

Research paper

Multi-scale assessment of metal contamination in residential soil and soil fauna: A case study in the Baltimore–Washington metropolitan region, USA



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H I G H L I G H T S

- Soil contamination of metals occurred in residences located in older, more urbanized areas.
- Earthworm burdens correlated with soil Pb and isopod burdens correlated with As, Cr, Ni, Pb and Zn.
- Bird blood Pb correlated with earthworm Pb burdens suggesting a high risk to wildlife.
- The persistence of Pb contamination in urban soils after decades of reduced emissions is alarming.

A R T I C L E I N F O

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This study examined the distribution of metals in residential soils from the scale of a residential yard to a metropolitan area by comparing residences along an urbanization gradient in the Baltimore–Washington area, USA. In addition, earthworms and terrestrial isopods were sampled from residential yards to measure body burdens of metals. Soil metal concentrations from lawns and planting bed (road, foundation, and yard) patches were compared (1) among land-use types (inner urban, outer urban, suburban, and rural); (2) between pre- and post-1940 built residential structures; and (3) among yard patch types. Lawn soil concentrations of As, Cd, and Pb varied statistically among the land-use types. Differences between inner urban and rural lawn soils varied almost eight-fold for Pb, three-fold for Cd, and more than two-fold for As. Bed patches exhibited a slightly stronger relationship than lawns across the urbanization gradient. A similar relationship was shown for pre- and post-1940 structures with older having higher concentrations than post-1940 structures. Earthworm body burdens were statistically correlated with soil Pb, while isopod burdens exhibited a significant relationship with soil As, Cr, Ni, Pb, and Zn. A post-hoc analysis with bird blood Pb data that was available for the residences, showed a significant relationship with earthworm Pb body burdens. This study suggests that despite policy efforts to reduce metal emissions, contamination of soil persists in urban residences at levels that have health implications for people and wildlife living in the Baltimore–Washington, DC area.

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1. Introduction

Urban landscapes are conspicuously contaminated by trace metals and thus pose a potential health risk to people and wildlife

(Mielke, 1999; Wong, Li, & Thornton 2006; Luo, Yu, Zhu, & Li 2012). Although many studies have demonstrated that trace metals accumulate in soils of urban landscapes, less is known about their spatial distribution at finer scales (Schwarz et al., 2012) and potential for accumulating into soil fauna and the animals that feed on these organisms (Roux & Marra, 2007). Metal accumulations in urban soils have been associated with roadside environments and vehicular emissions (Manta, Angelone, Bellanca, Neri, & Sprovieri

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2002; Zhang, 2006; Werkenthin, Kluge, & Wessolek 2014), interior and exterior paint (Trippler, Schmitt, & Lund 1988), stack emissions (Govil, Reddy, & Krishna 2001; Walsh, Chillrud, Simpson, & Bopp, 2001; Kaminski & Landsberger, 2000), management inputs (Russell-Anelli, Bryant, & Galbraith 1999), distance from roads (Wang, Ren, Liu, Yu, & Zhang, 2006; Yesilonis, Pouyat, & Neerchal 2008; Schwarz et al., 2012), land use (Xia, Chen, Liu, & Liu 2011; Mao, Huang, Ma, & Sun 2014), and age of structures (Schwarz et al., 2012; Stewart, Farver, Gorsevski, & Miner 2014). The spatial pattern resulting from these factors is dependent on characteristics of the source such as whether the metal was emitted from a point or non-point source, the height of emission, and the particle size fraction of the pollutant (Cook & Ni, 2007).

Lead (Pb) for example has been emitted into the environment as a gasoline additive and in paint and has over time accumulated in soils. In the United States, both uses have been banned through policy actions for more than 30 years, but nonetheless, the ability of Pb and other metals to persist in the environment has resulted in a distinct spatial pattern of accumulation in soil and riparian sediments (e.g., Bain, Yesilonis, & Pouyat 2010; Schwarz et al., 2012; Datko-Williams, Wilkie, & Richmond-Bryant 2014). Given the aforementioned factors, the resultant spatial pattern of Pb or other trace metals in urban soils should, in part, be determined by the amount and length of exposure to emissions. For example, patterns differ because of the age of the structure (older structures have been exposed to emitted Pb for a longer duration and are more likely to have Pb based paint), the nature of the emitting source, such as the juxtaposition to a road and the volume of traffic on that road, and how far from the source the aerosols and particulates have been dispersed.

