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Spatial distribution of metals in soils in Baltimore, Maryland: Role of native parent material, proximity to major roads, housing age and screening guidelines

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Spatial distribution of metals in soils of an older US city (Baltimore) was affected by parent material, proximity to major roads, and housing age.

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ABSTRACT

We investigated the spatial distribution of heavy metal above-background (anthropic) contents of Cd, Co, Cu, Cr, Fe, Mn, Ni, Pb, Ti, V, and Zn in Baltimore City surface soils and related these levels to potential contaminating sources. Composite soil samples (0–10 cm depth) were digested using a nitric and hydrochloric extraction technique. Slightly more than 10% of plots exceeded United States Environmental Protection Agency screening guidelines for Pb. In a principal component analysis, the first component corresponded to Co, Cr, and Fe, which are constituents of local mafic rocks. The second component corresponded to Cu, Pb, and Zn which were significantly higher within than beyond a 100 m buffer of the major roads within the city; furthermore, Pb and Zn were higher in older residential lots.

1. Introduction

Soils in urban areas are contaminated by heavy metals (Wong et al., 2006). Since urban landscapes are complex in nature and have numerous sources of heavy metals, assessing the extent and degree of metal contamination presents a challenge to researchers and urban planners (Thornton, 1991; Mielke, 1999; De Kimpe and Morel, 2000). Heavy metal contamination may result indirectly from urban environmental sources (e.g., atmospheric deposition) or directly from management inputs (e.g., fertilizer or pesticide use), and waste disposal. The consequent pattern is a conglomeration of both effects, which result in a heterogeneous distribution of heavy metal concentrations in urban-influenced soils (Pouyat and Effland, 1999).

Regional patterns of heavy metal concentrations can be explained by geochemical properties of mineral soil and parent material, plant species composition, and atmospheric deposition (Friedland et al., 1986; Grigal and Ohmann, 1989; Bityukova et al., 2000). In the northeastern United States, regional studies of heavy metal contamination in forest soils showed a significant relationship between Pb concentrations and proximity to major urban areas (Andresen et al., 1980; Johnson et al., 1982), whereas variations in the amounts of Zn, Cu, Ni, and Cd did not appear to be related to atmospheric patterns of deposition except near point sources of pollution (Friedland et al., 1986). At the scale of a major metropolitan area, Pouyat and McDonnell (1991) and Airola and Buchholz (1984) reported increasing levels of Cu, Ni, and Pb in forest soils going from rural to urban areas in the New York City metropolitan area. Pouyat and McDonnell (1991) collected samples beyond the "splash zone" of roads and therefore concluded that the accumulation of Cu, Ni, and Pb in forest patches occurred primarily from regionally deposited atmospheric particulates derived from automobile and industrial sources. Others have found similar results in forest patches along other urban–rural gradients (Parker et al., 1978; Pavao-Zuckerman, 2003).

Elevated heavy metal concentrations are almost universally reported and often with high variances (Thornton, 1991; Li et al., 2004; Wong et al., 2006) for "urban soils" which, may be disturbed or transported soils. Most of the heavy metal sources at a city scale have been associated with roadside environments (Van Bohemen and Janssen van de Laak, 2003; Zhang, 2006), interior and exterior

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paints (Trippler et al., 1988), refuse incinerators (Walsh et al., 2001), industrial stack emissions (Govil et al., 2001; Kaminski and Landsberger, 2000), management (Russell-Anelli et al., 1999) and industrial waste (Schuhmacher et al., 1997). In Baltimore City (heretofore referred to as Baltimore), Pouyat et al. (2007) reported that without accounting for background levels of parent material, soil concentrations of 12 heavy metals were highly variable and generally did not correspond to land use or cover. The authors did find a significant relationship between mafic rock types and Al, Fe, Mg, Mn, Ni, Ti, and V concentrations, suggesting the importance of native parent material to the spatial distribution of these metals in Baltimore.

In this study, we investigated the spatial distribution of the above-background contents of the heavy metals Cd, Co, Cu, Cr, Fe, Mn, Ni, Pb, Ti, V, and Zn in surface soils in Baltimore and related these levels to potential contaminating sources. Specifically, using GIS mapping techniques in combination with principal component analysis (PCA) and univariate statistical procedures, we (1) related above-background levels of heavy metals to their proximity to major roads, the United States Environmental Protection Agency's (EPA) Toxic Release Inventory (TRI) emissions data, and age of housing stock and (2) developed maps using principal component scores to identify areas of heavy metal contamination in Baltimore. The TRI is a publicly available EPA database that contains information on toxic chemical releases and other waste management activities reported annually by regulated industry groups and federal facilities. We used above-background contents to spatially assess contamination of heavy metals. The above-background contents, which were not directly measured in this study, account for differences in bulk density and mineral content of parent materials that occur in the Baltimore. Kriging and PCA have been used successfully to determine the spatial distribution and relationships of metals with urban environmental factors (Facchinelli et al., 2001; Li et al., 2004). In addition, to assess the potential health effects of the measured soil heavy metals, we compared measured median concentrations of these metals with existing guidelines published by EPA (soil screening values) and The Netherlands (target values). The Netherlands guidelines were used to provide more stringent soil elemental concentration standards.

A previous investigation of garden soils in Baltimore region showed that Pb concentrations were related more to road density than to the age of housing stock, the latter of which was originally attributed to the use of Pb-based paint (Mielke et al., 1983). Similar to the results found by Mielke et al. (1983), we predicted that soils sampled in land use types other than urban gardens will have Pb levels related to vehicular traffic despite reductions in Pb emissions since the mid-1980s. Soil Pb signatures should persist due to the long residence times of Pb in urban soils (Yesilonis et al., 2007; Mielke et al., 1983; Mielke, 1999; Semlali et al., 2004) and the potential for particulates of Pb to persist and to be redistributed as street dust for decades after the source emission (De Miguel et al., 1997). However, by expanding the scope of investigation to soils other than those found in urban gardens, we expected other potential sources, such as industrial emissions and waste disposal, to show a relationship to soil Pb levels in Baltimore. Other metals, such as Cu, Cr, Zn, and Ni also have been found at elevated levels in Baltimore, but anthropic levels of these metals have not yet been related spatially to potential sources (Pouyat et al., 2007).

