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STATUS OF SELECTED HEAVY METALS DISPERSION FROM TOP SOIL IN AND AROUND AUTOMOBILE WORKSHOP AREAS IN ZANZIBAR MUNICIPALITY, TANZANIA

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ABSTRACT

The distributions of selected heavy metals in top soil in areas with and without automobile workshops in Zanzibar municipality were investigated. Sampling was done during dry and rainy seasons. The samples were digested with aqua regia (HCl and HNO_3, 3:1) and analysed by Atomic Absorption Spectrophotometry. The concentrations of zinc, copper and lead in the samples ranged 0.27–419.2, 0.10–167 and 0.02–271 mg/kg dry weight, respectively. The concentrations were higher in areas with automobile workshops than those without. The concentrations of the heavy metals were highest at point sources and decreased with increase in distance during the dry season. The concentrations increased with increase in distance during the rainy season, indicating that the dispersion was highly influenced by runoff water. There were no significant differences in the concentrations of the metals between the sampling periods. The zinc concentrations in 51.11% of the samples exceeded the Tanzania Bureau of Standards (TBS) permissible limit (150 mg/kg). The concentrations of copper in 40% of the samples from the point sources and the concentrations of lead in 18% of the samples exceeded the WHO permissible limit (100 mg/kg). Control measures should be taken to stop and clean-up the workshops in residential areas.

Keywords: *Heavy metals, Soil, Automobile, Tanzania*

INTRODUCTION

Heavy metals are among the important toxic pollutants that spread in the human residential and occupational settings as well as in other environmental spheres including air, water and soil. The common pronounced sources of heavy metals pollution in the environment include wastewater from industries, deposited wastes, mineral exploration and extraction, urbanization and vehicles (automobiles) which are all termed as anthropogenic pollutants (Cheng 2003, Achi *et al*. 2011, Warmate *et al*. 2011, Opaluwa *et al*. 2012). Automobile workshops (garages) may pollute the environment through various ways. Metals are often used as minor additives to oil and various auto-lubricants. Used engine oil also contains metals like copper, aluminium, chromium, iron, lead, manganese, nickel and tin which are in the forms of tinny fragments worn out from engine parts (Warmate *et al*. 2011, Ikhajiagbe *et al*. 2013). Engine oil from automobiles may end up into the environment close to, as well as far from, the workshops. Different agents such as rain water can take these metals to water bodies such as oceans, rivers and lakes (Achi *et al*. 2011, Warmate *et al*. 2011). By this scenario, heavy metals are not only a threat to terrestrial organisms but also to aquatic organisms. Studies have shown that different environmental components are contaminated with heavy metals which in turn lead to instability, disorder, harm and discomfort to the physical systems and

organisms (Gulati *et al*. 2010, Dospatliev *et al*. 2012, Ikhajiagbe *et al*. 2013). Even though some of the heavy metals are beneficial to plants and animals at certain levels, they are potentially toxic to their health in certain forms or at high concentrations. If released in the environment, heavy metals accumulate in the soil and sediments which are then absorbed and transferred through the food chains to higher trophic levels (Cheng 2003, Begum *et al*. 2009).

In Zanzibar municipality, there are open automobile workshops very close to or within the peoples' residential areas. The automobile workshops provide services such as replacement of used engine and lubricating oils, repair of fuel tanks, repair of braking and clutch systems, scraping of old vehicle body coats and spray paintings. This suggests that the problems of releasing and dispersal of heavy metals from garage wastes to the environment and pollution around automobile workshops might be occurring. However, the status and knowledge of the pollution level of heavy metals in these areas was previously uninvestigated. Therefore, this study was carried out to establish the levels of environmental contamination by this source in the areas. Three heavy metals (zinc, copper and lead) were selected as they were considered to be important contaminants in relation to the automobile workshops.