In a review of the literature, Yesilonis et al. (2008) found that globally, urban soils consistently accumulated above background levels of Pb, copper (Cu), and zinc (Zn). The authors associated this relationship with automobile usage, specifically tire wear (Zn), brake and radiator wear (Cu), and the accumulation of Pb from historic use of leaded gasoline and paint. Moreover, due to the ability of soil to chemically or physically immobilize these metals, soils have the capacity to accumulate them (Yong, Mohamed, & Warkentin 1992). In addition to Pb, Cu, and Zn, other metals including nickel (Ni), arsenic (As), cadmium (Cd), and chromium (Cr) have been reported to accumulate in urban environments (Kay, Arnold, Cannon, & Graham 2008; Odewande & Abimbola, 2008; Zheng, Chen, & He 2008). Like Pb, Cu, and Zn, the presence of Ni, Cd, and Cr in the environment can be related to automobile use (Van Bohemen & Janssen Van de Laak, 2003) but also industrial sources (Rawlins, Lark, O'Donnell, Tie, & Lister, 2005), while As has been introduced into residential areas as a wood preservative (Townsend, Solo-Gabriele, Tolaymat, Stook, & Hosein 2003).

Since most of the emission sources for trace metals are associated with urban land uses, there have been a number of studies examining the relationship between metal concentrations in remnant patches, such as a forest patch, with distance to the urban core or some metric of urban land use, i.e., an urbanization gradient. For example, Pouyat et al. (2008) showed evidence of a depositional pattern of metals in forest soils along urbanization gradients in the New York City, Baltimore, and Budapest metropolitan areas. The authors found a two to three-fold increase in contents of Pb, Cu, and Ni in urban forest remnants compared to suburban and rural counterparts. A similar pattern but with greater differences was found by Inman and Parker (1978) in the Chicago, IL metropolitan area, where levels of heavy metals were more than five times higher in urban than in rural forest patches. Other urbanization gradient studies have shown a similar pattern (Watmough, Hutchinson, & Sager 1998; Sawicka-Kapusta, Zakrzewska, Bajorek, & Gdula-Argasinska 2003), although smaller cities or cities having

more compact development patterns exhibited less of a difference between urban and rural forest soils (Pavao-Zuckerman, 2003; Pouyat et al., 2008; Carreiro, Pouyat, & Tripler 2009). In all of these studies, regional relationships were established by sampling relatively undisturbed remnant patches and did not include more disturbed or managed soils associated with residential land use, which are the soils most implicated with metal exposure to humans, e.g., Pb in children (Mielke, Gonzales, Powell, & Mielke 2008). For urban soils in the United States, a recent review of the literature showed that for all studies considered, soils in urban centers had higher Pb concentrations than in suburban areas, with the exception of New Orleans (Datko-Williams et al., 2014).

Besides human exposure, the accumulation of trace metals in urban soils can also affect soil fauna. Indeed, soil invertebrates are often used as indicators of pollutant levels (Dallinger, 1994; Nahmani & Lavelle, 2002), although interpreting the results can be challenging due to the multiple factors involved (Beyer & Cromartie, 1987; Nahmani, Hodson, & Black 2007). Soil taxa exhibit differences in their response to trace metal exposure due to their physiology, feeding habits, mobility, and microhabitat preferences (Beyer & Cromartie, 1987; Pižl & Josens, 1995; Scharenberg & Ebeling, 1996; Kamitani & Kaneko, 2007). For instance, earthworms are less mobile and consume a mixture of soil and detritus while isopods and millipedes are more active and feed mainly on plant detritus. An additional effect of metals in soil fauna is the potential for the bioconcentration of metals up the food chain (Hopkin & Martin, 1985; Raczuk & Pokora, 2008). Soil organisms are a primary food source for many invertebrate and vertebrate predators, and thus in heavily contaminated areas there is an increased risk of secondary poisoning (Spurgeon & Hopkin, 1996; Reinecke, Reinecke, Musibono, & Chapman 2000; Maerz, Karuzas, Madison, & Blossey 2005). For example, earthworms are an especially important component of the diet of ground feeding birds due to their high energy and nutrition content (Török & Ludvig, 1988).

