



Spatial Distribution of Some Toxic Metals in Topsoil and Bioaccumulation in Wild Flora Around a Metal Scrap Factory: A Case of Southwestern Nigeria



ABSTRACT

There is increasing metal pollution in soil due to the spate of industrialization in developing countries. It is premised on this scenario that the topsoil around a metal scrap factory was studied to assess the spatial distribution of metal and resultant mobility in native flora. Concentrations of Cd, Zn, Pb and Fe in samples of topsoil and plants were determined by Atomic Absorption Spectrophotometry. Spatial modelling of metals was carried out using the Inverse Distance Weighted (IDW) technique of ArcGIS. Also, metal mobility in native flora was assessed by the transfer factor model. The analysis showed high levels of Cd, Zn, Fe and Pb that were beyond natural concentrations. Spatial mapping of the concentrations indicated that the immediate north and south of the factory were significantly polluted by Cd, Zn and Pb while Fe presence in the area was partially geogenic. The level of Cd, Zn and Pb in the three native species exceeded permissible limits whereas only Cd showed great mobility within the biomass of the three species. The mobility of Cd in the diagnostic species is an indication of possible potential health risk from Cd toxicity through plants ingestion. It is therefore important that measures should be geared towards improving the pollution control system of the factory to stem down the rate of contamination of soil.

Key words: Heavy metal, spatial distribution, metal mobility, metal accumulation

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INTRODUCTION

The spatial variability and characterization of metal concentrations in the topsoil may be affected by soil parent materials, anthropogenic sources and the multiple sources of introduction (Hanesch *et al.* 2001; Martin *et al.* 2006). Different anthropogenic routes of metal introduction into the soil are industrial emissions and wastes generation, agricultural activities, metalliferous mining and smelting, energy and fuel production, and vehicle emissions (Zeidler 2005; González and González-Chávez 2006). It is worthy of note that presence of metals in soil is not only toxic to plants and deteriorates soil optimal bio-productivity but also causes severe threat upon exposure to human health especially at elevated level. Some of these metals (Pb, Cd, Hg, As) are known carcinogens and causes a long term damaging effect on the central nervous system. They are non-biodegradable and they have the tendency to bioaccumulate and biomagnify from one trophic level to another (Khan *et al.* 2008; Oti *et al.* 2012). Plants grown in polluted environment accumulate toxic metals more than others grown outside such an environment and may pose health hazards to biota and humans, which impose harmful impacts upon increased level of exposure (Ikeda *et al.* 2000; Khan *et al.* 2008; Luo *et al.* 2011).

Scrap metals is becoming a booming municipal solid

wastes (MSW) business in Nigeria; through scavenging activities, scrap metals are recovered from MSW and recycled into products like iron and steel, copper, brass, aluminum (Olanrewaju and Ilemobide 2009; Onwughara *et al.* 2010). The scrap metal recycling process involves remelting of scraps in an electric arc furnace or induction furnace using up to 90-100% scrap and reheating the formed steel bars in a reheating furnace to produce iron rods. The adoption of furnace systems in metal recycling process always results in the generation of dust and metal fumes depending on the operating temperature and raw material contents (EPA 2001). Metal particles in the gaseous emissions, if not properly treated through proper/stringent pollution control may be released and deposited into the environment. Depositions may be at different distances away from the factory depending on the wind speed, particle size, atmospheric temperature and residence time of metal particle (IFS 2007; Luo *et al.* 2011). Some of the toxic metals released during metal scrap recycling process are Cd, Pb, Zn, Cr, Co and Cu (IFS 2007; OSHA 2008; Luo *et al.* 2011).

Due to the potential ecological hazards that the presence of these toxic metals at high concentrations in the soil may cause, this study aimed at investigating the spatial distribution in topsoil and bioaccumulation of some of these

metals in wild flora around Prism Steel Mills Company, a 7-year old steel recycling factory. The previous study on the assessment of ecological vulnerability to trace metals in topsoil around this recycling factory by *Ogunkunle et al. (2016)* has indicated potential ecological hazards from Cd and Pb. This study aims to assess the spatial distribution of Cd, Zn, Pb, and Fe in the topsoil; and to evaluate metal mobility and accumulation in the native flora around the factory.