MATERIALS AND METHODS

Study areas

The sampling sites for this study were categorized into two groups. One group was located in areas with open automobile workshops which are found within the residential areas while the second group was located in the areas that were far away from the automobile workshops in Zanzibar municipality. The selected automobile workshops were located at Darajani, Kidutani/Kibokoni, Lumumba, Kijangwani and Kikwajuni (Figure 1). These areas were selected due to the fact that the automobile workshops have been there for a long time (>10 years) and the soil acts as long term sink for heavy metals which can remain there for many years depending on the nature of the heavy metals and type and characteristics of the soil. The automobile workshops were also very close to peoples' residences, schools and playgrounds. On the other hand, Makadara and Mtoni Mazrui were selected as areas for comparison since they did not host any automobile workshop.

Sample collection

The sampling was conducted in mid February 2013 (during dry season) and repeated in mid May 2013 (during rainy season). The samples were collected using clean plastic spoons after being dug using stainless shovel and stored in plastic containers (polyethylene bags). Samples were collected from points located at the automobile workshops (point sources) and from two opposite directions up slope (up gradient or parts sloping upward) and down slope (down gradient or parts sloping downward) at 10 m intervals up to 70 m (in some areas) and at 10 to 15 cm depths from the surface. The sampling points were located depending on the sizes of the workshop areas and distance from the residential areas, play grounds, schools and other settings. A total of 108 soil samples were collected and they comprised 54 samples from each season. The samples were transported to the Chemistry Department, University of Dar es Salaam for processing and analysis.



Figure 1: Map showing the study areas in Zanzibar Municipality (Source: Google map, modified by the researchers)

Sample preparation and digestion

Soil samples were oven-dried at 110 ^{o}C for 2 h, cooled in desiccators for 20 min and ground using mortar and pestle to fairly uniform sizes (Warmate *et al*. 2011). Soil samples were digested using aqua regia prepared by mixing concentrated hydrochloric acid and nitric acid in the ratio of 3:1. Aqua regia (15 mL) was added into the flask that contained 3 g of the sample and heated on a hot plate for 2 h (Sastre *et al*. 2002). The mixture was cooled to room

temperature, then filtered through filter paper into a 100-mL volumetric flask and diluted with distilled water up to the mark (Opaluwa *et al*. 2012). The extract was poured into a clean polyethylene bottle ready for analysis.

Analytical quality assurance

All the chemicals used were of analytical grade and good to high purity. The chemicals used included HNO_3 AR (69-71%, HOPKIN & Williams Ltd, England), HCl AR (35-38%, LOBA CHEMIE Pvt, Mumbai, India) and metal standards (≥ 99% purity, BDH Chemicals Ltd, Poole, England). The calibration standard solutions were prepared by dilution of the stock metal solutions and their concentrations ranged from 0.25 to 15 mg/L. The glassware were rinsed with 10% (w/v) HNO_3 solution, then washed with distilled water and dried in the oven. Reagent blanks were prepared using aqua regia (15 mL) with each batch of the digestion, treated and analysed in the same way as the samples to check for background contamination. Recovery and precision determinations were performed using a standard reference material (SRM) (BCSS-1; marine sediment manufactured by the National Research Council, Canada). Subsamples of the material (3 g each) were processed and analysed using the same procedures as for the soil samples. The values obtained were evaluated for agreement with the certified values. The mean concentrations of the metals obtained from the SRM were used to calculate the precision and accuracy of the analytical method used in this study. Satisfactory precision and accuracy were required to be within ≤ 20% relative standard deviation (RSD) and 80 to 120% recovery, respectively for all the elements (Chen and Ma 2001). The detection limits for the metals were established based on a 3:1 signal to noise ratio. The results from the blanks were all below the detection limits. The results from the standard reference material (sediment BCSS-1) were as presented in Table 1. Both the accuracy and precision for the concentrations of zinc, copper and lead were found to be within the acceptable limits with recovery (accuracy) ranging from 107.6 to 111% and precision of 0.78 to 2%. The detection limits for zinc, copper and lead were 0.0008, 0.0002 and 0.0005 mg/kg, respectively.