Because soil fauna can accumulate trace metals from soil and relatively few studies have compared metal concentrations of residential soils at multiple scales (residential yard to metropolitan region), the objectives of this study were threefold: (1) compare metal concentrations of soils in residential yards that varied in age from inner urban to rural areas in the Baltimore–Washington metropolitan area; (2) compare metal concentrations of soils in patch types that differ in location and soil cover within individual residential yards; and (3) correlate soil metal concentrations with the body burden of metals in earthworms (*Annelida:Oligochaeta*) and terrestrial isopods (*Crustacea:Isopoda*), both of which are potential food sources for several bird species in the region (Roux & Marra, 2007). In the first case, a comparison of soil metal concentrations of residences along an urbanization gradient will reflect, in part, the age of the structures (e.g., before and after policy changes in Pb additives to paint and gasoline) and the effect of vehicular traffic occurring in residential areas in the Baltimore–Washington metropolitan area. In the second and third cases, a comparison of metals in soils, earthworms, and isopods associated with five yard patch types comprised of either lawn (front and back) or planting beds (building foundation, yard or road) along an urbanization gradient will reflect spatial patterns at regional and lot, or yard, scales.

For the first and second objectives, we expected metal concentrations would be greater in more densely populated areas due to higher emissions, and that soils associated with patch types within yards that were closer to roads and adjacent to house foundations would have greater accumulations of metals than in the front or back lawns, particularly for Pb, Cu, and Zn. Moreover, we expected these differences to be magnified by the time of exposure (i.e., age of structure). For objective 3, we expected that the body burdens of earthworms would reflect concentrations of metals in the soils

they were collected from, while body burdens of soil isopods would reflect metal concentrations to a lesser degree because of their greater mobility and different diet.

2. Methods

2.1. Study area

Baltimore City is a historically industrial city located on the Chesapeake Bay in the Mid-Atlantic region of the USA (39°17'11"N, 76°36'54"W). During the last several decades, Baltimore has undergone a significant loss in population. Between 1950 and 2000, the city's population fell from 949,708 (<http://www.census.gov/population/www/documentation/twps0027/tab18.txt>, verified 1/28/2015) to 651,154 (<http://censtats.census.gov/data/MD/05024510.pdf>, verified 1/28/2015). The depopulation of Baltimore has resulted in a large number of residential housing vacancies and vacant lots. Most of the housing stock in Baltimore was built prior to 1950 in contrast to the surrounding Baltimore County area which experienced a substantial increase in housing growth after 1950. Only 60 km from Baltimore, Washington, DC is located near the Chesapeake Bay in the Mid-Atlantic region of the USA on the Potomac River, a tributary to the Bay. Unlike Baltimore, Washington has not had a history of heavy industrial use or a loss of population in the previous 50 years.

The Baltimore–Washington metropolitan area has hot humid summers and cold winters with average annual air temperatures ranging from 14.5°C in the inner urban areas to 12.8°C in the surrounding rural areas. This difference in air temperature in the metropolitan area is attributed to the heat island effect (Brazel, Selover, Vose, & Heisler 2000). Precipitation is distributed evenly throughout the year for the entire study area and ranges from an annual average of 106.8 cm in Baltimore and Washington to 103.1 cm in the surrounding metropolitan area (NOAA, http://www.nws.noaa.gov/climate/local_data.php?wfo=lwx, verified on 1/28/2015).

The Baltimore and Washington metropolitan region lies near or along the Chesapeake Bay between two physiographic provinces: the Piedmont Plateau and the Atlantic Coastal Plain. Most of the metropolitan area is characterized by nearly level to gently rolling uplands dissected by narrow stream valleys. Portions of the Piedmont Plateau in the region are underlain by mafic and ultramafic rock types (Crowley & Rhinhardt, 1979). The Coastal Plain is underlain by much younger, poorly consolidated sediments. Soils in the Coastal Plain are typically very deep, somewhat excessively drained and well-drained upland soils that are underlain by either sandy or gravelly sediments or unstable clayey sediment. The dominant coastal plain soils in the metropolitan area consist of Typic Hapludults. Soils in the Piedmont Plateau of the Baltimore–Washington metropolitan area are very deep, moderately sloping, well-drained upland soils that are underlain by semi-basic or mixed basic and acidic rocks (NRCS, 1976, 1998). The dominant piedmont soils in the metropolitan area consist of Ultic Hapludalfs. Highly disturbed soils and impervious surfaces make up more than 60% and 41% of the land area of Baltimore and Washington, DC, respectively (Pouyat, Groffman, Yesilonis, & Hernandez 2002; Nowak & Greenfield, 2009).