2. Materials and methods

2.1. Study area

Baltimore is a historically industrial city that is located on the Chesapeake Bay in the Mid-Atlantic region of the USA (Fig. 1). The city is at the junction of US interstate highways 70 and 95 and has 25 facilities reporting chemical atmospheric releases for metal and metal compounds from 1987 to 1995, totaling an estimated 197 tonnes of stack emissions of various pollutants (www.epa.gov/triexplorer/). The Toxic Release Inventory (TRI) sites were considered because they accounted for all metals being investigated (Ti, Cr, Mn, Co, Ni, Cu, Zn, Pb, Cd, V, and Fe).

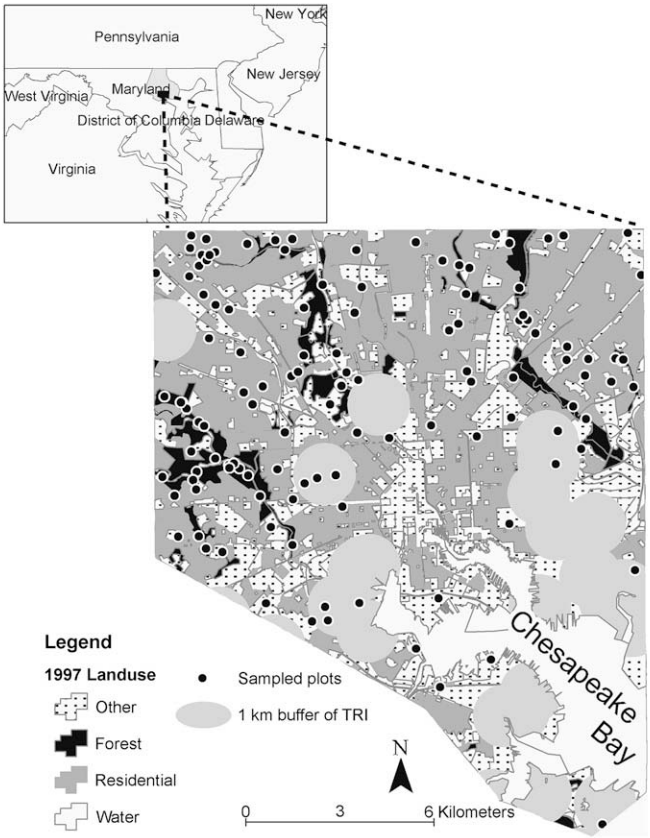

Fig. 1. Sampling locations, road buffer strips, and a subset of Toxic Release Inventory (1-km buffer circles) of Baltimore, MD, USA.

As with all USA cities that developed during the industrial revolution, emissions from industrial sources have decreased substantially since the enactment of the Clean Air Act and the concurrent loss of manufacturing industries since the early 1970s. The remaining industrial sites currently in Baltimore tend to be clustered around the harbor and major waterways (Fig. 1).

Baltimore lies on the Chesapeake Bay along the boundary between two physiographic provinces: the Piedmont Plateau and the Atlantic Coastal Plain (Hunt, 1967). Sheer relief is caused by the physiographic province boundary between the Piedmont and Coastal Plain and often results in waterfalls. These falls occur from Alabama to New Jersey, and this boundary is commonly referred to as the "Fall Line". The north–northeast trending Fall Line separates the two provinces, dividing the city approximately into half. Most of the city is characterized by nearly level to gently rolling uplands, dissected by narrow stream valleys. The Piedmont Plateau in the city is underlain by mafic and ultra-mafic rock types (Crowley and Reinhardt, 1979). The Coastal Plain in the city is underlain by much younger, poorly consolidated sediments. Soils in the Coastal Plain of the city are very deep, somewhat excessively drained to well-drained upland soils that are underlain by either sandy or gravelly sediments or unstable clayey sediment. Soils in the Piedmont Plateau of Baltimore are very deep, moderately sloping, well-drained upland soils that are underlain by semi-basic or mixed basic and acidic rocks (NRCS, 1998). Highly disturbed soils make up more than 60% of the land area of the city (Pouyat et al., 2002).

2.2. Study design and laboratory methods

Plots were located within Baltimore by a stratified random sampling design using land use and cover: forest, residential, and other (Fig. 1). The "other" land use category includes commercial, industrial, institutional, extractive, open urban land, brush, wetlands, bare ground, and transportation". These plots were established in 1999 as part of the Baltimore Ecosystem Study (Pickett et al., 2001) to collect data needed to calibrate the Urban Forest Effects (UFORE) model (Nowak et al., 2004). As previously mentioned, analysis of all soil data collected in an earlier study showed that heavy metal concentrations at a 0–10 cm depth did not correspond to these land-use cover type designations (Pouyat et al., 2007). Therefore, for the purposes of this study, we used the original randomly stratified design to consider the spatial relationships of the 122 plots with other potential sources.

Following Nowak et al. (2004), a circular plot with a radius of 11.35 m (0.04 ha) was re-established at each sample location. Two techniques were used to acquire soil samples within the plots: (1) three undisturbed 5-cm diameter by 5-cm depth cores were used to measure bulk density (bulk density cores were only taken at 0–5 cm and not 5–10 cm) and (2) a composite soil sample (0–10 cm depth) was obtained with a 2-cm diameter stainless steel sampling probe. If present, the depth of organic horizon was recorded but was omitted from the sample. Each composite sample was air dried and sieved in the laboratory with a stainless steel 2-mm mesh sieve. The undisturbed cores were used to determine soil bulk density (Blake and Hartge, 1986).

