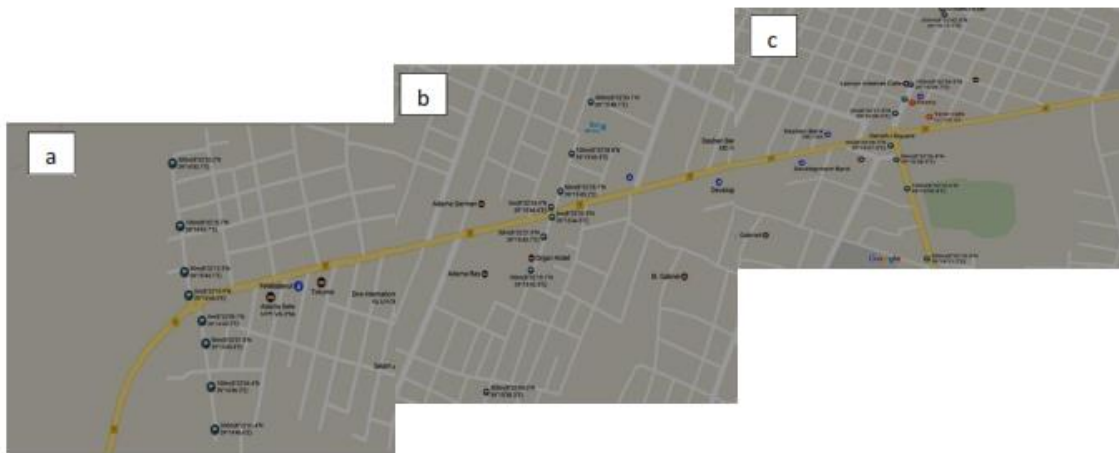


Figure 1: The three sampling sites a) Awra Godana site, b) Dembela site, and c) Derartu site with sampling distances to the road edge designed as 0-50 m, 50-100 m and 100-300 m from both left and right side of the road

Chemical analysis

pH of the samples were determined in a 1:2.5 soil/water suspension prepared by mixing 20 g soil sample and 50 ml distilled- deionized water in a 100 ml polythene wide-mouth type bottle. Then, the mixtures were placed on shaker for 2 hours allowed to stand for about 5 minutes for the soil particles to settle.

The heavy metals analysis was carried out after sample digestion following the established method⁴⁰. About 1.0 g of the sieved sample was reacted with 15 ml of concentrated HCl, 5 ml of HNO₃ and 0.3 H₂SO₄. The samples were then filtered through Whatman No. 42 filter paper, washed, and made up to 50 mL with deionized water. The sample solution was then aspirated into the FAAS at intervals and then analyzed for the presence of Pb, Cu, Zn and Cd.

Recovery test

To assess and validate the accuracy of the analytical procedures used, spiking experiments were done and the percent recoveries were calculated. These procedures were done by digesting 1.0 g of soil samples spiked with standard solutions of the metals (Pb, Cu, Zn and Cd). Then, the sample was mixed well and subjected to acid mixtures of 15 ml of concentrated HCl, 5 ml of HNO₃ and 0.3 H₂SO₄. The digested sample was filtered and transferred into 50 mL volumetric flask and diluted to the mark with deionized water. Each recovery test for soil samples was performed in triplicate. Finally, the solution was analyzed with AAS. The percentage recovery of the sample was calculated as:

$$\% \text{ Recovery} = \frac{\text{Amount after spike} - \text{amount before spike}}{\text{amount added}} \times 100\%$$

The percentage recovery of metals under investigation was Pb= 99%, Cd = 96%, Cu = 98% and Zn = 97%, which confirms the robustness of the extraction procedure using in this investigation.

Statistical Analysis

Means and standard deviations were calculated for all the data and means were compared among treatments (samples taken from three different distances to the road edge) by applying one way ANOVA. Differences between the means were considered to be statistically significant at $p \leq 0.05$. Correlation between heavy metals was calculated by Pearson correlation test at 0.05 significant levels. All statistics were computed using data analysis and graphic software (SPSS, version 20.0)

Results and Discussion

pH of the soil samples

The pH value for different distances from the roadside edges were found to be with the range of 7.8 – 8.0, indicated it is slightly alkaline (Table 1). The result of this study reported the pH of the roadside soil dust of the studied sites ranged from 7.5-8.0. The result implied that the observed pH of the soil solution might lead to low mobility of heavy metals so that persist in the soil for long time⁴¹. Complexions with organic matter, chemisorption on oxides and silicate clays, and precipitation as carbonates, hydroxide or phosphate are all favored at higher pH and may decrease solubility of heavy metals⁴².