METHODOLOGY

Study area

The study was carried out around Prism Steel Mills Company, Ikirun in Southwestern Nigeria (**Figure 1**). The location of the factory, climate and geology of the study area have been previously described in a previous study by *Ogunkunle et al. (2016)*.

Sampling and chemical analysis

Seventy-five samples of topsoil (0-15 cm) were collected within 1-km radius from the perimeter fence of the factory (*Ogunkunle et al. 2016*) with the aid of GPS equipment (Garmin Etrex 20, USA). Samples of plant species (root + shoot) that were encountered within 1 m radius of each sampling point were collected and enumerated to species level. After identification, 35 samples of *Chromolaena odorata*, 20 samples of *Tithonia diversifolia* and 25 samples of *Sida acuta* were used for the

study based on the species composition/abundance.

Soil pH was determined in a soil suspension (1:2.5 w/v) using a glass electrode pH meter (PHC-3C model) and the total organic matter (OM) content was determined by loss on ignition (*Reddy et al. 2009*). Digestion of soil and plant samples were done using aqua regia method (*Chen and Ma 2001*). Briefly as described in *Ogunkunle et al. (2016)*, 1 g of each sample (soil or plant) was weighed into a 250-mL Pyrex glass beaker, and a mixture of 2 mL concentrated HNO_3 and 6 mL HCl were added and heated on a hot plate for 2 h at a temperature of 110°C until the solution becomes clear. The solution was allowed to cool, filtered using Whatman No. 42 filter paper into a 50-mL volumetric flask and made up to mark with deionized water. Cd, Zn, Pb and Fe in the digest were determined by Atomic Absorption Spectrophotometry using Peckin Elmer A Analyst 200. Quality Assurance/Quality Control (QA/QC) was ensured by replicate digestion, use of blanks and percentage recovery of elements using certified reference materials for soil and plant (IAEA-SL-1 and IAEA 359, respectively) were between 84-95% and 92-110%, respectively.

Statistical analysis and data analysis

Kolmogorov–Smirnov tests was used to evaluate if data were normally distributed since normal distribution of data has been proven to theoretically favour geo-statistical modeling (*Cressie 1993*). Kruskal-Wallis test was used to determine significant differences ($P < 0.05$) among the mean

