

Evaluation of accumulation and concentration of heavy metals in different urban roadside soil types in Miranda Park, Sydney

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Abstract

Purpose The overall objectives of this study were to examine the relationship between the concentrations of heavy metals such as Pb, Zn, Cu, and Cd in roadside soil derived from three different geological parent materials, Hawkesbury Sandstone, Wianamatta Shale, and Mittagong Formation and also to examine the influence of rainfall events on heavy metal concentrations in both the topsoil and the subsoil in all three soil types. In this paper, the focus is on lead and zinc.

Materials and methods The results obtained from the samples taken from an initial transect were used to select the location of the study sites. Soil samples were collected using a stainless steel auger at distances of 1, 5, and 10 m from the edge of two major roads of similar traffic volumes bordering a suburban park. At each of five study sites, samples were collected at depths of 0–10 and 10–30 cm, three times pre-rainfall (after extended periods of no rain) and three times post-rainfall (after intensive rainfall periods). The modified aqua regia digestion method was applied for heavy metal concentrations measurement. To determine the temporal dynamics of trace elements in the soils, sequential extractions were applied to all the topsoil samples according to the modified three-step sequential extraction procedure.

Results and discussion The corresponding concentrations of Pb and Zn were different for the soil derived from Hawkesbury Sandstone and Wianamatta Shale and also

Mittagong Formation. The highest concentration of Pb was in the soil from Wianamatta Shale, 159.32 mg/kg and the highest concentration of Zn was in the soil from the Mittagong Formation, 254.12 mg/kg, all at a distance of 1 m from the roadside. From the sequential chemical extraction results, the rainfall substantially influenced the exchangeable fraction (F1) of Pb at a distance from the road of 1 m. A significant reduction of F1 was found for the soil derived from Mittagong Formation which also had the most significant reduction of total Zn concentration.

Conclusions The interpretation of the results showed that there was a clear correlation between the concentration of Pb and Zn with the distance from the roadside and depth in all soil types. However, the results also showed that there are variable concentrations between the soil types. The heavy metal concentrations at the same distance for the three soil types are different. The rainfall events do influence the heavy metal concentration differently in both topsoil and subsoil of the three soil types at the same distance from the roadside.

Keywords Heavy metals · Rainfall events · Soil type · Urban park

1 Introduction

In recent years, many researchers have investigated the presence of heavy metals in urban roadside soils, especially where contamination might have a detrimental role to public health (Cicchella et al. 2008; Azeez et al. 2014; Vural 2013). Curran-Cournane et al. (2015) stated that public health impacts occur with the frequency of human contact with roadside soils through inhaling suspended road dust or by direct bodily contact with heavy metals accumulating over time. An understanding of the spatial and temporal distribution of heavy

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metals in roadside soil provides the advantage of rapid evaluation of contamination variability in an area and accurate determination of areas which may contain excessive amounts of toxic elements (Sun et al. 2010; Škrbić and Đurišić-Mladenović 2013; Karim et al. 2014). The ensuing results from such a database compilation of urban trace metal concentrations could benefit decision-making such as developing soil protection guidelines used to determine soil suitability for specific land development.

Heavy metal concentrations in urban roadside soil are influenced and determined by two critical factors: local geologies weathered to form the parent material of the soil and also anthropogenic activities, including vehicle emissions, coal and fuel combustion, paint, and local industry (Al-Chalabi and Hawker 2000; Werkenthin et al. 2014). From current forecasts, approximately 64.8 % of the projected 8.3 billion global population will move and live in urban areas (Owusu and Oteng-Ababio 2015). Thus, it is vital to evaluate and quantify the concentrations of contaminants in urban soils to protect environmental and human health.

Elevated metal concentrations such as Pb, Zn, Cu, Cd, Ni, and Cr in roadside soils have been consistently found in cities around the world. In most studies, traffic emission was recognized as the most important factor that would lead to significant quantities of metals deposited in roadside soils, for example, in China (Chen et al. 2010; Wei and Yang 2010; Lu et al. 2012), Turkey (Guney et al. 2010; Kadioğlu et al. 2010), Greece (Massas et al. 2009), Pakistan (Karim et al. 2015), and Nigeria (Azeez et al. 2014). Some studies have focused on the concentrations of heavy metals in roadside soils derived from vehicular emissions and demonstrated their detrimental impacts on the surrounding environment (Khan et al. 2011; Yan et al. 2012; Vural 2013). In most studies, specific roadside soil pollutant metals such as Pb, Zn, Cu, and Cd were mainly investigated because of their known effect on human health. However, in current years, other metals such as platinum group elements (PGE) and antimony (Sb) have raised the attention of researchers in urban soil contamination (Kaur and Katnoria 2014; Liu et al. 2015; Wichmann and Bahadir 2015).