Table 1: The mean pH value with standard deviations for the right and left road side soil taken at different distance from roadside edges

Depth	Distance	pH	
		Right	Left
0-5cm	0-50 m	8.03±0.05	7.93±0.06
	50-100 m	7.92±0.09	7.94±0.03
	100-300 m	7.90±0.03	7.77±0.05

Level of heavy metals in the Road side soil as affected by distance

The ANOVA result of level of heavy metals in the soil sample at different distance of the roadside edges was described in Table 2. ANOVA result shows, on left side of the road there is significance difference with respect to the level of Pb and Zn between and within group means (at $p < 0.05$), whereas ANOVA analysis with respect to Cu shows there was no significant difference between and within group means (at $p < 0.05$). On the right side of the road ANOVA result

shows there was significant difference (at $p < 0.05$) between and within group means for all metals (Pb, Zn, and Cu).

The most abundant heavy metal is Pb compared to the other heavy metals in both sides of surface soil (Table 2). The Cu concentrations are lower due to the continuous removal of heavy metals by the trees grown in this area and also due to leaching of heavy metals into the deeper layer of the soil. But different abundances were recorded on the two sides of roads. In left side soil the heavy metal was found in the order of $Pb > Zn > Cu > Cd$ (Table 2). On the otherhand, in the right side soil the levels metals for every distance from the road side edges is in the order of $Pb > Cu > Zn > Cd$.

The concentration of Cd is below detection limit of the instrument. The difference in the order of risk level with respect to abundance of Zn and Cu might be because of the presence of different trees and plants in right side of road that up taken Zn preferably through their root, stem and leaves.

Table 2: ANOVA of the level of heavy metals concentration (mg/kg) in surface soil (0-5 cm) collected at different distances from road side edges

Left side of road						
		Sum of Squares	df	Mean Square	F	Sig.
Lead	Between Groups	737.373	2	368.687	8.441	.004
	Within Groups	655.200	15	43.680		
	Total	1392.574	17			
Zinc	Between Groups	324.529	2	162.264	15.247	.000
	Within Groups	159.640	15	10.643		
	Total	484.169	17			
Copper	Between Groups	219.202	2	109.601	1.761	.206
	Within Groups	933.454	15	62.230		
	Total	1152.656	17			
Right side of road						
		Sum of Squares	df	Mean Square	. F	Sig
Lead	Between Groups	1351.294	2	675.647	8.663	.003
	Within Groups	1169.946	15	77.996		
	Total	2521.240	17			
Zinc	Between Groups	101.397	2	50.698	4.524	.029
	Within Groups	168.111	15	11.207		
	Total	269.508	17			
Copper	Between Groups	1729.501	2	864.751	62.318	.000
	Within Groups	208.145	15	13.876		
	Total	1937.647	17			

Table 3: Mean concentrations of Pb, Zn, and Cu levels (mg/kg) in surface soil (0-5 cm) with the factor of road distance from road side edges on both side of road

Parameter	Distance	Level of heavy metals	
		Left	Right
Pb	0-50m	61.98a	90.51a
	50-100m	61.06a	84.93a
	100-300m	47.97b	69.99b
Zn	0-50m	46.06a	37.53a
	50-100m	42.75a	34.76ab
	100-300m	35.86b	31.72b
Cu	0-50m	36.11a	56.37a
	50-100m	28.88a	41.60b
	100-300m	28.55a	32.59c
Cd	0-50m	Nd	Nd
	50-100m	Nd	Nd
	100-300m	Nd	Nd

NB: Only significant means (at $P \leq 0.05$) were indicated using different letters within a column (a, b), Nd refers to not detectable