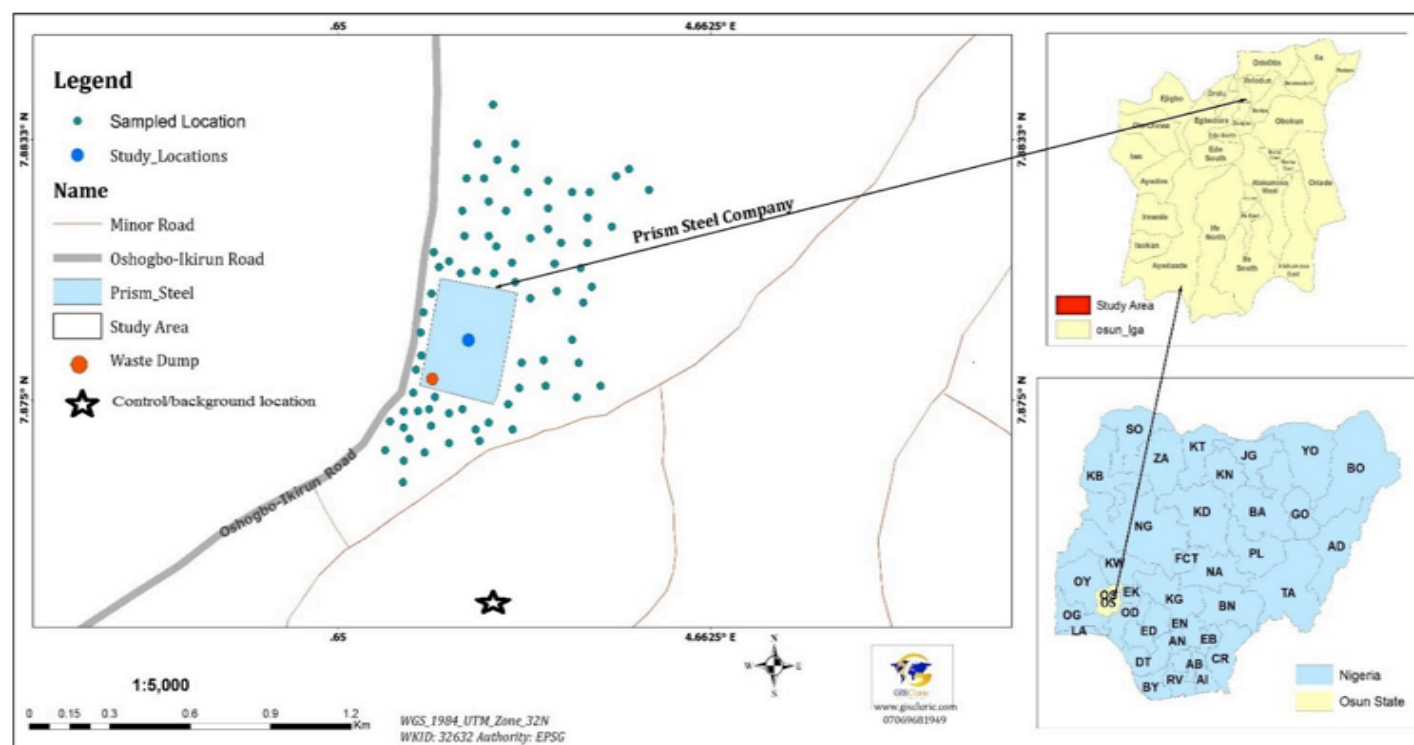


Figure 1. Map of study area showing Prism Steel Mills Company, Southwestern Nigeria (*Ogunkunle et al. 2016*).

concentrations of metals in plant biomass as the distribution of plant data failed the Normality test. Statistical Software for Social Sciences (SPSS) for Windows version 16.0 was used for the analysis.

Spatial distribution of total concentration of Cd, Zn, Pb and Fe in the soil was carried out by Inverse Distance Weighted (IDW) technique of ArcGIS 10 [Environmental System Research Institute (ESRI), Redlands, Canada]. It relies mainly on the inverse of the distance raised to a mathematical power which controls the significance of known points on the interpolated values based on their distance from the output point (Watson and Philip 1985).

Assessment of ecological vulnerability through metal transfer in the flora was studied by assessing the mobility of metal (Transfer factor) into plants (Intawongse and Dean 2006).

Transfer factor (TF) is defined as the ratio of metal concentration in plant biomass to concentration of total metal in soil (Cui *et al.* 2004; Chojnacka *et al.* 2005) and expressed in equation 1.

$$TF = \frac{C_p}{C_{ts}} \quad \text{Equation 1}$$

Where:

C_p = the concentration of a particular metal in plant biomass
 C_{ts} = the concentration of a particular metal in soil

RESULTS AND DISCUSSION

Level and spatial distribution of Cd, Zn, Pb and Fe in the topsoil around the factory

The chemical characteristics of the topsoil has been previously reported in Ogunkunle *et al.* (2016). Soil pH was slightly acidic to neutral and the average soil OM showed a relative fertility of the soil (Table 1). These two attributes render metal ions readily bound to soil and reduce bioavailability. Solubility, bioavailability and adsorption of metals to solid phases in soil are influenced by soil properties like the pH and OM (Alloway 1995; Vega *et al.* 2004; Stehower *et al.* 2006). Despite the vegetation within the site,

the OM content in the soil was considerably lower than the soil found within the same region of the state (Awotoye *et al.* 2011; Salami and Sangoyomi 2013), but yet higher than some other soils (Salami and Sangoyomi 2013) in other areas.