Further studies have investigated the factors addressed from different aspects that have influenced the heavy metal accumulation and distribution in roadside soils. For example, Chen et al. (2010) had explored the relationship between distance from the traffic and the heavy metal concentration levels in roadside soil and have shown that the concentration decreases with increasing distance from the road. De Silva et al. (2015) investigated the impacts of road age, traffic density, and vehicular speed on heavy metal concentration; the results of this study from Pearson's correlations analysis indicated that roadside soil metal concentrations have significant correlations with vehicular speed ($R=0.90$), road age ($R=0.82$), and traffic density ($R=0.68$).

Sydney, the economic and cultural center of the state of New South Wales in Australia, has experienced rapid industrialization and urbanization in the last decades. However, the rapid development has had severe negative effects on the urban environment such as the increase in urban traffic resulting in contamination of roadside soils (Guney et al. 2010; Li et al. 2015). Although since 2002 leaded petrol has been prohibited in Australia, roadside soils are continuously being affected by lead contamination. Previous works carried out by Bakirdere and Yaman 2008 and Ayrault et al. 2012 found that the concentration of Pb is still retained at a high level in urban roadside soils due to the long period of use of leaded fuel and a continuing source from the wearing of metal parts of vehicle bodies. In the available literature, previous research undertaken has not considered the variation in concentration of heavy metals in roadside soil derived from different geologies impacted by wet and dry conditions.

Thus, the purpose of this study herein was to investigate whether there was a relationship between the concentration of heavy metals in roadside topsoil and subsoil derived from three different geological parent materials, Hawkesbury Sandstone, Wianamatta Shale, and Mittagong Formation. In addition, the influence of rainfall events was investigated and, while the concentration of a number of heavy metals was determined, the focus of this paper is on lead and zinc.

2 Experimental

2.1 Study area

The site, Miranda Park, was chosen for the following specific reasons: similar traffic volumes (approximate AADT 30,000, the traffic volume was calculated based on Traffic Authority New South Wales (2005) on each of the major roads adjacent to the park: The Boulevard (northern side) and The Kingsway (southern side); after initially consulting the geology of the Sydney 1:100,000 sheet (Herbert 1983) and soil landscape of the Wollongong-Port Hacking 1:100,000 sheet (Hazelton and Tille 1990), this park is sited in a conjunction of three different geological areas; from the examination of aerial photographs, there were no obvious changes to the landscape from 1930 to the present. Then, the site was visited from which it was confirmed that different geologies were present (Hawkesbury Sandstone, Wianamatta Shale, and Mittagong Formation) from which the three different soils (yellow podzolic, red podzolic, and yellow earth) were derived. A transect that intersected these three geologies was established (Fig. 1). By considering the results from the initial transect, three sampling locations were selected; there were AB for Hawkesbury Sandstone, CE for Wianamatta

Fig. 1 Research area, *AB*—Hawkesbury Sandstone, *CE*—Wianamatta Shale, *D*—Mittagong Formation sampling 1 to 9, was the initial transect to determine the background heavy metal level



Shale, and D for Mittagong Formation). They were sampled for 18 months during periods of variable rainfall events. Site information is shown in Table 1.

2.2 Soil sample collection

Samples were collected at depths of 0–10 and 10–30 cm at distances of 1, 5, and 10 m from the edge of both major roads at each of the five sites. Six soil collections were conducted according to rainfall events, three times pre-rainfall collections

(after extended periods of no rain) and three times post intensive rainfall events over a period of 18 months from November 2013 to May 2015. All the sample locations were recorded using GPS in order to continually collect samples at the same location during the whole sampling period. In order to prevent any sampling contamination of the soil, samples were collected using a stainless steel auger in which the topsoil (0–10 cm) and subsoil (10–30 cm) were able to be collected at the same time. Then, the topsoil and subsoil samples were separated.