For heavy metal analysis, the 0–10 cm composite samples were digested at the Baltimore Ecosystem Study and University of Maryland Baltimore County Lab using a nitric and hydrochloric extraction technique (modified EPA method 3050B; US EPA, 1996). With this technique a subsample of 2.0 g ± 6–8% (2.12 g–2.16 g) of soil was placed into a beaker and 10 mL of 1:1 HNO_3 was added and covered with a watch glass. The samples were heated to 95 °C ± 5 °C and refluxed for 10–15 min without boiling. The samples were cooled, and 5 mL of concentrated HNO_3 was added and refluxed for 30 min. Again, the samples were cooled, and 5 mL of concentrated HNO_3 was added and refluxed for 30 min and then allowed to sit for 24 h. After 24 h 2 mL of water and 3 mL of 30% H_2O_2 were added to the solution and then heated. Seven millilitres of 30% H_2O_2 in 1-mL aliquots were added with warming. The acid–peroxide digestate was heated at 95 °C ± 5 °C without boiling for 2 h. Ten millilitres of concentrated HCl were added to the sample and heated for 15 min at 95 °C ± 5 °C. The digested material was passed through a #42 Whatman filter and stored in polyethylene bottles. These extracts were sent to the Cornell University Nutrient Analysis Laboratory to determine acid-soluble elements with an Inductively Coupled Plasma Atomic Emissions Spectrophotometer (model: SPECTRO CIROS CCD-CIRcular Optical System).

2.3. Statistical analysis

Depending on the statistical analysis, elemental data are presented as a concentration per oven dry-weight of soil ($\mathrm{mg\,kg^{-1}}$) or as acid-soluble element contents ($\mathrm{kg\,ha^{-1}}$) to a depth of 10 cm, the latter of which accounts for differences in bulk density. Elemental data were also interpreted by subtracting out background levels of individual heavy metals, heretofore referred to as anthropic levels. Anthropic contents assess the amount of “contamination”, and measured concentrations reflect potential exposure to humans.

Anthropic levels ($\mathrm{Conc_{anthropic}}$) were calculated by subtracting the Anticipated Typical Concentration ($\mathrm{Conc_{ATC}}$) from the measured soil acid-soluble metal concentration ($\mathrm{Conc_{sample}}$)

$$\mathrm{Conc_{anthropic}} = \mathrm{Conc_{sample}} - \mathrm{Conc_{ATC}}$$

$\mathrm{Conc_{ATC}}$ represents the mean concentration plus one standard deviation for values obtained from the State of Maryland Department of the Environment’s (MDE) Cleanup Standards for Soil and Groundwater, August 2001 (Table 1). The ATC represents a value that is equal to or greater than the majority of background concentration samples.

MDE determined “background level” by statistically evaluating previously collected soil trace metal concentration data. The state was divided into three provinces based on differences in geologic formations: Eastern Maryland, Central Maryland, and Western Maryland. The Central Maryland background values were used for west of the Fall Line and the Eastern Maryland background values for east of the Fall Line.

Table 1

Mean heavy metal concentrations ($\mathrm{mg\,kg^{-1}}$) of major rock types [a] and the Anticipated Typical Concentrations of Central and Eastern Maryland

Element	Geologic rock type ($\mathrm{mg\,kg^{-1}}$)			Maryland province soils	
	Ultra-mafic	Mafic	Sandstone	Central Maryland[b]	Eastern Maryland[b]
Cd	0.12	0.13	0.05	1.1	0.73
Co	110	35	0.3	33	11
Cr	2980	200	35	30	28
Cu	42	90	30	42	12
Fe[c]	94 000	–	9800	26 000	15 000
Mn	1040	1500	460	1 400	480
Ni	2000	150	9	22	13
Pb	14	3	10	61	45
V	40	250	20	35	30
Zn	58	100	30	73	63

Dash (–) indicates no values were reported for the element.

[a] Adapted from Krauskopf (1967) and Rose et al. (1979).

[b] Anticipated Typical Concentrations (MDE, 2001).

[c] Cannon (1978).

As an exploratory and data reduction technique, the elemental data were first submitted to a PCA factoring in a correlation matrix using the SAS package (SAS Institute, version 8.0, 2003). A PCA can take several soil properties and express them in terms of a smaller number of common components (Pielou, 1984). The first principal component (PC1) explains the maximum possible variance of the data set, the second component (PC2) explains the maximum variance subject to being uncorrelated with PC1, the third component (PC3) explains the maximum variance subject to being uncorrelated with PC1 and PC2, and so on (Usher, 1976).

We ran the PCA using the content data ($\mathrm{kg\,ha^{-1}}$) of Cd, Cu, Co, Cr, Fe, Mn, Ni, Pb, Ti, V, and Zn representing either the measured (includes background) or anthropic (excludes background) contents of each sample. We used a kriging procedure in ArcMap™ (ESRI®, 2004) to interpolate the most explanatory first two principal component scores, allowing visualization of the spatial distribution of heavy metal variance corresponding to each component (e.g., Lin and Chang, 2000).

Potential contaminant sources were statistically tested between individual elemental variables using non-parametric Wilcoxon Two-Sample Test and the most explanatory principal components using Tukey’s *t*-test. Comparisons were made between anthropic contents of soils between plots located within and outside buffer zones from major roads (100 m) and TRI stack emission locations (1 km) (Fig. 1). We chose the 100-m road buffer based on studies reporting that most contamination from automobile exhaust occurs up to 100 m from the edge of a roadway (Adriano, 1986). The buffered roads within Baltimore were selected using the 2000 Census Tiger Line Data for the Baltimore Region road features. The buffer was created around the following roads: primary roads with and without limited access (includes interstate and US highways), secondary and connecting roads, and state highways. The TRI buffer distance was determined to be 1 km from an individual stack, which was based on findings of smelter studies that typically showed substantial decreases in soil contamination beyond 1 km from the emission source (e.g., Hutchinson and Whitby, 1974; Godin et al., 1985; Martley et al., 2004).

Two guidelines were used to evaluate the potential public health risk of soil contamination: The Netherlands and EPA. The Netherlands target values reflect concentrations to be achieved to fully recover the functional properties of the soil for humans, plants, and animals (VROM, 2000; Swartjes, 1999), and EPA screening values are risk-based concentrations derived from standardized equations combining assumptions on exposure with EPA toxicity data of human health (http://www.epa.gov/superfund/resources/soil/fact_sht.pdf). Frequency distributions were created to determine the proportion of plots to exceed both guideline values for measured concentrations.