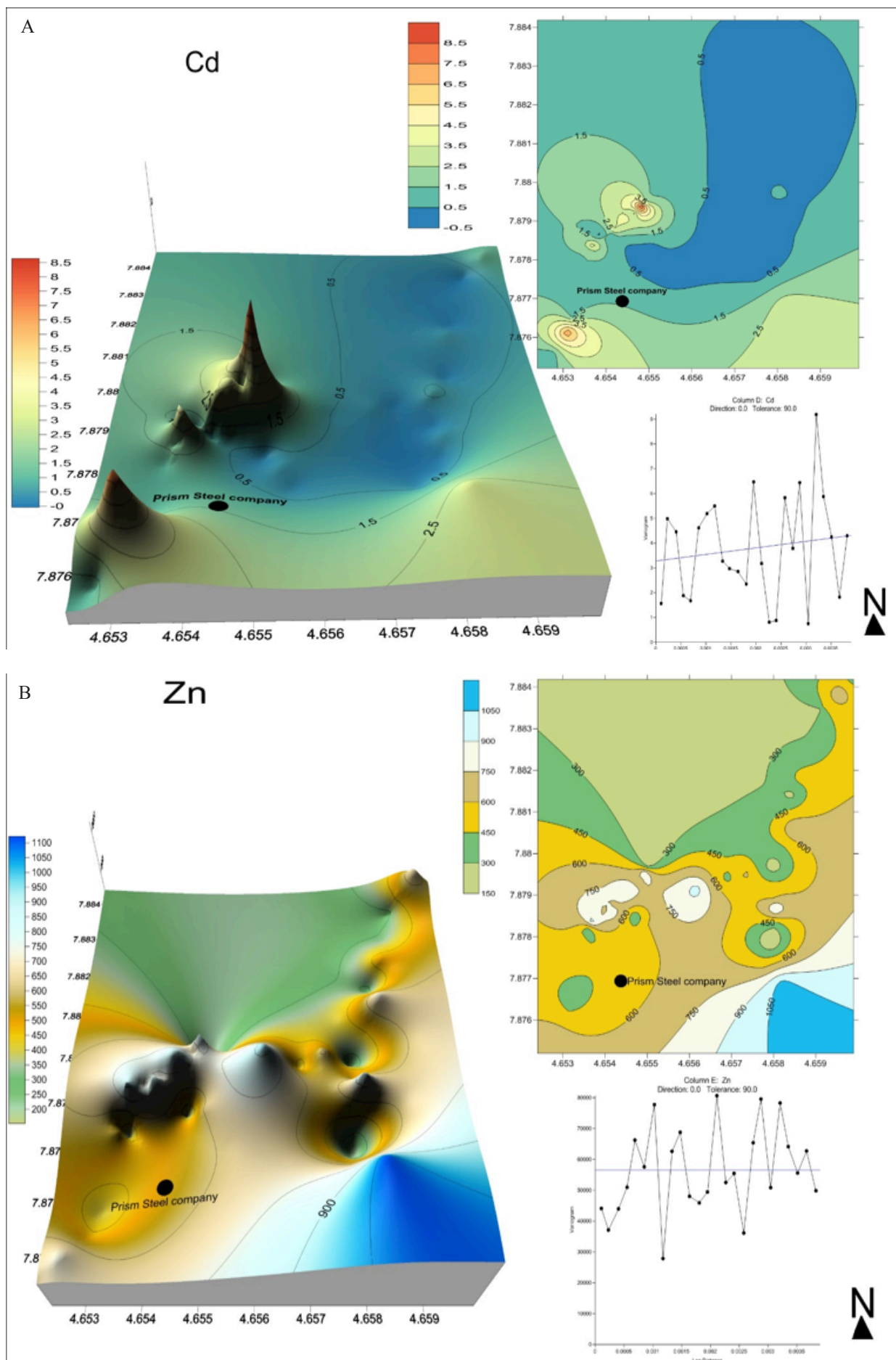
Concentrations of trace metals in the soil varied with some hotspots but the average concentration of the metals followed the order Fe > Zn > Pb > Cd. The results indicated that most of the metal concentrations were above the Environmental Quality Standards for Soils as previously reported by Ogunkunle *et al.* (2016). These concentrations were also found to exceed the environmental soil quality guidelines set by the National Environmental Standards Regulations and Enforcement Agency of Nigeria. The levels of Cd, Zn and Pb in the soil surrounding the recycling factory exceeded the crustal value by 4.3, 7.6 and 2.9 folds, respectively; this is an indication of increasing contamination of the topsoil around the factory. Though, the concentrations of metals in the soil may be influenced by the geology of the parent materials, but elevated concentrations above tolerable level is a strong indication of anthropogenic influence on the environment (Al-Khashman and Shawabkeh 2006; Awotoye *et al.* 2011). Furthermore, the mean concentrations of Pb and Zn in the study exceeded concentrations reported in similar studies on metal recycling factories in Spain (Acosta *et al.* 2011), Algeria (Maas *et al.* 2010) and Greece (Voutsas *et al.* 1996). In addition, the level of Cd in this study was higher than the concentrations found in studies on metal recycling factories in Algeria (Maas *et al.* 2010) and Spain (Acosta *et al.* 2011) but lower than Cd concentration (289 mg kg⁻¹ and 11.9 mg kg⁻¹) reported for Fe and Zn smelters in Bulgaria and China, respectively (Li and Huang 2007; Schulin *et al.* 2007).