Table 1 Sample site description

	Site AB	Site CE	Site D
Close highway name	Kingsway	Kingsway	The Boulevarde
AADT (% heavy) 2014 ^a	30,148	30,148	28,424
Speed limit (km/h)	60, 40 in school time	60, 40 in school time	60
Road type, lanes	1 + 1	1 + 1	1 + 1
Annual mean precipitation (mm) ^b	348.7	348.7	348.7
Topsoil texture ^c and clay content (%)	Sandy loam, 7–20	Silt loam, 12–27	Sandy clay loam, 20–35
Subsoil texture and clay content (%)	Light clay, 35–40	Silty clay, more than 40	Sandy clay, more than 35
Geology ^d	Hawkesbury Sandstone	Wianamatta Shale	Mittagong Formation
Topography	Slope from 10 m	No slope	No slope
Road inclining	Flat	Flat	Flat
Annual wind direction	West	West	West

^a The AADT data is calculated based on RTA Traffic Volume Data for Sydney region 2005

^b The annual mean precipitation is cited from WillyweatherTM

^c Soil texture is conducted in the field

^d The geology information is cited from Herbert (1983)

2.3 Sample preparation and analysis

Inclusions such as leaves and stones were removed from the field soil samples; the soils were then air-dried, ground, and passed through a 2-mm sieve in order to obtain a dry particle-size fraction smaller than 2 mm. For the chemical analyses, all the reagents used were analytical grade reagents. The glass that was in contact with the samples was cleaned with 10 % nitric acid and rinsed with Milli-Q water. In addition, the quality assurance and quality control (QA/QC) procedures were conducted by certified reference material (CRM, DC73309) which is approved by the China National Analysis Centre. CRM was paralleled and analyzed with roadside soil samples by identical procedures; duplicate and blank samples were analyzed as part of the methodology to ensure repeatable results.

The modified aqua regia digestion method (Gaudino et al. 2007) was applied for heavy metal measurement. Firstly, 1.5 mL H₂O₂, 4.5 mL HCl, and 1.5 mL HNO₃ were added to the soil. Hydrogen peroxide was used to enhance the destruction of organic matter. After adding the chemical reagent, all the samples were transferred to microwave digestion. The sequence of each digestion cycle is referred to in Gaudino et al. (2007). The solution was transferred from the beaker to the volumetric polypropylene tube (25 ml) via filter media and the solution was made up to the mark with dilute nitric acid. The concentrations of heavy metals were measured by ICP-MS (Inductively Coupled Plasma-Mass Spectrometry).

To examine the temporal dynamics of trace elements in soils, sequential extraction was applied for all the topsoil samples according to the modified BCR three-step sequential extraction procedure (Pueyo et al. 2008). In the first step, the exchangeable and weak acid soluble fraction can be extracted. In the second step, the reducible fraction can be extracted and the oxidizable fraction can be extracted in the third step. Moreover, the residue from the third step is the residual fraction. Initial statistical analysis (mean, standard error) was conducted by Excel 2010 (Microsoft Inc., Redmond, USA) and SPSS v.22.0 (SPSS Inc., Chicago, USA). Further statistical analysis will be presented and discussed in Zhuang and Ball (in prep).

3 Results and discussion

3.1 Heavy metal spatial and temporal distribution

The QA/QC values for each metal were approximately equal to each CRM value (range 91.3–106.4 %) (Table 2), inferring that the procedures used for soil heavy metal digestion are relatively accurate and reliable. These slight deviations occurring in the procedures may be attributed to incomplete digestion, silicate sample matrices.

The results in Table 3, which are the mean values of Pb and Zn on three separate pre-rainfall samplings and three

Table 2 Validity check for digestion methods

Unit: mg/kg	Pb	Zn
Mean	605	397
SD	45	31
% SD	7.4	7.8
Certified value ^a	636	373
% recovery	95.1	106.4

Recovery rates and repeatability standard deviation for the determination of heavy metals in Certified Reference Material DC73309; concentrations of element are given in milligrams per kilograms (ppm) of dry weight; $n = 16$