3. Results

Measured and anthropic Pb concentrations of heavy metals varied widely in the Baltimore landscape. The distribution of all metals is right-skewed with the mass of the distribution concentrated to the left, i.e. low elemental concentration values (Table 2). Even with skewed distributions, contamination of at least one metal has occurred for most of the plots sampled in this study. For example, at least 50% of the plots had anthropic and measured Pb concentrations of 60.5 and 89.3 $\mathrm{mg\,kg^{-1}}$ or greater, respectively. In fact, all of the metals showed contamination at the 50th percentile with Cr, Cu, Fe, Ni, Pb, and Zn showing above-background levels at the 25th percentile (Table 2).

Frequency distributions of measured metal concentrations showed that levels often exceeded guidelines established by the EPA and the Netherlands (Fig. 2). Cadmium and Pb had the greatest number of plots in excess of the Netherlands target values by 54.9 and 50.8%, respectively, while 10.7% of plots exceeded EPA screening guidelines for Pb (Fig. 2). Likewise, Cr exceeded the Netherlands guidelines in 17.2% of plots, and 0.82% exceeded EPA screening guidelines. Of the remaining guidelines available for comparison, concentrations of Co, Ni, and Zn exceeded the Netherlands guidelines in 23.8, 28.7, and 22.1%, respectively, of the plots measured.

3.1. Inter-metal comparisons

A clear relationship was discernable in the PCA for both anthropic and measured heavy metal contents ($\mathrm{kg\,ha^{-1}}$ at 0–10 cm) of soils in Baltimore (Table 3). For the anthropic contents, almost 74% of the variation was explained by the first three components of the PCA, with PC1 (eigenvalue of 3.8) accounting for 38% of the variation (Table 3A). Eigenvalues correspond to the elements

Table 2
Percentile concentrations (mg kg^{-1}) of anthropic concentrations and concentrations with background levels

	Element (mg kg^{-1})									
	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn
Anthropic concentrations										
Minimum	0.003	0.004	0.004	0.009	0.064	0.008	0.030	0.013	0.011	0.005
10th percentile	0.003	0.004	3.00	3.44	0.064	0.008	1.36	1.49	0.011	2.38
25th percentile	0.003	0.004	9.31	9.28	1710	0.008	3.92	17.8	0.011	18.0
50th percentile	0.400	0.738	21.2	18.9	6220	0.008	8.07	60.5	5.56	42.8
75th percentile	1.06	6.27	49.4	36.1	13 100	172	15.6	154	20.5	97.8
90th percentile	1.56	13.5	152	61.8	20 700	382	35.9	375	46.3	266
Maximum	2.53	50.3	779	195	44 600	1520	324	5620	91.9	1080
Measured concentrations										
Minimum	0.003	0.452	8.30	6.41	6540	5.59	4.85	4.07	6.81	5.61
10th percentile	0.245	3.92	21.2	18.1	11 600	124	9.52	30.2	14.1	39.9
25th percentile	0.470	5.89	25.3	24.9	14 500	200	13.3	46.6	17.8	60.0
50th percentile	0.886	12.1	38.3	35.2	22 300	422	18.4	89.3	31.4	80.7
75th percentile	1.57	19.4	69.4	53.5	29 300	615	27.3	181	46.5	131
90th percentile	2.10	28.0	172	80.8	38 700	904	44.8	402	72.3	298
Maximum	3.12	65.3	794	216	62 600	2130	336	5650	118	1110

which a factor represents. For example, a factor associated with an eigenvalue of 3.8 indicates that the factor accounts for as much variance in the data as four variables (Kachigan, 1991). A slightly higher percentage of the variance (77%) was explained by the first three components of the PCA when using measured heavy metal data (Table 3B). For both analyses, positive loadings of PC1 corresponded to Co, Cr, and Fe. Anthropic levels of Ni also were related to PC1, while measured contents of Mn and V were related to PC1 (Table 3). All of the metals associated with PC1 are constituents of mafic rock types found in the Baltimore region (Table 1). A kriging of PC1 overlain on the surface geology map of Baltimore (Fig. 3) shows good correspondence between mafic rock types and the measured contents for metals associated with this parent material (Table 1). Fig. 3 is a map of the main component of the Piedmont portion of Baltimore City, Mount Washington Amphibolite, and the Coastal Plain, unconsolidated strata. The Mount Washington Amphibolite is “chiefly uniform, medium- to coarse-grained amphibolite

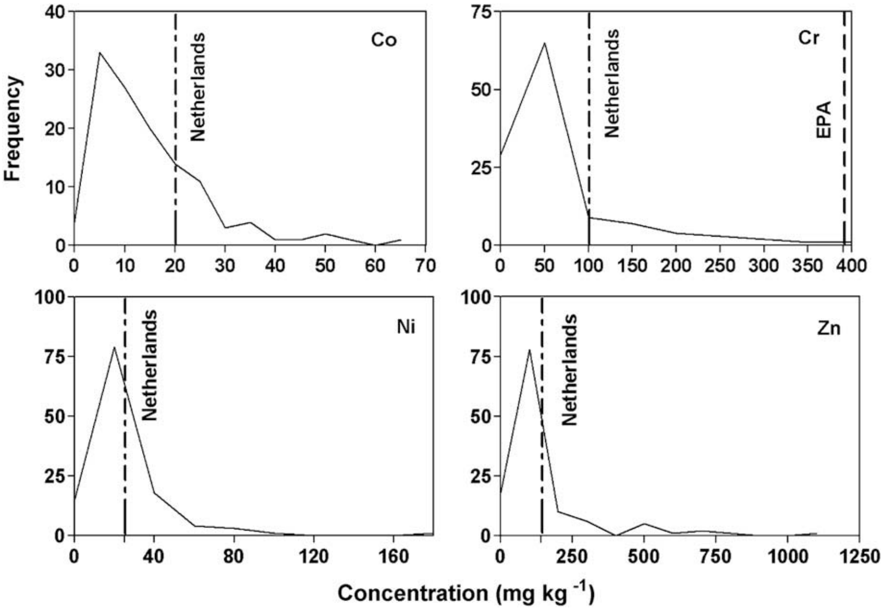

Fig. 2. Plot frequency distributions of metal concentrations (mg kg^{-1}) measured for Baltimore with U.S. Environmental Protection Agency (EPA) soil screening levels and Netherlands soil guideline values. There are no EPA guidelines for Cd, Ni, and Zn because the values were higher than the maximum abscissa concentration.