All residences that were sampled were associated with Neighborhood Nestwatch, a citizen science program operated through the Smithsonian Migratory Bird Center at the National Zoo in Washington, DC (<http://nationalzoo.si.edu/scbi/MigratoryBirds/Research/NeighborhoodNestwatch/>, verified on 1/28/2015). Nestwatch program participants help monitor nests and bird sightings in their own yards (Evans et al., 2005). In order to participate in the Nestwatch program, residents must live within a 75-mile

radius of Washington, DC. As of 2010, there were 220 participating sites. The residences selected for our study represent a diversity of the Nestwatch participants due to the different degrees of urbanization sampled. The urban–rural comparisons of residential land use encompassed the Washington, DC and Baltimore, MD metropolitan areas and extended to Caroline and Carroll County, MD. The urban–rural comparison of residential yards consisted of four land-use types defined by population density (no. of individuals per square mile): (1) inner-urban (>8000); (2) outer-urban (7999–2000); (3) suburban (1999–300); and (4) rural (<299). The total number of residential yards sampled included 8 inner-urban (IU), 4 outer-urban (OU), 4 suburban (S), and 7 rural (R) for a total of 23 residential yards (Fig. 1). These yards were randomly selected within the Nestwatch project. The residences were far enough apart that independence between them could be assumed.

Each yard was subdivided into patches delineated by management, cover, and location with respect to the foundation of the house and the nearest road, the two areas on residential lots where trace metals have been shown to accumulate in soil (Mielke, 1999; Yesilonis et al., 2008; Schwarz et al., 2012). In addition, front and back lawns were sampled separately because of likelihood of differences in exposure to emission sources, use, and management. The resultant delineations included front lawn (FL), back lawn (BL), roadside bed (RB), yard bed (YB), and foundation bed (FB) patch types. House, or structural, age and landscaping practices were recorded for each residence. Residences were subsequently divided into two age categories: pre- and post-1940 homes (leaded gasoline began use in the 1930s and commuter traffic increased after 1940 in the metropolitan area). In all cases, the structures were no taller than three floors and thus eddy air flow patterns had potential to transport particles emanating from roads into back yards.

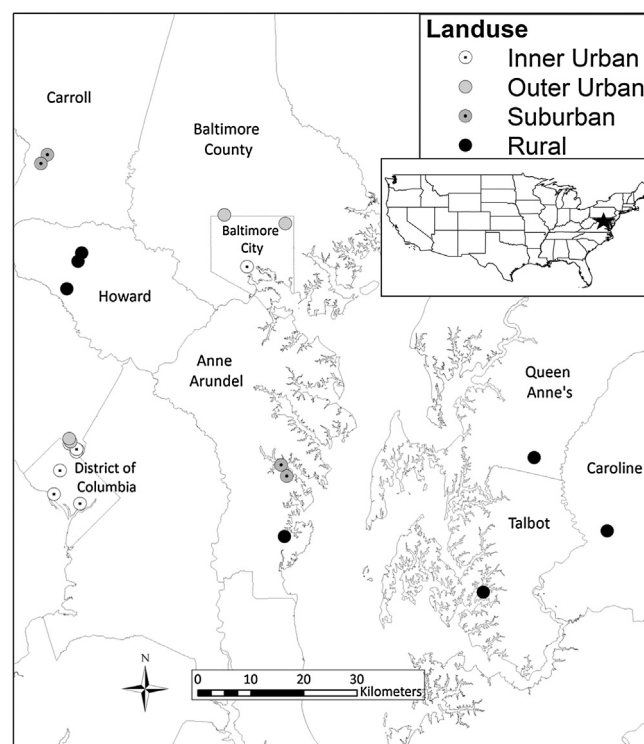


Fig. 1. The Baltimore–Washington metropolitan area showing 23 residences that were included in the study. The residential urban–rural comparisons encompassed the Washington, DC and Baltimore, MD metropolitan areas and extended to Denton County, MD and Westminster County, MD.