SD standard deviation; % SD percentage of standard deviation

^a Certified values from certified given values

separate post-rainfall samplings, can be summarized as follows: The concentrations of Pb and Zn in all the locations decreased with distance from the road. This trend not only occurred in the topsoil but also in the subsoil. The highest concentration of Pb and Zn are all found at a distance of 1 m from the road. There was a percentage change of Pb concentration from pre-rainfall to post-rainfall at a distance of 1 m for the topsoil at location AB of 16.5 %, CE 12.5 %, and D 1.5 %. The percentage change at a distance of 1 m for the subsoil is AB 9.3 %, CE 20.3 %, and D 13 %. The results in Table 3 also indicate that a percentage change in Zn concentration from pre-rainfall to post-rainfall at a distance of 1 m for the topsoil at location AB of 4.4 %, CE 9.4 %, and D 15.9 %. The percentage change at a distance of 1 m for the subsoil is AB 6.0 %, CE 14.5 %, and D 16.9 %. The corresponding concentrations of Pb and Zn are different for soil derived from Hawkesbury Sandstone (AB), Wianamatta Shale (CE), and Mittagong Formation (D). From Table 3, similar comparisons can be made for topsoils and subsoils at distances of 5 and 10 m from the road. As the field study period post-dates the introduction of lead-free petrol and the subsequent banning of leaded petrol, the elevated levels of Pb and Zn concentrations were considered to be the results of weathering of crash barriers and abrasion of vehicular bodies; this is consistent with the published literature (Davis et al. 2001; Legret and Pagotto 2006; Loganathan et al. 2013; Karim et al. 2014). The results also indicate the concentrations of metals substantially decrease with depth in the soil profile. The influence of soil depth on mean concentrations of heavy metals agreed with many studies (Imperato et al. 2003; Yang et al. 2012; Curran-Cournane et al. 2015). Yang et al. (2012) reported that the concentrations of Pb, Zn, Cu, Hg, and As were substantially greater in topsoil (0–10 cm) than subsoil (20–40 cm) along two districts crossing the Yangtze River, China. The low concentration in the subsoil may be a result of low mobility of these analytes in soil (Curran-Cournane et al. 2015).

Table 3 Summary of measured mean value for lead and zinc isotope ratios of roadside soil samples in Miranda Park, Sydney (unit: mg/kg dry weight)

Sampling profile layer	Sampling time	Distance from road (meter)	Pb			Zn			
			Sampling location			Sampling location			
			AB	CE	D	AB	CE	D	
Topsoil	Pre-rainfall	1	103.63	159.32	137.50	223.41	254.12	264.23	
		5	47.36	56.31	50.74	94.53	100.32	113.21	
		10	21.15	16.21	17.35	42.36	33.64	51.32	
	Post-rainfall	1	78.87	139.09	135.69	213.35	230.24	222.36	
		5	44.14	42.32	55.18	97.34	103.24	101.42	
		10	19.24	20.34	24.31	39.32	39.35	45.62	
	Subsoil	Pre-rainfall	1	86.08	103.23	97.35	164.78	138.54	142.64
			5	36.41	40.21	48.61	66.24	55.34	71.23
			10	18.21	19.16	22.74	27.35	24.36	33.21
Post-rainfall		1	94.32	124.32	110.34	174.33	158.64	166.21	
		5	47.11	44.27	57.41	70.31	62.31	77.41	
		10	20.74	24.12	27.15	23.32	30.74	27.54	

The concentration of Zn is greater than the concentration of Pb in all the three different soil types. The highest concentration of Pb was in Wianamatta Shale-derived soil, 159.32 mg/kg and the highest concentration of Zn, 254.12 mg/kg occurred in Mittagong Formation all at a distance of 1 m from the road. The rainfall would change the metal concentration in subsoil; however, the rainfall reduced the metal concentration in the topsoil in all the locations. This could be explained by the rain leaching the metal deep into the soil profile. For the topsoil, the rainfall has been shown to produce the most significant percentage change in Hawkesbury Sandstone-derived soil for Pb (23.89 %) and Mittagong Formation-derived soil for Zn (15.85 %) at a distance of 1 m. On the contrary, for the subsoil, due to heavy metal leaching by rainfall from the upper layer, the concentrations of both Pb and Zn increased substantially. The greatest percentage change for Pb is 20.43 % and Zn is 19.97 % in location CE (Wianamatta Shale-derived soil). The high level of metal concentration may be the result of the higher clay content in the subsoil of Wianamatta Shale which has the greatest clay content (more than 40 %); the clay plays a critical role in soil absorption of heavy metals.