Lead

A decrease in the mean concentration of Pb was observed as sampling distance from the roadside edges increases from 0-50 m to 100-300 m (Table 3). However, the decrease in the mean value of Pb were not significant (at $P \leq 0.05$) with distance in 0-50 and 50-100 m from the road edge at the two locations along main road (left) and (right). The result is in agreement with the findings of earlier work⁴³, which indicated that there was a marked decrease in the Pb content within a distance of few meters from the road. Moreover, another reported work⁴⁴ revealed that Pb content amounted to 83.70 mg/kg at a distance of 50 m and 22.86 mg/kg at a distance of 100 m from the road side. The Pb levels observed in this study were significantly higher than those reported by two researchers^{45,46}. However, the result of this study is lower than the WHO maximum permissible limit of 300 mg/kg with respect to Pb on surface soil. The result indicated low quality fuel and exhaust gases of petrol engines may be the major causes of the increased lead contamination in the roadside soil^{47,48}.

Zinc

There was a decrease in the mean concentration of Zn as sampling distance from the roadside edges increases from 0-50 m to 100-300 m (Table 3). The highest Zn level were observed in 0-50 m (46.06 mg/kg, 37.5 mg/kg) and the lowest Zn level was observed at 100-300 m (35.86 mg/kg, 31.7 mg/kg) distance from the road edge of both left and right side. However, the decrease in the mean value of Zn was not significant

(at $P \leq 0.05$) with distance in 0-50 and 50-100 m from the road edge at the two locations along main road (left) and (right) (Table 3). Similar trend with a decrease in the Zn content within a distance of few meters (0-10 m) and (10-50 m) from the road edge was reported⁴⁹. Vehicular source such as corrosion of vehicle parts, tire wears, leaks of engine oil, and/or exhaust fumes may be the potential sources of the observed Zn concentration in the study area. The mean concentration of zinc in the study area is lower than WHO limit of 200 mg/kg of soil.

Copper

As shown in Table 3, the mean values of Cu were found to decrease significantly as sampling distance from the roadside edges increase from 0-50 m to 100-300 m on the right side of the road. Whereas the decrease on the left side was not significantly different with sampling distance, indicates dispersion of pollutants from the point of source. The range of concentrations of copper observed was also considerably lower than the value of 114 mg/kg set by the WHO regulatory standard for copper in soil.

The Pb and Cu level on the right side of the road is higher than the left side (Table 3). The highest mean values found on right side of road might be related to the existence of tire shops, garages, banks and hotels where vehicles stopped and also portal and exit points to different cities. For instance, brake wear from vehicles are reported to have an important source of Cu and Pb concentrations in roadside soils.

Comparisons of Pb, Zn and Cu level of the Road side soil with distance and other studies

As shown in Figure 2, the highest average concentrations of all the metals (Pb, Zn, and Cu) were found in a distance of 0-50 m close to the road edge, while the lowest concentrations were detected at 100-300 m away from the roadside.

The level of Pb, Zn, and Cu in a surface soil at distance from the road edge was in the order of 0-50m > 50-100m > 100-300m. The highest levels were found on the 0-50 m (Figure 2). The lowest heavy metal levels were found on the 100-300 m distance from the road edges for all the metals. This result is in agreement with the earlier findings⁴⁹ which stated that concentrations of heavy metals in soil samples collected from roadside appeared to be in the spatial pattern of distribution with the order of 0 m > 50 m > 100 m > 1000 m. Moreover, it was reported⁵⁰ that highest concentration of metals near the roadside and on the surface of soil while the content of trace elements in soil decreased with the increase in distance from roadside. This indicates that the concentration of heavy metals in soil along the main road have been shown to decrease rapidly with distance from the roadside and

with decreasing traffic density. Moreover, the result confirmed that vehicular emission played a significant role in the deposition of heavy metals on surface soil of Adama city road.