Table 1: Results from the analyses of standard reference material (sediment BCSS-1)

Metal	Certified value (μg/g)[a](NRCC 1995)	Measured values (μg/g)[b]	Accuracy (Recovery %)	Precision (% RSD)
Zinc	119 ± 12	128 ± 1.0	107.6	0.78
Copper	18.5 ± 2.7	20 ± 0.4	108.1	2.00
Lead	22.7 ± 3.4	25.2 ± 0.2	111.0	0.79

[a]Mean ± standard deviation; [b]Mean of two values ± standard deviation

Heavy metal analysis

The digested samples were analysed for zinc, copper and lead using an Atomic Absorption Spectrometer (Thermo Scientific iCE 3000 AAS) employing flame atomization techniques at the Chemistry Department, University of Dar es Salaam. Lamps containing zinc, lead and copper were used. Wavelengths of the monochromator used were 213, 328.8 and 217.0 nm for zinc, copper and lead, respectively. Calibration curves were prepared by running several standards of the metals under the same conditions as the samples. The concentrations of the metals in the sample solutions were determined using

the standard calibration curves. Average values of three replicates were taken for each determination.

Data analysis

The data were statistically analysed using SPSS version 16 and the GraphPad InStat software (Motulsky 1998). Paired *t*-test was used to compare the concentrations of the metals in the soil samples between the two sampling periods. One way Analysis of Variances (ANOVA) and Tukey post-test were also used to compare the concentrations of the metals among the sites with automobile workshops.

RESULTS AND DISCUSSION

Concentrations of heavy metals in soil

The concentrations of the heavy metals determined in the soil samples collected from areas with automobile workshops are summarized in Table 2 and the concentrations of the heavy metals in samples collected in areas without automobile workshops are presented in Table 3.

Table 2: Concentrations of heavy metals in soil samples in areas with automobile workshops (mg/kg dry weight)

Sites and Sampling points	Category	Zinc		Copper		Lead	
		Feb	May	Feb	May	Feb	May
Darajani; 6 points (n = 12), 0–30 m, up & down slope	Max	291.10	349.30	17.60	8.23	223.0	110.13
	Min	131.80	125.20	0.21	0.16	38.70	27.40
	Mean ± SD	192.00 ± 59.49	191.34 ± 83.15	7.65 ± 7.53	4.23 ± 3.33	89.0 ± 67.54	66.70 ± 30.89
Kidutani; 8 points (n = 16), 0–40 m, down slope	Max	286.80	216.73	108.70	66.30	143.0	177.12
	Min	99.51	99.13	15.41	11.03	39.90	29.74
	Mean ± SD	189.80 ± 62.32	161.50 ± 46.93	51.70 ± 42.37	30.52 ± 18.46	81.10 ± 32.27	85.20 ± 51.96
Kikwajuni; 11 points (n = 22), 0–70 m, up & down slope	Max	419.20	316.00	103.10	74.54	154.70	262.00
	Min	100.60	83.90	8.09	6.20	45.30	35.51
	Mean ± SD	189.45 ± 94.50	185.00 ± 70.34	20.30 ± 27.72	22.60 ± 19.86	81.80 ± 37.40	103.60 ± 61.72
Kijangwani; 10 points (n = 20), 0–70 m, up & down slope	Max	328.90	315.90	167.00	63.42	71.20	111.53
	Min	67.51	52.90	20.30	2.02	7.20	0.02
	Mean ± SD	192.60 ± 90.35	157.22 ± 78.27	51.04 ± 43.55	23.30 ± 19.53	33.83 ± 19.36	32.54 ± 35.77
Lumumba; 10 points (n = 20), 0–50 m, up & down slope	Max	136.00	212.40	34.32	106.00	135.14	271.20
	Min	18.24	6.60	0.54	2.45	26.50	23.60
	Mean ± SD	76.00 ± 40.38	79.52 ± 72.14	12.70 ± 10.80	23.81 ± 33.40	57.20 ± 32.88	80.30 ± 91.32

Max = maximum concentration; Min = minimum concentration; SD = Standard Deviation;
n = total number of samples collected from each site