It was realized that all trace metals (Cd, Zn and Pb) except Fe were heterogeneously distributed around the factory with peculiar hotspots indicating potential anthropogenic source. These localized hotspots were mainly located at the north and north-eastern part of the factory. For instance, the mapping of Cd showed low concentration of Cd in the North of the factory while the South of the factory showed an even spread of high concentrations of Cd. Localized 'hotspots' (areas with isolated extreme

Table 1. Descriptive statistics of metal concentrations in the topsoil surrounding the factory (Ogunkunle *et al.* 2016).

	Metal Concentration (mg kg ⁻¹)					
	pH (H ₂ O)	OM	Cd	Zn	Pb	Fe
Mean	6.9±0.1	1.5±0.05	1.3±0.9	573.2±141.1	40.5±30.7	20 246.0±5 792.1
Minimum	6.7	1.4	0.5	137.5	3.3	10 875
Maximum	7.2	1.5	9.6	1 125.0	118.7	37 825
CCME ^a	NA	NA	1.4	200	70	NA
Mean crust ^b	NA	NA	0.3	75	14	47200 ^c

^aCanadian Council of Ministers of the Environment (2007); ^bBowen (1979); ^cTurekian and Wedepohl (1961)



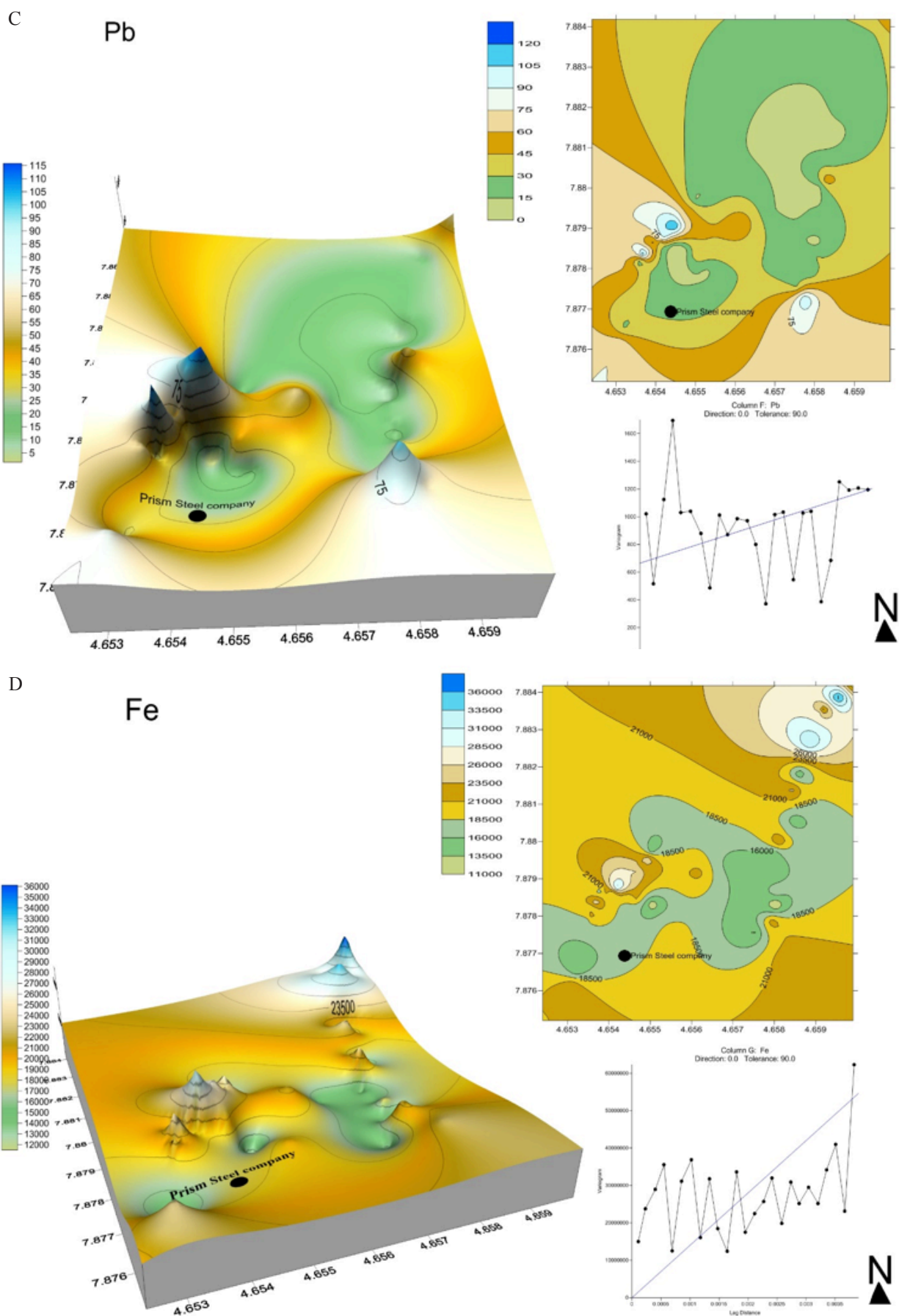


Figure 2. Spatial distribution of (a) Cd, (b) Zn, (c) Pb and (d) Fe in the topsoil using Inverse Distance Weighted technique (ArcGIS 10).

concentrations) of Cd were also noticed in the north and south-east parts of the factory. Kriging graph of Cd distribution indicated relative variability in the concentrations of Cd around the factory (**Figure 2a**). Similarly, low concentrations of Zn were noticed in the far-north of the factory while high concentrations were observed at the immediate north, north-eastern and south-western part of the factory (**Figure 2b**), whilst extremely high concentrations of Zn were also observed at the far south-eastern part of the factory. Variability in the spatial distribution of Pb was noticed in the immediate north and north-eastern part of the factory with some isolated hotspots. Low distribution of Pb was observed at the far north-east of the factory while the far south-eastern was largely distributed with relatively high Pb loads (**Figure 2c**). Mapping of Fe concentrations in the topsoil