Table 3
Principal component factor scores and eigenvalues of untransformed (A) anthropic content data (kg ha^{-1}) and (B) measured content data (kg ha^{-1}) including background levels

	Principal component		
	1	2	3
(A) Anthropic content (kg ha^{-1})			
Eigenvalue	3.8	2.4	1.2
% Variance	38	24	12
Element			
Cd	0.39	0.54	*0.57*
Co	*0.87*	−0.34	−0.14
Cr	*0.77*	0.20	−0.33
Cu	0.45	*0.64*	−0.31
Fe	*0.84*	−0.26	0.34
Mn	0.60	−0.23	−0.07
Ni	*0.73*	−0.13	−0.42
Pb	0.11	*0.70*	0.23
V	0.65	−0.35	*0.53*
Zn	0.29	*0.89*	−0.06
(B) Measured content (kg ha^{-1})			
Eigenvalue	4.2	2.3	1.2
% Variance	42	23	11
Element			
Cd	0.35	0.54	*0.62*
Co	*0.91*	−0.29	−0.06
Cr	*0.73*	0.24	−0.38
Cu	0.50	*0.60*	−0.33
Fe	*0.87*	−0.23	0.26
Mn	*0.77*	−0.23	0.13
Ni	0.71	−0.09	−0.48
Pb	0.09	*0.70*	0.24
V	*0.76*	−0.31	0.38
Zn	0.23	*0.91*	−0.07

The eigenvalue corresponds to the number of elements which the principal component represents. The high loadings are italicized.

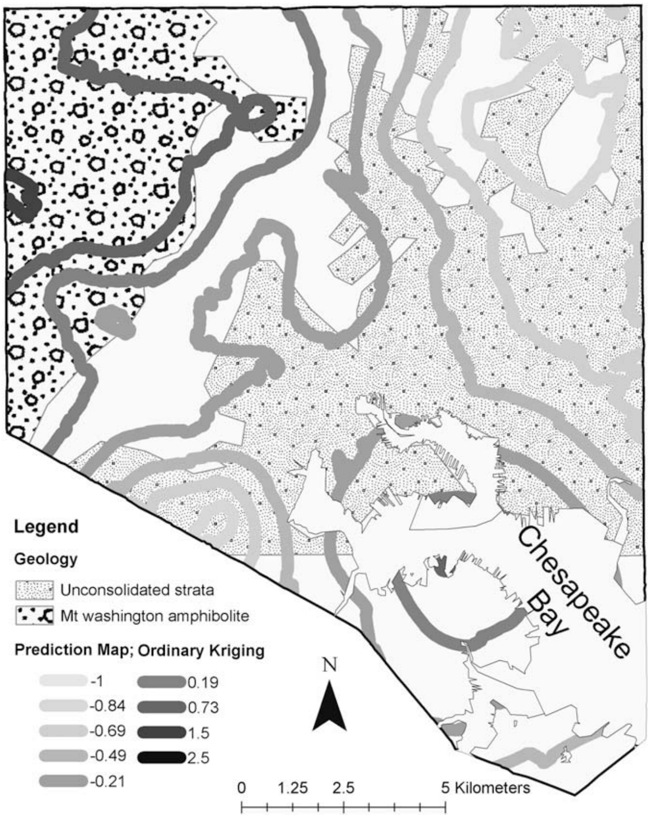

Fig. 3. Ordinary kriging prediction map showing the first principal component (PC1) superimposed over the surface geology of Baltimore. PC1 correlates to the measured content values (kg ha^{-1}) of Mn, Fe, Co, and V.

consisting of plagioclase plus actinolite and/or hornblende" (Crowley and Reinhardt, 1979).

The second principal component explained between 23 and 24% of the variation for the anthropic and measured analyses, respectively (Table 3). In both cases, positive loadings of PC2 corresponded to high contents of Cu, Pb, and Zn (Table 3). A kriging of PC2 overlain on a map of Baltimore (Fig. 4) shows good correspondence between older areas of the city that have a high density of roads and PC2, or anthropic levels of Cu, Pb, and Zn (Fig. 5). For both the measured and anthropic levels, PC3 explained about half the variation explained by PC2, which corresponded to Cd and V for anthropic levels and high contents of Cd for measured levels (Table 3).

Correlations among contents of individual metals were consistent with the PCA (Tables 3 and 4). Anthropic contents of metals associated with mafic rock types (PC1) were positively related (e.g., Co and Mn, $r = 0.77$, $P < 0.0001$; Co and Fe, $r = 0.58$, $P < 0.0001$; and Co and Ni, $r = 0.56$, $P < 0.0001$). Likewise, metal anthropic contents related to PC2 tended to be positively related; Pb was positively correlated to Zn contents ($r = 0.79$, $P < 0.0001$), while Zn was positively correlated to Cu contents ($r = 0.54$, $P < 0.0001$).

3.2. *Relationship to roads, housing age, and tri data*

There was a distinct relationship between anthropic levels of heavy metals and distance to major roads in Baltimore. Anthropic levels were 3.5- and 2.8-fold higher inside the 100-m buffer zone of major roads for Pb and Zn, respectively, and these differences were statistically significant at $P < 0.001$ using a Wilcoxon Two-Sample Test (Table 5). Cu and Zn were closely correlated with Pb contents ($r = 0.49$, $P < 0.0001$ and $r = 0.79$, $P < 0.001$, using Pearson correlation). Likewise, PC2 significantly differed ($P = 0.002$) with higher loadings occurring inside the road 100-m buffer zone. By contrast, V had higher concentrations outside the 100-m buffer zone (Table 5).

The same metals having higher contents inside the 100-m road buffer also were correlated to the year a residential plot was developed. For residential plots, Pb ($r = -0.70$, $P < 0.001$), Zn ($r = -0.55$, $P < 0.0001$), and PC2 ($r = -0.40$, $P = 0.004$) anthropic contents showed a negative correlation with the year of development (Fig. 5). However, when all land-use types were considered, these variables were not statistically related to year of development.

There was no clear relationship between proximity of sample site to TRI stacks using the TRI data reported by the EPA. In fact, TRI data show that Cr ($P = 0.013$), Ni ($P = 0.051$), and V ($P = 0.011$) were all higher outside the 1-km buffer surrounding individual TRI sources (Table 6).