Table 3: Concentrations of heavy metals in soil samples collected from areas without automobile workshops (mg/kg dry weight)

Sites and sampling points	Sampling point/ Distance (m)/ Category	Zinc		Copper		Lead	
		Feb	May	Feb	May	Feb	May
Makadara;	First point (0)	149.51	184.50	8.43	5.41	34.44	21.83
5 points (n	10	135.33	182.03	6.65	8.70	25.93	94.80
= 10)	20	147.80	137.40	5.50	5.73	45.40	38.32
	30	148.70	112.42	5.71	5.20	88.00	31.71
	40	143.80	196.70	8.72	9.10	174.42	31.71
	Mean concentration	145.03	162.61	7.00	6.83	73.64	43.70
Mtoni	First point (0)	24.20	277.00	0.10	24.41	11.80	113.00
Mazrui;	10	34.60	5.24	0.83	0.50	11.10	3.94
4 points (n	20	38.40	13.24	1.90	0.80	12.52	13.40
= 8)	30	22.74	0.27	0.14	0.55	27.70	3.40
	Mean concentration	30.00	74.00	0.74	6.60	15.80	33.40

n = total number of samples collected from each site

Concentrations of zinc

The concentrations of zinc in all the sites with automobile workshops varied from 6.6 to 419.2 mg/kg dry weight (dw) with mean concentrations ranging from 76 to 192.6 mg/kg dw (Table 2). These values are higher than those observed at Mtoni Mazrui which is free from automobile workshops where the concentrations of zinc varied from 0.27 to 277 mg/kg dw with average concentrations ranging from 30 to 74 mg/kg dw (Table 3). At Makadara, which is also free from automobile workshop, the case was different. The concentrations of zinc in the samples from that site ranged from 112.42 to 196.7 mg/kg dw with mean concentrations ranging from 145.03 to 162.61 mg/kg dw (Table 3). The reasons for these high concentrations at the area free from automobile workshops could not be established under this study, but possible reasons could be due to runoff water from other areas passing through that site since the area is located at lowland, as well as due to atmospheric deposition. There are no rivers or streams crossing that area at Makadara. The elevation of the concentrations of zinc at areas with automobile workshops indicated that there were anthropogenic contributions to the observed levels. Used oil, spray paints and vehicle body paints containing zinc oxide and other garage wastes were the probable sources of this elevation. A similar study by Pam *et al*. (2013) revealed that the average concentrations of zinc at auto mechanic workshop clusters in Benue State, Nigeria ranged from 295.5 to 553.3 mg/kg dw, which are higher than those found in this study. Anapuwa (2014) reported concentrations of zinc ranging from 11.00 to 27.75 mg/kg dw (mean = 16.74 mg/kg dw) in soil from automobile workshops in Delta State, Nigeria, which are lower than those found in this study.

In general, there were significant differences in the concentrations of zinc among the

study areas with automobile workshops in the samples collected in February ($F(4, 40) = 4.625$, $p = 0.0037$). This implied that the activities taking place at these areas differ among the workshops; hence different amounts of the heavy metal wastes are deposited. However, the mean concentrations of zinc between most of the paired individual sites were not significantly different while the concentrations of zinc in samples from Lumumba were significantly lower than in samples from all other sites ($p < 0.05$). For the samples collected in May, there were significant differences in the concentrations of zinc among those sites ($F(4, 40) = 3.708$, $p = 0.0117$) and there were significant differences between the following paired sites: Darajani vs Lumumba and Kikwajuni vs Lumumba ($p < 0.05$). These results could be due to the same reasons explained for the observations on February samples.