surrounding the factory indicated extremely low variability (as shown by the Kriging graph) with limited number of hotspots around the northern area of the factory (**Figure 2d**). The low variability indicated by the Kriging graph indicates that Fe was homogenously distributed in the topsoil and that Fe contents in the soil were geogenic in nature, since control soils within Ikirun were <50% of the values in this study as per *Abidemi (2013)* and *Abiala et al. (2013)*.

The heterogeneous distributions of Cd, Zn and Pb in the maps showed that most of the metal contents in the topsoil were from artificial sources, and the main source is likely the factory. Steel and iron manufacturing has long been demonstrated as a major source of trace metals (e.g. Cd and Pb) but depends largely on the type of technology used for processing (*Pacyna 1987*). Cadmium is used in the plating of iron, steel, and other metals, whilst lead is used to increase durability of steel, Zn is used as an alloying and coating element and Fe is also part of the scrap materials used in steel production (*Pacyna 1987; Janke et al. 2000; Arruti et al. 2010*). These metals are often released as particulate matter ($PM_{2.5}$, PM_{10}) associated with dust or as off-gas during pre-treatment and melting stages of steel production. In addition, metal-containing dust particulates are generated from unprotected storage of metal scrap within site for pre-treatment (*Janke et al. 2000; Jiang et al. 2002*). The emitted dust and off-gas would eventually settle on nearby soils by rain or wind and would account for the hotspots of metal concentrations around the immediate vicinity of the factory. The heterogeneous distribution of Cd, Zn and Pb could be attributed to the wind action as the wind directions in the region are south-westerly and north-easterly in the rainy and dry seasons, respectively. Wind transport of dust has previously been pointed as an important factor that influences the spread of pollutants from industrial foci (*Al-Khashman and Shawabkeh 2006; Tembo et al. 2006; Owoade et al. 2014*).