3.2 Chemical portioning of heavy metals in soil

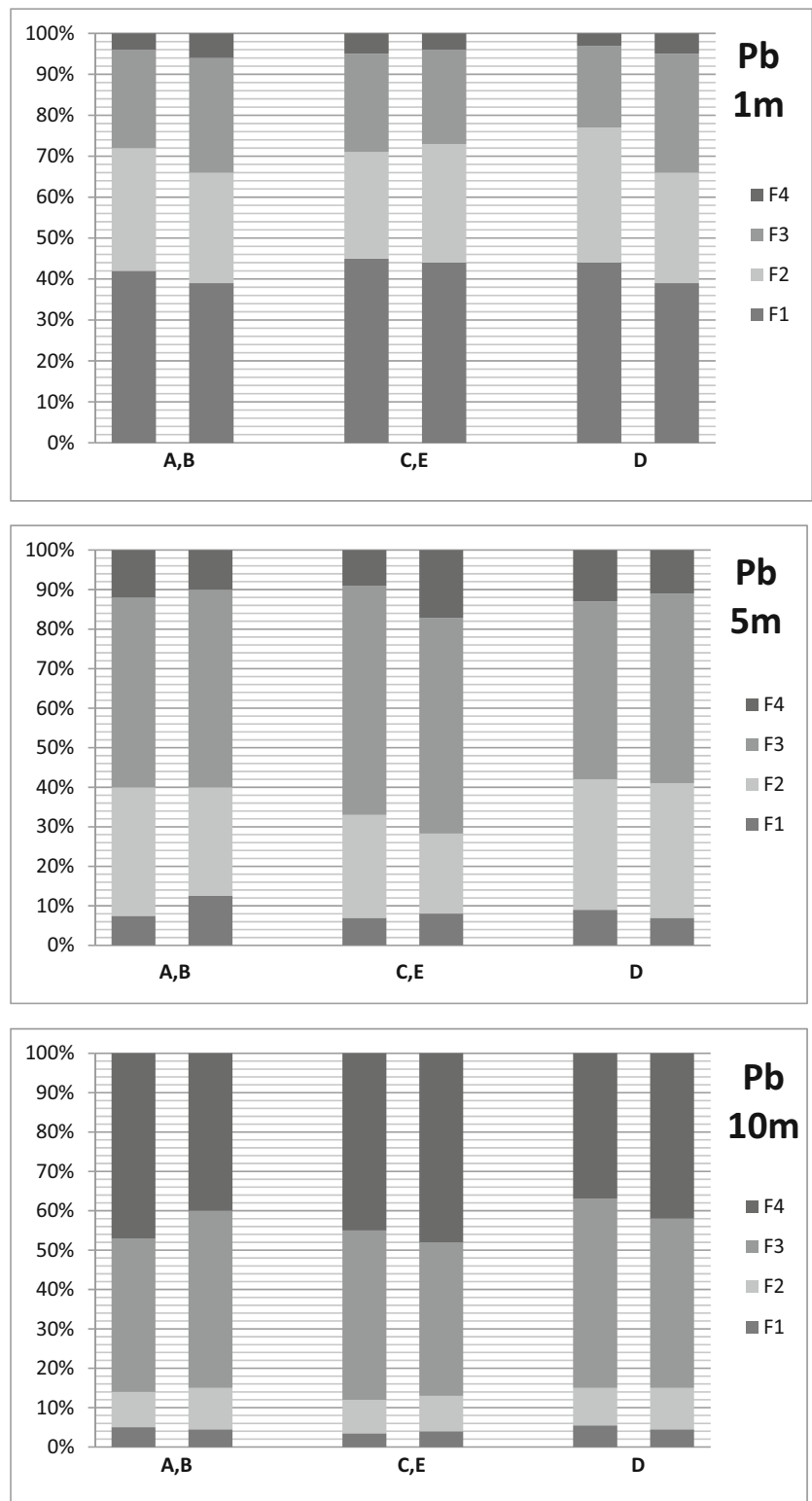
The general metal partitioning form of Pb and Zn in the topsoils of the three different soil types sampled at a distance of 1, 5, and 10 m from the road was obtained from the sequential chemical extraction (BCR) process. These results are shown in Figs 2 and 3. The BCR results are generally described as the percentage distribution of each fraction. The chemical-partitioning pattern showed the variation among individual

soil samples. However, the averaged partitioning patterns of both Pb and Zn can reflect several variations with regard to different soil types and also can indicate several clear changes as a result of distance from the road and rainfall influence.