The concentrations of zinc in 51.11% of the samples collected from all the sites were above the Tanzania Bureau of Standards (TBS) permissible limit of 150 mg/kg (TBS 2007). The point sources at most of the sites and points located down slope (parts sloping downward) in the study areas were observed to contain higher concentrations of zinc than the TBS permissible limit. This is partly due to large amounts of garage waste at the point sources. The high concentrations of zinc at points away from the point sources are influenced by runoff water that takes away the garage waste and other wastes down the slope. The concentrations of zinc in some (9.3%) of the samples at Darajani, Kikwajuni and Kijangwani exceeded the World Health Organization (WHO) permissible limit of 300 mg/kg (Lacatusu 2000). At Makadara, 30% of the soil samples had concentrations of zinc higher than the TBS permissible limit.

Concentrations of copper

Copper was detected in all the soil samples and its concentrations varied from 0.16 to 167 mg/kg dw with mean concentrations ranging from 4.23 to 51.7 mg/kg dw at the areas with automobile workshops (Tables 2). These concentrations were higher than those found in the samples from the areas without automobile workshops (Mtoni Mazrui and Makadara) which ranged from 0.1 to 24.41 mg/kg dw with mean concentrations ranging from 0.74 to 7 mg/kg dw (Table 3). The probable reasons for this elevation are the same as those explained on zinc. The findings of this study indicated that there was low distribution of copper around the areas with automobile workshops. This could be due to the fact that high proportion of copper that is contained in used oil sinks into the ground through leaching leaving very small amount on the top soil (Anapuwa 2014). There were significant differences in the concentrations of copper in the soil samples among the study areas with automobile workshops in February samples ($F(4, 40) = 3.935$, $p = 0.0087$). There was no significant difference in the concentrations of copper in soil among the study areas in May ($F(4, 40) = 1.276$, $p = 0.2954$).

The concentrations of copper in some (40%) of the samples at the point sources of Kidutani, Kikwajuni, Kijangwani and 50 m away from the point source of Lumumba were above the WHO permissible limit (100 mg/kg) but below the TBS permissible limit (200 mg/kg) (Lacatusu 2000, TBS 2007). The concentrations of copper in the samples from Darajani were below the WHO and TBS permissible limits. The studies of Ojiako and Okonkwo (2013) and Anapuwa (2014) in Nigeria reported concentrations of copper ranging from 0.6851 to 2.0259 mg/kg dw and 0.25 to 1.20 mg/kg dw, respectively, which are lower than the levels observed in this study. In contrast, the study by Pam *et al*. (2013) at auto mechanic workshop clusters in Benue State, Nigeria

reported higher mean concentrations (ranging from 254.1 to 1,384.1 mg/kg dw) than those observed in this study.

Concentrations of lead

The concentrations of lead in the samples in areas with automobile workshops ranged from 0.02 to 271 mg/kg dw with mean concentrations ranging from 32.54 to 103.6 mg/kg dw (Table 2). These concentrations were higher than those found in samples from Mtoni Mazrui, an area without automobile workshop, which ranged from 3.4 to 113 mg/kg dw with mean concentrations ranging from 15.8 to 33.4 mg/kg dw (Table 3). As for zinc, at Makadara which does not host any automobile workshop, the concentrations of lead in February samples were higher than those found in some areas with automobile workshops. This could be due to the fact that Makadara is located in urban area where there are possibilities of being polluted by other factors such as emissions from cars, atmospheric deposition as well as commercial and other activities in the area and neighbouring areas. There were significant differences in the concentrations of lead among the study areas with automobile workshops in the samples collected in February ($F(4, 40) = 3.299$, $p = 0.0199$). There were no significant differences in the concentrations of lead in the soil samples among all the study areas with automobile workshops in May ($F(4, 40) = 1.924$, $p = 0.1252$).

Some points, especially point sources and points located down slope up to 70 m away from the point sources were observed to have concentrations of lead greater than the TBS and WHO permissible limits (Lacatusu 2000, TBS 2007). The concentrations of lead in 18% of the soil samples in and around all the automobile workshop areas were greater than the WHO permissible limit (100 mg/kg). The concentrations of lead in 7.41% of the soil samples at Kikwajuni, Darajani and Lumumba were greater than the TBS permissible limit (200 mg/kg), while the concentrations of lead at other sites were below the TBS permissible limit. The mean concentrations of lead found in this study from all the sites with automobile workshops were comparable to those reported in similar studies. For example Pam *et al*. (2013) and Ojiako and Okonkwo (2013) reported lead concentrations ranging from 283 to 665 mg/kg dw and 3.604 to 33.248 mg/kg dw, respectively.