Accumulation and transfer of Cd, Zn, Pb and Fe in flora around the factory

Metal accumulation in native flora around the factory resulted to the following pattern: Cd: *C. odorata*=*T. diversifolia*=*S. acuta*; Zn: *T. diversifolia*> *Chromolaena odorata*= *Sida acuta*; Pb: *C. odorata*=*T. diversifolia*=*S. acuta*; and Fe: *T. diversifolia*> *C. odorata*=*S. acuta* at $P<0.05$ (**Table 2**). Level of Cd and Pb in the three native flora exceeded the permissible limit in plant (0.2 mg kg^{-1} and 2.0 mg kg^{-1} , respectively). Concentrations of Zn in *C. odorata* and *T. diversifolia* exceeded the permissible threshold by the Canadian Council of Ministers of the Environment (CCME) while Zn accumulation in *S. acuta* was below the threshold (200 mg kg^{-1}). In comparison with another study, the levels of metals in plants in this study were significantly higher than the reported concentrations in plants by the study of *Owoade et al. (2014)* around a similar facility in southwest Nigeria. The possible reason for this variation is pinned on the factory's duration of production. In this study, the factory is operating for seven years as against the less than 4 years of production in the report of *Owoade et al. (2014)*. The near neutral pH (6.91) and OM content (1.4%) are responsible for reducing potential mobility and bioavailability of some of the trace metals despite the high concentrations found in soil. However, within the soil matrix, fractions of individual metal are bioavailable to biota, whilst residual fractions are non-bioavailable (restricted by pH and OM) at particular time, however if the concentration of such trace metals increases, the fraction of individual metals bioavailable or mobilized increases owing to carrying capacity of the soils (*Papadopoulos et al. 2007; Zeng et al. 2011; Adamo et al. 2014*). Although not investigated, metals often exist in four major forms in soils: dissolved, mineral, organic and sorbed (bound to soil). Most of the metals remain bound in soil mineral and organic matter, and is non-bioavailable to biota such as plants (*Jones and Jacobsen 2009; Zeng et al. 2011*).

The metal transfer to the native flora in terms of transfer factors (TFs) were analyzed (**Figure 3**). Transfer of Cd in *C. odorata* was greater than 1.0 ($TF_{Cd}>4.0$) and 54% of sampled plant species recorded $TF>1.0$ while TF_{Zn} , TF_{Pb} and TF_{Fe} were below 1.0. *T. diversifolia* also recorded TF_{Cd} greater than 1.0 ($TF_{Cd}>6.0$) and 56% of the plant species that were sampled gave $TF>1.0$ while TFs for Zn, Pb and Fe were below 1.0. The same pattern of TF recorded in *T. diversifolia* was the same in *S. acuta*. TF_{Cd} was greater than 1.0 ($TF_{Cd}>8.0$) and 74% of sampled plants recorded $TF>1.0$ while TF_{Zn} , TF_{Pb} and TF_{Fe} were below 1.0. Since greater than 50% of total number of sampled plants had $TF_{Cd}>1.0$ in all the plant species, these species can be referred to as potential accumulator of Cd. Several studies

Table 2. Metal accumulation in plant species growing around Prism Steel Mills factory.

Plant Species	Metal Accumulation in Plant Biomass (mg kg ⁻¹)				
	N	Cd	Zn	Pb	Fe
<i>Chromolaena odorata</i> ¹	35	0.9±0.5 ^{a*}	225.6±80.7 ^b	9.3±3.91 ^a	746.1±100.6 ^b
<i>Tithonia diversifolia</i> ¹	20	0.8±0.6 ^a	323.5±134.6 ^a	12.4±6.8 ^a	1 213.1±266.1 ^a
<i>Sida acuta</i> ¹	25	0.8±0.5 ^a	184.4±53.9 ^b	10.8±6.9 ^a	453.1±140.6 ^b
Permissible limit in plant ²		0.2	200	2.0	NA

*Values with the same letter are not significantly different at $P<0.05$ by Kruskal Wallis test.