3.2.1 Lead

The percentage of exchangeable fraction (F1) of Pb decreases with an increase in the distance from the road. The percentages of F1 decreased from approximately 40 % down to approximately 5 %. At a distance of 1 m, there is a clear reduction of exchangeable fraction in all locations in the post-rainfall period. This could explain the reduction of total Pb concentration in all the locations. The exchangeable fraction stands for the metal mobility in order to impact on the metal movement in the soil profile. In contrast, the oxidizable fraction (F3) and residual fraction (F4) predominantly come from the parent material. The lead in the roadside soil is strongly associated with organic matter and Fe-Mn oxide phases (ca. 70 %) with small amounts in the residual and carbonate fraction at a distance of 5 m from the traffic. The percentage of exchangeable Pb is very low (ca. 5 %) in roadside soil in all these three locations at a distance of 10 m from the roadside. However, there is a significant exception occurring in location AB at a distance of 5 m from the roadside where the exchangeable Pb percentage increased significantly. From Fig. 2, the rainfall significantly influenced F1 at a distance of 1 m from the roadside, because at this distance, the roadside soil is critically impacted by the deposition from vehicles onto the road surface as dust in the form of aerosols. Moreover, the rainfall changed the exchangeable fraction especially at distance 1 and 5 m from the roadside.

Fig. 2 The mean lead BCR results of three times pre-rainfall sampling and three times post-rainfall sampling in three different locations at distance of 1, 5, and 10 m are shown. The mean chemical partitioning is shown in percentage. The *left columns* indicate the pre-rainfall results and the *right columns* indicate the post-rainfall results. *F1* exchangeable and weak acid soluble fraction (soluble and carbonate fraction); *F2* reducible fraction (associated to Fe-Mn oxides); *F3* oxidizable fraction (associated to organic matter); *F4* residual fraction