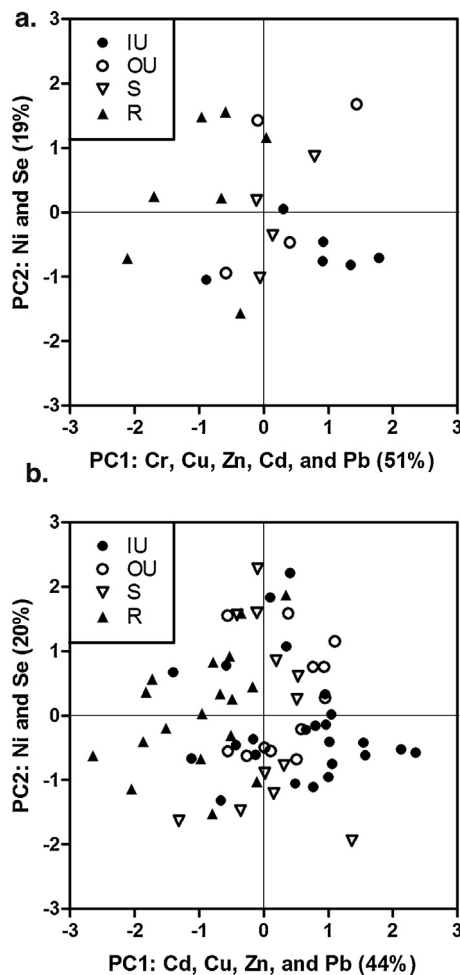


Fig. 2. Scatter plot of first (PC1) and second (PC2) principal components for lawn (a) and bed (b) patches using land-use type symbols. Symbols represent inner-urban (IU), outer-urban (OU), suburban (S), and rural (R). Data were log transformed.

2.2. Soil sampling and analysis

In each yard patch type (FL, BL, RB, YB, and FB), 15 soil cores from a depth of 0–10 cm were collected, and thatch and other fabric material were removed prior to placing cores in a paper bag. These 15 cores were taken in areas that most represented the yard patches. Soil cores from each bag were air-dried, ground, and sieved through a 2-mm mesh sieve. A subsample was ashed in a muffle oven for four hours at 450 °C to determine ash-free dried weight. Additional subsamples were digested at the Baltimore Ecosystem Study/University of Maryland Baltimore County Laboratory using a nitric acid extraction (modified EPA method 3050B). Soil extracts were analyzed at Towson University in Maryland to determine acid soluble extractable As, Cu, Cr, Ni, Pb, and Zn using an Inductively Coupled Plasma Mass Spectrophotometer (ICP–MS). All samples were spiked with 1 µg/L Indium internal standard solution to monitor and correct for instrumental drift and matrix effects. Each batch of 20 samples was typically run with at least one NIST Standard Reference Material (SRM) to monitor recovery and one duplicate sample to monitor reproducibility. All SRMs for the data reported were within 85–105% recovery, and duplicate samples did not deviate by more than 10% from the mean measured value. Elemental data are presented as a concentration per dry weight of soil (mg kg⁻¹).

2.3. Earthworm and Isopod Sampling

In all 23 residences, the RB, YB, and FB planting beds were sampled for earthworm abundance using a 25 × 25-cm quadrat treated with 8L of 0.05% formalin solution (Raw, 1959). The quadrat was placed in a representative area for that patch. Collected earthworms were killed in 70% ethanol and immediately placed in 4% formalin solution. Initial attempts to sample FL and BL patch types for earthworms resulted in only a few individuals, and therefore we discontinued sampling for earthworms in these patch types. In addition, about 80% of the total sample was *Lumbricus* sp. juveniles or Lumbricidae juveniles and therefore only lumbricid earthworms were used in the tissue analysis. At each site, soil moisture (Dynamax TH₂O soil moisture probe), soil temperature (Taylor Precision TruTemp Thermometer), leaf litter depth, slope, and other site characteristics were recorded. In addition, all isopods visibly present within a thirty-minute sampling period were collected from road, foundation and yard bed patch types.