Sources and distribution of heavy metals contamination in the study areas

The results obtained in the samples from the sites with automobile workshops generally showed that the concentrations of all the heavy metals at the point sources were higher than at the periphery points in all the sites in February samples, while in May the highest concentrations were found in samples collected from points located down slope in all the sites (Figure 2). This therefore indicated that the activities taking place in areas with automobile workshops were responsible for the heavy metals (zinc, copper and lead) contamination in soils in the study areas and the dispersion was influenced by runoff during the rainy season. However, in samples from the areas that do not host any automobile workshops, zinc, copper and lead were also detected. This is probably due to the fact that these sites are in urban areas where the metals have other sources including traffic-related emissions from roads and atmospheric deposition from windblown dusts. Heavy metals also occur naturally in soils although at relatively low concentrations (Mbah and Anikwe 2010).

The concentrations of the metals in soil samples collected in February from all the sites with automobile workshops laterally decreased with increase in distance from the impacted points on both sides (up and down slopes), which is consistent with a similar study in Nigeria by Anapuwa (2014). This

indicated that the dispersion of the metals in soil from the point sources was limited due to the absence of rainfall. The results for the samples collected in May showed that the concentrations of the metals increased with increase in distance down slope from the point sources. The mean concentrations of zinc, copper and lead in samples from most sites were generally higher in samples collected in February than in May. The concentrations of the heavy metals showed similar trends in samples from most of the sites during the dry and rainy seasons, and were generally in the order zinc > lead > copper. The decrease in the concentrations of the metals in May (rainy season) samples was probably due to heavy rainfall that rained during mid March to May. The rainwater caused dilution and took away some portions of the heavy metals as observed by Fagbote and Olanipekun (2010). The elevation of the concentrations of the metals in samples from Kikwajuni and Lumumba in May could be due to runoff water during the rainy season which carried the metals from neighbouring areas and deposited them in soils at these areas since they are located down slope. The soil textures in the study areas were observed to be heavy sandy and sandy loam soils with low to moderate organic matter contents in most of the sampling points and high organic matter contents at some points. Such soil parameters could favour both the vertical (leaching) and lateral distribution of the heavy metals in the areas.

Statistically, there were no significant differences in the concentrations of the metals between the soil samples collected in February (dry season) and May (rainy season) (t = 0.152–1.37, df = 7–9, p = 0.213–0.988 for zinc; t = 0.99–1.733, df = 9, p = 0.177–0.835 for copper and t = 0.713–0.907, df = 9–10, p = 0.386–0.934 for lead). This indicated that, the dry and rainy seasons had no significant impacts on changing the overall concentrations of the heavy metals in the study areas during that period.