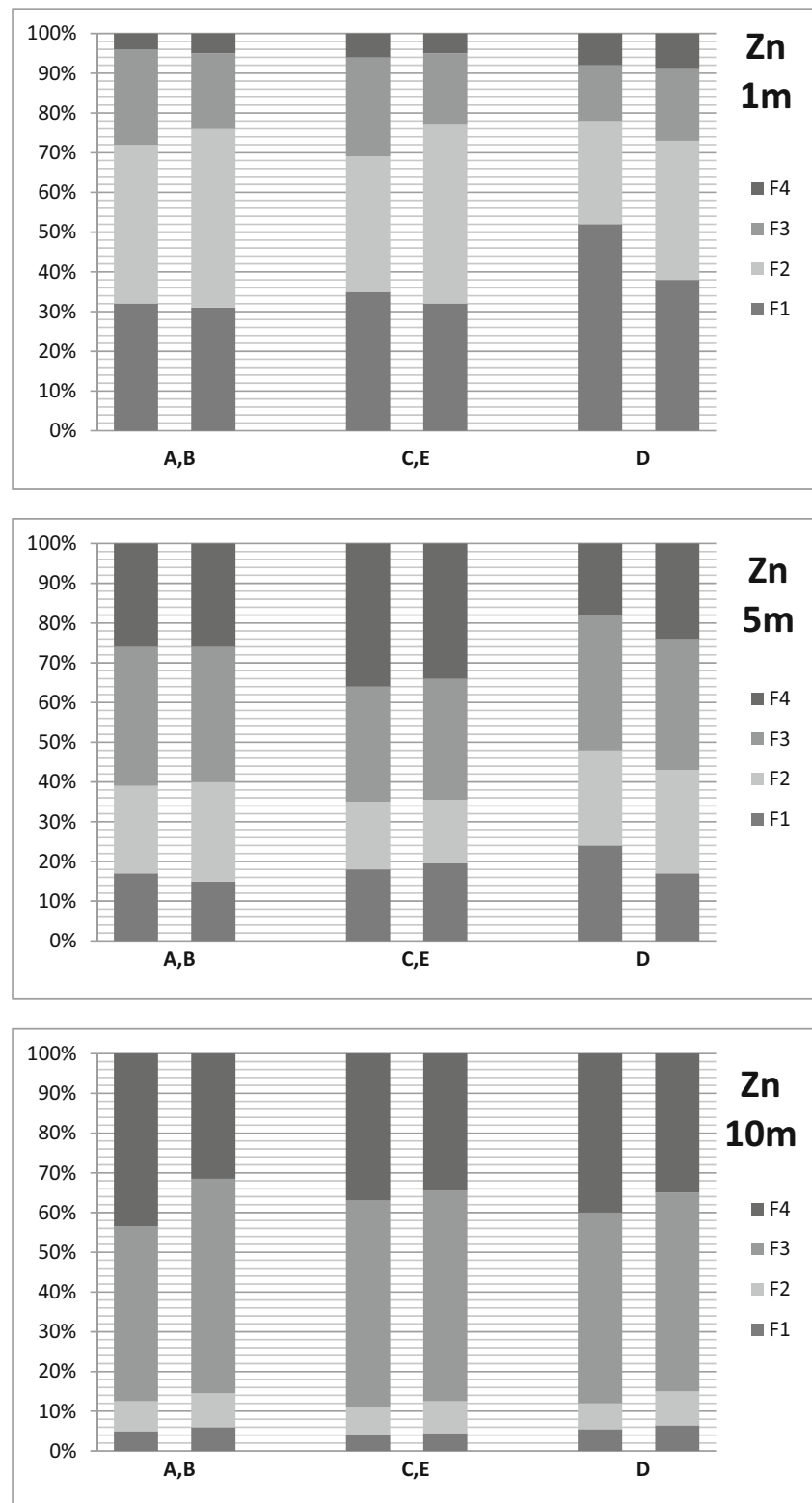


3.2.2 Zinc

In this study, according to Fig. 3, at a distance of 1 m from the roadside, the exchangeable fraction varied from approximately 30 to approximately 52 %. In the comparison between

different soil types, the significant reduction of F1 was found in location D. Similarly, the most significant reduction of total Zn concentration was also in this soil type. However, at a distance of 5 m from the roadside, the predominant fraction for Zn is the Fe-Mn oxide phase (ca. 40 %). The next

Fig. 3 The mean zinc BCR results of three times pre-rainfall sampling and three times post-rainfall sampling in three different locations at distance of 1, 5, and 10 m are shown. The mean chemical partitioning is shown in percentage. The *left columns* indicate the pre-rainfall results and the *right columns* indicate the post-rainfall results. *F1* exchangeable and weak acid soluble fraction (soluble and carbonate fraction); *F2* reducible fraction (associated to Fe-Mn oxides); *F3* oxidizable fraction (associated to organic matter); *F4* residual fraction



important fractions are the exchangeable fractions, (ca. 30%). The exchangeable Zn in location D in the post-rainfall period is above 50%, which may need further study to investigate its bioavailability and ecological implications. At a distance of

10 m, the Zn chemical composition is very similar to Pb; this implies the metal concentration is mostly contributed from the parent materials and other stable forms. Furthermore, comparing the chemical fraction composition at different distances,

the total Zn elevation is mainly due to the increase of exchangeable fraction and it also could be influenced by rainfall.

4 Conclusions

The present study was focused on the concentrations of Pb and Zn in relation to soil types and rainfall events. It was found that the concentrations of both Pb and Zn showed a clear negative correlation with distance and soil depth in all soil types as determined in many other previous studies. However, there are differences in the results between the soil types. The heavy metal concentrations at the same distance in different soil types are different, particularly at the distance of 1 m. Also the rainfall events do influence the heavy metal deposition differently in both topsoil and subsoil of the three soil types. The results of BCR indicate the Pb and Zn input by traffic are mainly as the exchangeable fraction in the topsoil because the percentage of exchangeable fraction also decreases with increase in distance. The rainfall could importantly decrease the percentage of exchangeable fraction (F1) in topsoil and that could explain the total metal concentration reduction after rainfall. The F1 of both Pb and Zn has a similar percentage at a distance of 10 m in this study. The concentrations of Pb and Zn have regressed to the background level at 10 m. The concentration of exchangeable fraction tends to be very low at a distance of 10 m from the BCR results for all the soil types. The rainfall would carry the metals into a deeper layer; as a result, the total heavy metal concentration decreased in the topsoil and increased in the subsoil due to the rainfall event. However, different soil types have different ability to reflect this variation. Therefore, there is a relationship between heavy metal concentrations, the influence of rainfall events on heavy metals deposition and movement and soil type.

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