In the laboratory, the collected earthworms (total of 338) and isopods (more than 500) were dried at 70 °C to a constant weight. A randomly chosen subsample of isopods ($n = 38$) and adult earthworms ($n = 17$) were selected for analysis. Each earthworm was dissected, and guts were removed to examine tissue metal content. Samples less than 0.5 g dry weight were digested directly. If the total sample mass exceeded 0.5 g, tissues were homogenized using a Teflon mortar and acid-washed glass pestle on a powered drive. A subsample of the homogenized tissue was then digested. Invertebrates were digested at Towson University with 6 M trace metal grade HNO₃ in Teflon vessels at 150 °C overnight. The solution was then evaporated to dryness and digestion continued with 3 mL of 30% high purity H₂O₂ (Ultrex II grade, J.T. Baker). The digestate was then diluted with 0.2 M trace metal grade HNO₃ for analysis.

2.4. Statistical analysis

2.4.1. Soil data

As an exploratory and data reduction technique, the elemental data were first submitted to a principal component analysis (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 few common components (Pielou, 1984). The first principal component (PC1) explains the maximum possible variance of the dataset, 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 used a scree plot along with the eigenvalues to determine the number of principal components that were kept. We ran separate PCA runs using concentration data (mg kg⁻¹) of Cd, Cu, Co, Cr, Fe, Mn, Ni, Pb, Ti, V, and Z for lawn (FL and BL) and planting bed types (RB, FB, and YB).

To test for differences among residences in IU, OU, S, and R land-use types, means for individual soil properties and the principle components resulting from the PCA (PC1, PC2, and PC3) were analyzed using a restricted likelihood estimation technique in SAS Proc Mixed and using GT2 Hochbergs pair-wise comparisons. Residences were far enough apart such that no spatial correlation was considered. Land-use type was a fixed effect, and there were no random effects (SAS Institute, 2003). The effect of land use on soil concentrations of the residences sampled along the urban-rural gradient was analyzed by Proc Mixed. Separate Proc Mixed tests were run for the lawn (FL and BL) and planting bed (RB, FB, and YB) patch types because we needed to keep the soil samples separate (for correlating with soil organism body burdens) and because of large differences in surface area between the yard patch types. The 15 subsamples were not individually considered in the statistical

analysis because they were composited during field collection. In addition, residences were subdivided by age structure (pre- and post-1940) irrespective of land-use type. A mixed model was used to test for differences in log-transformed elemental concentrations between pre- and post-1940 structures. Since the number of residences sampled per population category and structure age was unequal, Hochberg's Method for unequal sample sizes was used to determine significant differences between means. Soil variables were $\log_{10}$ transformed to stabilize the variance of individual properties where necessary.

2.4.2. Body burden comparisons

Body burden metal concentrations were correlated with soil metal data from the same yard patch type the organism was collected from using Pearson correlation ($n = 17$ for earthworms and $n = 38$ for isopods). In addition, blood concentrations of Pb were available for several species of birds in 12 of the residences sampled in this study (Roux & Marra, 2007). We took advantage of this overlap to conduct a post-hoc analysis correlating Pb concentrations in soil, earthworm, and isopod bodies with bird blood concentrations of Pb in the commonly sampled residences using $\log_{10}$ transformed data in a Pearson correlation.

3. Results

Measured concentrations of trace metals varied widely in this study. 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 1). Even with skewed distributions, contamination of at least one metal has occurred for many of the yards sampled in this study. For example, at least 20% of the patches sampled had Pb concentrations of 93 mg kg^{-1} or greater, which is slightly greater than the guidelines for soil Pb in The Netherlands, or 90 mg kg^{-1} (Table 1). In fact, at the 80th percentile, Cu, Ni, Pb, and Zn had concentrations above-background levels for all of the yard patches sampled (Table 1).

3.1. Principal component analysis (PCA)

A clear relationship was discernible in the PCA for both lawn (front and back) and planting bed (road, foundation, and yard) soils among land-use types and age categories in the Baltimore–Washington metropolitan area (Fig. 2a and b). For the lawn soils, 80% of the variation was explained by the first three components of the PCA, with PC1 accounting for 51% of the variation. For the planting bed soils, a slightly lower percentage of the variance (77%) was explained by the first three components of the