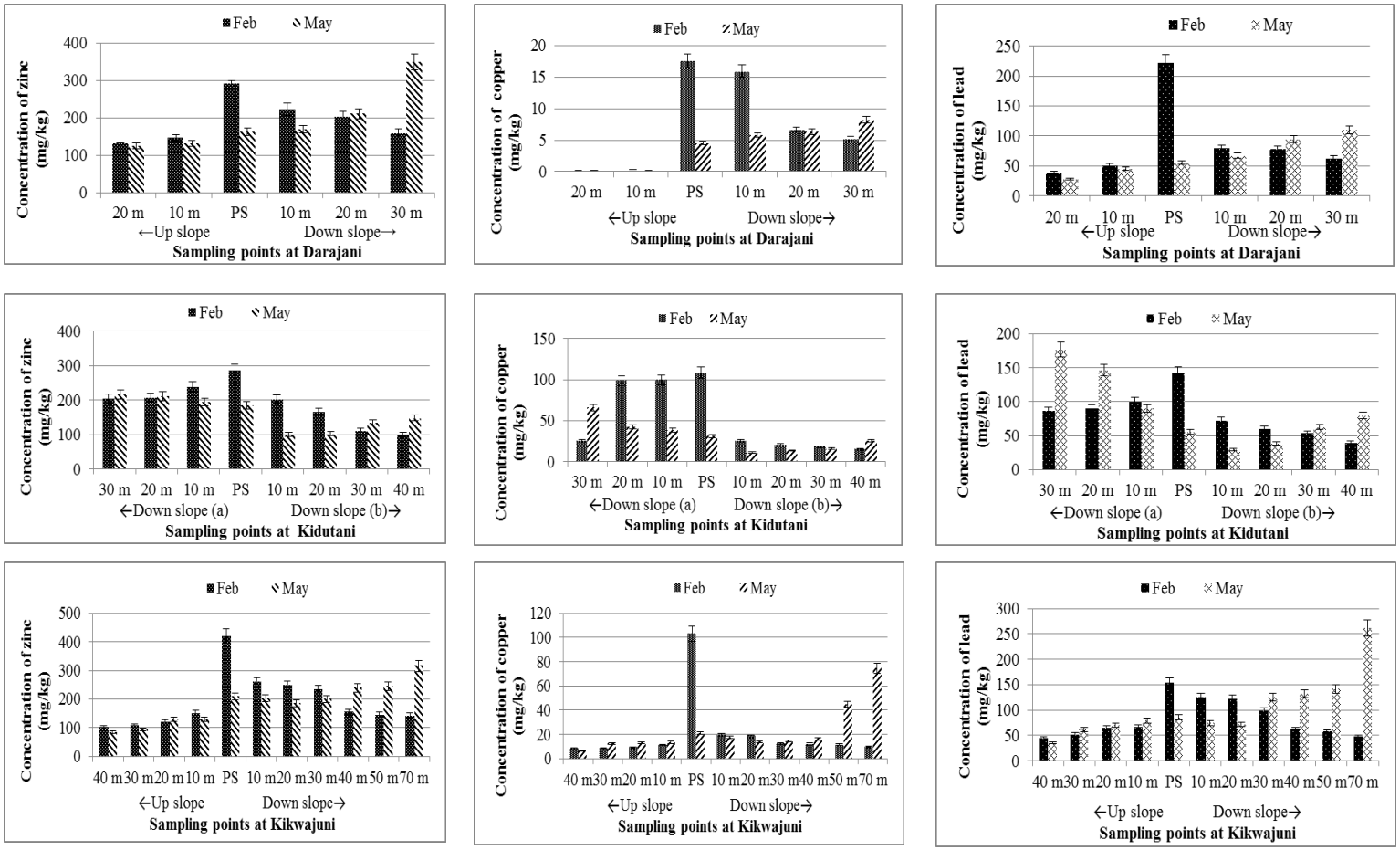


Figure 2: Distribution of concentrations of heavy metals in soil in areas with automobile workshops (PS = point source)