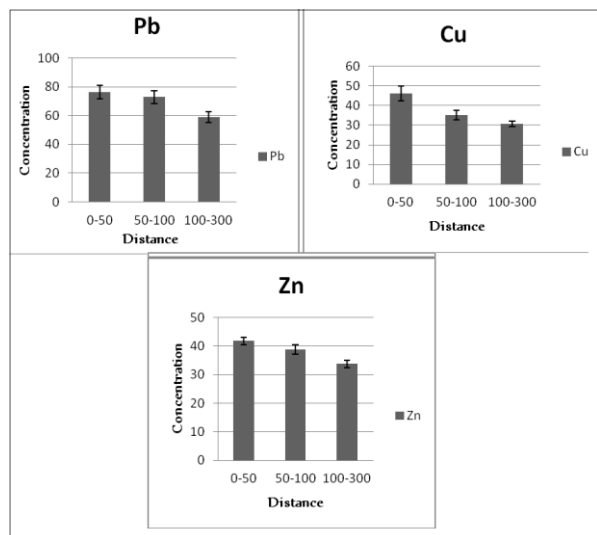


Figure 2: Mean concentration (mg/kg) of Pb, Zn and Cu in different road distance (bars indicate standard error of mean)

The level of Pb, Zn, and Cu in a surface soil at distance from the road edge was in the order of 0-50m > 50-100m > 100-300m. The highest levels were found on the 0-50 m (Figure 2). The lowest heavy metal levels were found on the 100-300 m distance from the road edges for all the metals. This result is in agreement with the earlier findings⁴⁹ which stated that concentrations of heavy metals in soil samples collected from roadside appeared to be in the spatial pattern of distribution with the order of 0 m>50 m>100 m>1000 m. Moreover, it was reported⁵⁰ that highest concentration of metals near the roadside and on the surface of soil while the content of trace elements in soil decreased with the increase in distance from roadside. This indicates that the concentration of heavy metals in soil along the main road have been shown to decrease rapidly with distance from the roadside and with decreasing traffic density. Moreover, the result confirmed that vehicular emission played a significant role in the deposition of heavy metals on surface soil of Adama city road.

Compared to the levels of heavy metals reported in different locations, Adama traffic volume is categorized within the medium traffic volume area (OMI) ~1000 vehicle per day, but the level of Cu found in this study is higher than the range (2.02 to 11.03 mg/kg) reported in OMI⁵¹. Moreover, the Cu level in this study is lower than those found in French Major Highways⁵² (14.0 mg/kg), Anand City, India⁵³

(20.04 mg/kg) and Beijing, china 29.70 mg/kg (Chen et al.,2010). The level of Pb content found in this study is also higher than Anand city, India (3.77 mg/kg), and Beijing, China (35.40 mg/kg). Whereas the Zn level found in Adama city is lower than the reported values of the aforementioned city.

Table 4: Pearson’s correlation between concentrations of Cu, Zn and Pb on the left and right side road soil

Left side of the road			
	Pb	Zn	Cu
Pb	1		
Zn	.876**	1	
Cu	.573*	.487*	1
Right side of the road			
	Pb	Zn	Cu
Pb	1		
Zn	.211	1	
Cu	.319	.654**	1

** . Correlation is significant at the 0.01 level (2-tailed)
 * . Correlation is significant at the 0.05 level (2-tailed).

As shown in Table 4, Heavy metal concentrations showed significantly positive correlation with each other on the left side road at the 0.01 level and at the 0.05 level. On the right side of the road significantly positive correlation was observed between Zn and Cu (0.654) only. This significantly positive correlation between themselves reflects the association between elements and the homology of their pollution sources. This implies road traffic is the most probable source of these metals since there are no other sources such as industries and farmlands in the study areas. The observed positive correlation between Zn and Cu on the right side road related to the existence of tire shops, garages, banks and hotels where vehicles stopped for maintenance and contribute for non-exhaust emissions. Whereas the distribution of Pb confirms its availability has direct relationship with tailpipe emissions (exhaust emissions) of high traffic density⁵⁴.

Conclusion

The study confirms that the highest levels of Pb, Cu and Zn were detected in Adama roadside soil samples collected from the border zone (1m-50m) of the roads side edges and the value decreased with road distance. Decreasing the level of heavy metals in the soil samples with distance indicate the possible cause of contamination by vehicular emissions and are significantly affected by vehicles traffic volume. The most abundant heavy metal is Pb compared to the other heavy metals in both side of surface soil but this

concentration is lower than the permissible standard limit set by world health organization. The highest level of Pb, Cu, and Zn in the soil samples close to the main roadside as compared to other reference sites and observed positive correlation among the metal concentrations indicated pollution due to vehicles emission will be a potential threat for the city unless frequent monitoring on the level of heavy metals and strict measures were taken accordingly.

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