4. Discussion

Although measured soil metal concentrations were reported in a previous study to be unrelated to land use and cover (Pouyat et al., 2007), in the current analysis we found that the measured concentrations were consistently greater than background levels (Fig. 2), which suggest that the sampled sites were affected by human activity. Even though metal concentrations were above-background levels, the skewed distributions suggest that there were relatively few highly contaminated sites (Table 2, Fig. 2), which may in part be due to the relatively few industrial or urban open sites we were given permission to sample ($n = 12$ plots). As a comparison to a study that sampled an area with an industrial history, average

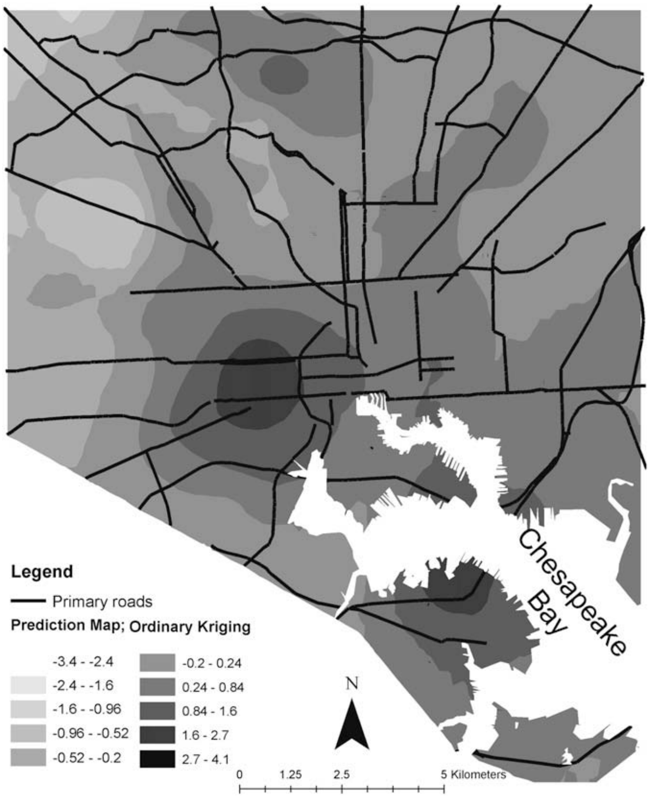

Fig. 4. Ordinary kriging prediction map showing the second principal component (PC2) superimposed over major roads within Baltimore. PC2 correlates to the anthropic content values of Cu, Pb, and Zn (kg ha^{-1}).

concentrations of Cd, Cr, Cu, Ni, Pb, and Zn (no other metals were common between the studies) were considerably lower in this study compared to the western end of East St. Louis, Illinois (Kaminski and Landsberger, 2000). However, compared to other studies (Zhang, 2006; Madrid et al., 2002; Li et al., 2004; Bityukova et al., 2000; Birke and Rauch, 2000; Facchinelli et al., 2001; Pouyat and McDonnell, 1991; Romic and Romic, 2003; Klein, 1972), Baltimore has high soil concentrations of Co, Cr, and Pb (Table 7).

Our comparison of the distributions of measured concentrations of Cd, Co, Cr, Ni, Pb, and Zn to guidelines established by EPA and the Netherlands, suggests that a substantial number of areas in Baltimore are contaminated with levels that are potentially harmful to human health. Almost 11% of the plots sampled had concentrations above EPA's Pb soil screening guideline of 400 mg/kg (Fig. 2). In comparison to the "soil target value" guideline of the Netherlands, more than 50% of the plots exceeded 85 mg Pb/kg. For Cr

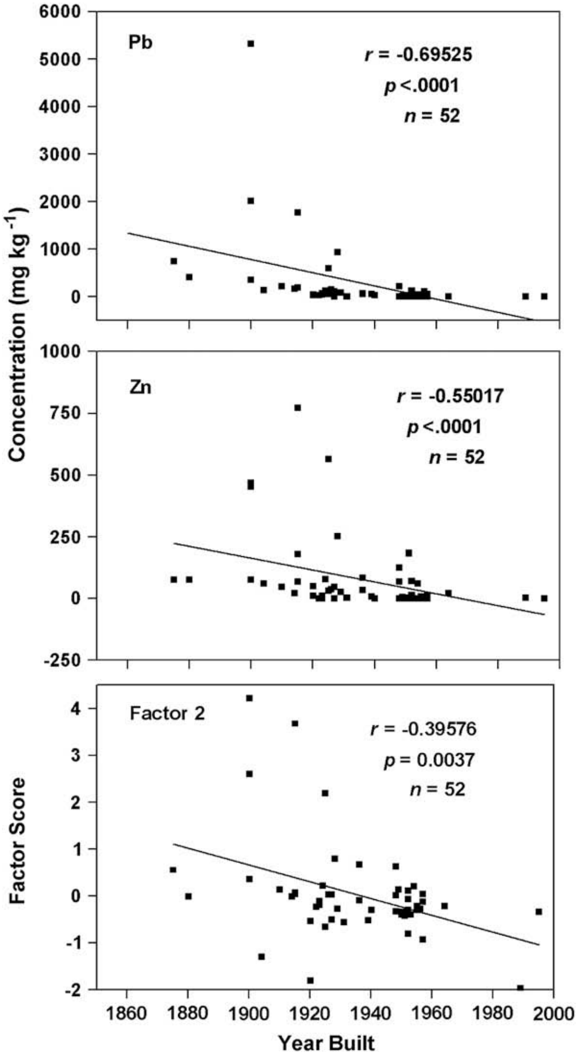

Fig. 5. Spearman correlation coefficients of metal concentrations (mg kg^{-1}) and year structure were built. The data represent anthropic metal data from medium- and high-density residential areas.