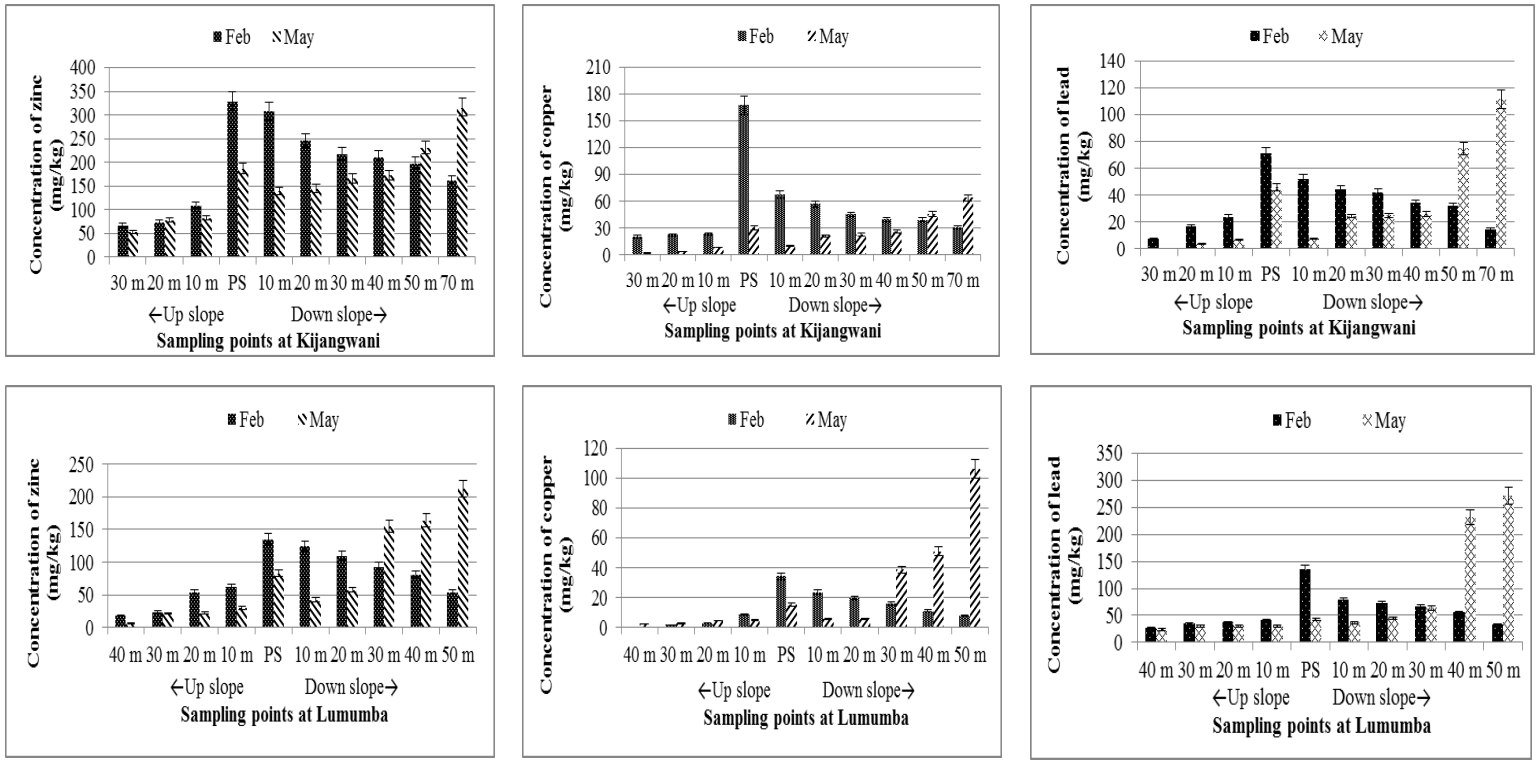

Feb
May
Concentration of zinc (mg/kg)
Concentration of copper (mg/kg)
Concentration of lead (mg/kg)
30 m 20 m 10 m PS 10 m 20 m 30 m 40 m 50 m 70 m
40 m 30 m 20 m 10 m PS 10 m 20 m 30 m 40 m 50 m
←Up slope
Down slope→
Sampling points at Kijangwani
Sampling points at Lumumba

Figure 2 (ctd)

CONCLUSIONS

The findings of this study indicated that the automobile workshops were indeed polluted with high levels of zinc, copper and lead and their concentrations were higher than those obtained in areas without such workshops. The metals were found to be dispersed to other points and at increasing levels down slope in samples collected during the rainy season, indicating that runoff water highly contributed to the dispersion of the metals from the sources. Therefore, the automobile workshop wastes were the sources of the elevation of zinc, copper and lead concentrations in the study areas. No significant differences were observed in the concentrations of these metals between the sampling periods. Some of the concentrations of the heavy metals were greater than the permissible limits indicating risks and concerns for the environment and public health. Control measures should be taken to prevent further contamination.

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