¹Refers to perennial herb; ²FAO/WHO (1996); NA- Not applicable

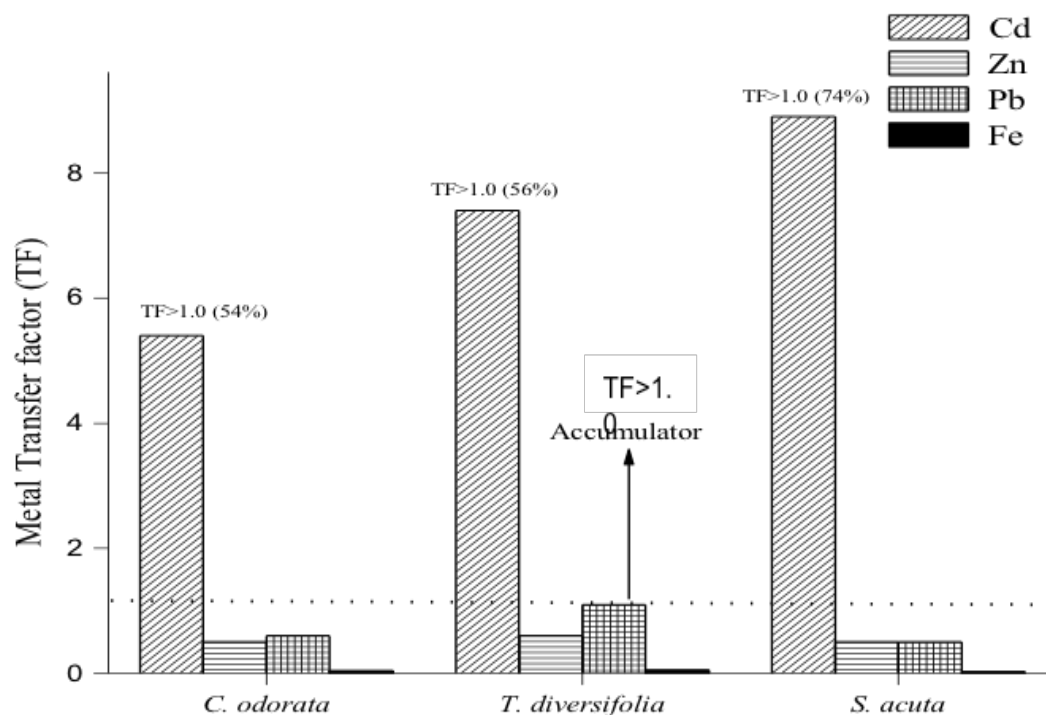


Figure 3. Transfer factor (TF) of metals in biomass of plant species around Prism Steel Mills factory.

have also reported the potential of *S. acuta* to accumulate and phytoextract Cd from polluted soil (Gupta and Sinha 2007; Ogunkunle et al. 2014) and the possible reason for greater accumulation of Cd by the three species could be linked to the nature of Cd, it is less retained by the soil than other toxic metals (Lokeshwari and Chandrappa 2006). It has also been reported that plants could accumulate Cd more efficiently than other trace metals in the ecosystem (Amoo et al. 2005; Fagbote and Olanipekun 2010). Ecological implications of the high mobility of Cd in the flora around the factory could be felt primarily on herbivores that browse on these plants through biomagnification of Cd and therelative transfer to humans through the food chain.

CONCLUSION

The study has evaluated the metal status of the surrounding topsoil of metal scrap factory after seven years of production. It was found that Cd, Zn and Pb have been greatly enriched in the soil and the level of pollution was high but still manageable if consideration to pollution

control is given by the factory. Spatial mapping of metal concentrations showed that the immediate environment of the factory is at risk of progressive contamination which may portend potential ecological hazards. Plant species found at the surrounding showed reduced accumulation of Zn, Pb and Fe while Cd showed significant mobility in the studied species; which is suggestive of being potential Cd accumulator. The high mobility of Cd in the soil seems to indicate serious ecological problem because of the potential transfer of Cd along the food chain. It is recommended that increased efforts should be put into the pollution control system of the factory so that metal levels in the soil especially Cd and Pb would not exceed tolerable threshold.

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