Table 4
Spearman correlation matrix of anthropic contents (kg ha^{-1})

	Pb	Cu	Zn	Cr	Mn	Fe	Co	Ni	Cd
Pb									
Cu	0.488								
	<0.0001								
Zn	0.793	0.544							
	<0.0001	<0.0001							
Cr	−0.100	0.0941	−0.0176						
	0.273	0.303	0.848						
Mn	0.00178	0.278	0.0675	0.182					
	0.985	0.0019	0.460	0.0452					
Fe	−0.0607	0.239	0.0567	0.498	0.332				
	0.507	0.0081	0.535	<0.0001	0.0002				
Co	−0.0706	0.334	0.0510	0.367	0.768	0.577			
	0.440	0.0002	0.577	<0.0001	<0.0001	<0.0001			
Ni	0.0625	0.453	0.163	0.605	0.422	0.481	0.561		
	0.494	<0.0001	0.0728	<0.0001	<0.0001	<0.0001	<0.0001		
Cd	0.209	0.262	0.258	0.311	0.246	0.358	0.125	0.273	
	0.0207	0.0035	0.0042	0.0005	0.0062	<0.0001	0.171	0.0024	
V	−0.245	−0.0574	−0.166	0.589	0.191	0.705	0.378	0.302	0.223
	0.0066	0.530	0.0675	<0.0001	0.035	<0.0001	<0.0001	0.0007	0.0135

Table 5
Medians and probability (*P*) values of 100-m road buffer anthropic contents (kg ha^{-1})

Element	Road buffer[a] (kg ha^{-1})		*P* value[b]
	Outside	Inside	
Cd	0.323	0.537	0.0568
Co	0.904	0.004	0.100
Cr	22.2	16.9	0.429
Cu	15.7	23.1	0.0757
Fe	6690	4270	0.109
Mn	0.008	0.008	0.286
Ni	8.24	6.93	0.342
Pb	38.7	134	<0.0001
V	6.98	0.956	0.0429
Zn	33.3	93.4	0.0003
Factor 1[c]			0.962
Factor 2			0.0018
Factor 3			0.556

[a] $n = 93$ and $n = 29$ for plots outside and inside the 100-m buffer, respectively.

[b] The metal *P* values are the Wilcoxon Two-Sample Test, *t* approximation, one-sided Pr > *Z* value.

[c] The *P* values were calculated using the normal Gaussian distribution ANOVA Proc Mixed test.

Table 6
Toxic release inventory site 1-km buffer anthropic medians (kg ha^{-1}) and probability (*P*) values

Element	TRI[a] (kg ha^{-1})		*P* value[b]
	Outside	Inside	
Cd	0.400	0.554	0.396
Co	0.763	0.004	0.133
Cr	25.1	10.3	0.0127
Cu	18.6	21.0	0.370
Fe	6530	5150	0.263
Mn	0.008	0.008	0.430
Ni	8.24	4.27	0.051
Pb	60.7	60.3	0.296
V	6.34	0.011	0.0107
Zn	40.8	89.3	0.0959

[a] The metal *P* values are the Wilcoxon Two-Sample Test, *t* approximation, one-sided Pr > *Z* value.

[b] $n = 113$ and $n = 9$ for plots outside and inside the 1-km buffer, respectively. Factors 1, 2, and 3 were not significant.

contamination, a lower number of plots exceeded EPA and the Netherlands guidelines (5.7 and 17.2%, respectively). For Cd, Co, Ni, and Zn, none of the plots exceeded EPA guidelines; however, the target value guideline of the Netherlands was exceeded by approximately 55, 24, 29, and 22% of the plots for Cd, Co, Ni, and Zn, respectively (Fig. 2). Overall, 16 and 85% of the plots exceeded at least one of the guidelines put forth by the EPA soil screening and the Netherlands target values, respectively. If only the residential plots are considered (total of 52), 19 and 98% of the plots exceeded EPA and the Netherlands values, respectively. These results suggest that the potential for human exposure to heavy metals in Baltimore is high. However, a more complete analysis of exposure factors at higher resolutions and at a shallower sampling depth than that used in this study would be required to assess the risk of human exposure.

Although most of the metals were higher than background concentrations, results of the PCA suggest that the underlying parent material in Baltimore explained much of the variation of the anthropic contents of metals associated with mafic parent material (Table 3A). This result was surprising since we derived the anthropic contents of each metal to calculate the approximate amount of deposition (or contamination) of that particular metal by accounting for the mineral content of the parent material using data collected by MDE. It should be noted that no parent material samples were collected and analyzed by the nitric–hydrochloric acid method. A possible explanation is that the background levels used to calculate anthropic contents were derived from data at a state scale compared to the resolution of our data at the city scale; therefore, in some cases the background levels may have been underestimated. This possible underestimation also may explain why the anthropic and measured levels did not change the PC results, with the exception of one or two metals (Table 3). The fact that metals associated with PC1 (Co, Cr, Fe, and Ni) were particularly skewed to the lower range of the distribution, such that levels above-background did not occur until approximately the 75th percentile (Table 2), implies a low incidence of contamination by these metals.

In areas of the city that are underlain by mafic rock types, contamination could be explained by the ability of these metals to bind onto soil particles and subsequently get distributed throughout the cityscape and thus become concentrated by air and water movement. For example, urban development typically requires major site disturbances and movement of soil material that subject soils to erosional processes. As these materials are redistributed in the landscape, they may accumulate on structural and leaf surfaces, which can later be washed off and accumulated in soil or sediments (De Miguel et al., 1997).

Even though contamination of Co, Cr, Fe, and Ni was apparently low, a few plots had very high above-background contents of Co, Cr, Fe, and Ni near the harbor in the southern industrial sections of the city (Fig. 1). The spatial separation of contamination of these metals between the northwestern part of the city, which is underlain by mafic rock types, and the southeastern part underlain by unconsolidated sediment is likely due to the historic presence of

Table 7
Heavy metal elemental means of data reprinted in the literature

Author	City	*n*	Extraction solution	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	V	Zn
This study	Baltimore, Maryland	122	HNO_3/HCl	1.06	15	72	45	23 495	472	27	231	37	141
Zhang (2006)	Galway, Ireland	–	HF, $HClO_4$, HCl, HNO_3, agua regia	–	6	35	27	2	53	22	58	50	85
Madrid et al. (2002)	Seville, Spain	–	Agua regia	–	–	39	68	20 100	471	22	137	–	145
Li et al. (2004)	Hong Kong	152	HNO_3, $HClO_4$	0.62	3	23	23	–	–	12	95	–	125
Bityukova et al. (2000)	Tallinn, Estonia	198	XRF[a]	–	5	40	45	–	384	–	75	32	156
Birke and Rauch (2000)	Berlin, Germany	3299	EDXRF[b]	–	<5	27	39	–	–	–	79	–	151
Facchinelli et al. (2001)	Italy	50	Agua regia	–	19	46	58	–	–	83	16	–	63
Pouyat and McDonnell (1991)	New York Forest (Urban)	9	HNO_3	–	–	29	34	–	323	24	115	–	69
Romic and Romic (2003)	Zagreb, Croatia	331	Agua regia	0.66	–	–	21	27 041	613	50	26	–	78
Klein (1972)	Grand Rapids, Michigan	70	HNO_3, potassium permanganate	0.41	2	3	8	2 200	–	5	18	–	21

Dash (–) indicates no values were reported for study.

[a] XRF – X-ray fluorescence spectroscopy.

[b] EDXRF – energy-dispersive X-ray fluorescence spectroscopy.

industrial sources (Fig. 3). A significant amount of mafic and ultramafic ore was mined during the 1800s northwest of Baltimore and was transported to Baltimore harbor for processing (Olson, 1997) and may have been a local source of the Co, Cr, Fe, and Ni contamination.

Since empirically measuring industrial emissions were not possible, we used the EPA TRI database to relate industrial pollution sources to soil contamination levels. Our data showed a lack of correlation between TRI stack emissions and anthropic levels of metals in this study (Table 6). To our knowledge, no studies reported in the literature have found a statistically significant relationship between TRI data and soil levels. An earlier analysis of TRI data in Baltimore found relationships between TRI locations and the socio-economic status of current residents (Boone, 2002). The importance of this relationship is unclear since our analysis did not find a statistical relationship between soil metal contents and TRI metal emission data.

There are several potential reasons for the lack of a relationship between stack emission data and soil metal contents. First, the original experimental design was not set up to specifically investigate the relationship between TRI stack emissions and soil metal contamination and lowered the power of the analysis. Second, our analysis did not account for stack height and wind direction nor did it account for local topographic effects (proximity to buildings, landscape variables such as aspect and slope) and thus we assumed an isotropic distribution by using a circular buffer, when the distribution may be anisotropic. Third, addresses used in the TRI database may not represent the physical location of the stack emission but rather represent another location, such as the company's headquarters. Fourth, most of Baltimore's industrial plants were closed when the inventory was established under the Emergency Planning and Community Right-to-Know Act of 1986. Finally, there were only nine plots that fell within the TRI 1-km buffer compared to 113 that fell outside the buffer and that also decreased the statistical power. The small number of samples ($n = 9$) creates a low-power statistical design with increased probabilities of making a Type II error.

The second principal component was correlated highly with Pb and Zn contents (Table 3). The spatial distribution of this component suggests that contamination of these metals is occurring primarily in sections of the city that are older and have high road-traffic densities (Fig. 4). Several studies have reported a similar association of these metals in urban landscapes, suggesting a related source (Bityukova et al., 2000; Facchinelli et al., 2001; Manta et al., 2002; Madrid et al., 2002; Li et al., 2004; Zhang, 2006). Mielke et al. (1983) measured Pb concentrations in garden soils of Baltimore and reported a relationship between the older "inner" city of Baltimore and elevated Pb concentrations. This relationship is consistent with the statistically significant relationship that we determined between Pb content and housing age (Fig. 5). However, the authors suggested that vehicular traffic rather than paint was the primary cause of Pb contamination since housing in inner-city Baltimore was characterized by buildings with unpainted exteriors (brick) and older structures that experienced longer periods of exposure to automobile exhaust than younger structures outside the inner-city area (Mielke, 1999).

Indeed, Cu, Pb, and Zn soil contaminations can be tied to automobile by-products. Lead was an additive to gasoline in the USA until 1986 and was an important component of automobile exhaust (Mielke, 1999), whereas, Cu and Zn contaminations can occur from brake emissions and tire abrasion, respectively (Councell et al., 2004; Hjortenkrans et al., 2006). Emissions of all three metals (50, 220, and 162 tonnes yr^{-1} for Cu, Pb, and Zn, respectively) were higher than other metals measured from automobile use as estimated for the Netherlands in 1999 (Van Bohemen and Janssen van de Laak, 2003). Councell et al. (2004) estimated that 16,612 mg of Zn yr^{-1} are emitted by tire wear in the US. Consistent with these measurements, a number of studies have shown that the contamination of surface soils along or near roads by Pb and the biologically mobile Cu and Zn (Gish and Christensen, 1973; Jim, 1998; Morcelli et al., 2005). Moreover, these metals often are correlated positively in measurements of urban bulk deposition (Fergusson and Stewart, 1992).

A third contamination factor was vanadium, which explained only 12% of the variation in the data set. Possible sources of V in Baltimore include coal burning (fly ash), cement processing, and steel production. Moreover, the estimation of anthropic levels of V may be the result of an error in our estimation of background levels of this element. Estimates of V in parent material for the northwest section of Baltimore ranged from 35 mg kg^{-1} for mafic complex rock to 250 mg kg^{-1} for mafic rock (Table 1).

5. Conclusions

In conclusion, results of this study suggest that urban soils in Baltimore had concentrations that were elevated above-background levels with a large proportion of locations exceeding EPA soil screening or Netherlands soil target values for at least one metal. We propose that contamination was related primarily to vehicle sources as elevated Cu, Pb, and Zn anthropic contents occurred in sections of the city that were older and have high road densities. By contrast, industrial sources appeared to be related to localized soil contaminant sources, such as the harbor area of the city, and not to EPA's TRI emissions data. Finally, our finding that above-background contents of the trace metals Co, Cr, Fe, and Ni were related to parent material was surprising, since we derived the anthropic contents of each metal to calculate the approximate amount of deposition (or contamination) of that particular metal. Higher spatial resolution data of background levels are needed to increase the accuracy of the calculation of anthropic contents of the metals. Besides the potential for industrial sources, we conclude that the apparent contamination by these metals is possibly due to the ability of these metals to bind onto soil particles and subsequently get redistributed by air and water movement throughout the urban landscape.

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