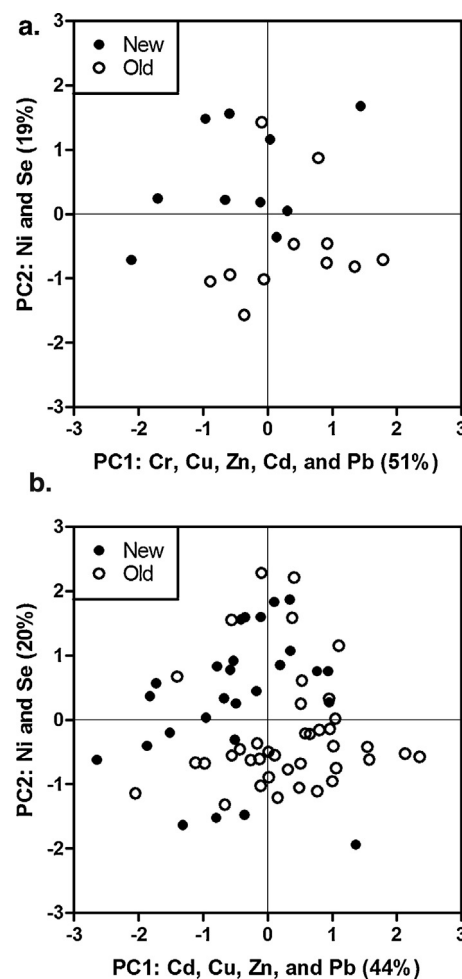


Fig. 3. Scatter plot of first (PC1) and second (PC2) principal components for lawn (a) and bed (b) patches using symbols for structure age categories. Symbols represent new (post-1940) and old (pre-1940) structure locations. Data presented log transformed.

PCA, with PC1 accounting for 44% of the variation. For both soils, positive loadings of PC1 corresponded to Cd, Cu, Pb, and Zn with the addition of Cr for the lawn soils (Fig. 2a and b). Likewise, the second principal component (PC2) was positively associated with Ni and Se for both the lawn and planting bed soils. Inspection of the scatter plots for the first two components (PC1 and PC2) with symbols representing land-use type showed a clustering of IU residences to the right of the origin (positive loadings of PC1 or higher concentrations of Cd, Cu, Pb, and Zn) while R residences clustered to the left (negative loadings) for both lawn and planting bed soils. We interpret the distribution of lawn and planting bed samples along PC1 to represent an urbanization gradient. The distribution of sample sites along PC2 is less clear. For the lawn samples, the IU soils clustered below the origin (negative loadings, or elevated concentrations of Ni and Se) and R soils tended to have positive loadings with respect to PC2 (Fig. 2a), whereas the planting bed soils did not exhibit a discernible pattern with respect to PC2 (Fig. 2b).

Inspection of the scatter plots for PC1 and PC2 using age categories (pre- and post-1940 structures) as symbols showed a clustering in the lower right quadrant (positive loadings of PC1 and negative loadings of PC2, or elevated levels of Cd, Cu, Pb, and Zn and lower levels of Ni and Se) of the older residences for both lawn (Fig. 3a) and planting bed soils (Fig. 3b). This pattern suggests that older structures have higher concentrations of Cd, Cu, Pb, and Zn

Table 1
Percentile concentrations (mg kg^{-1}) of trace metal concentrations for all samples taken in the study. Background concentrations for Cu, Cr, Cd, Ni, Pb, and Zn from Yesilonis et al. (2008).

Element	Cr	Ni	Cu	Zn	As	Se	Cd	Pb
N	107	107	107	107	107	107	107	107
Percentile								
Minimum	3.9	3.5	2.1	13.6	0.51	0.171	0.011	6.6
10	5.3	4.8	3.8	26.6	1.03	0.263	0.051	11.1
20	7.0	6.0	6.0	37.1	1.41	0.364	0.108	14.4
30	10.1	6.9	8.4	46.3	1.61	0.403	0.129	19.7
40	12.0	7.9	10.5	56.7	1.96	0.475	0.148	27.0
50	13.5	9.7	13.6	68.5	2.47	0.522	0.195	33.8
60	16.1	10.8	15.5	89.7	2.92	0.597	0.246	43.6
70	18.5	12.4	22.0	108	3.23	0.669	0.296	65.5
80	21.8	14.0	25.6	136	3.59	0.764	0.391	93.0
90	28.7	18.5	32.5	226	6.38	0.974	0.656	351
Maximum	82.3	40.1	158	534	26.0	1.96	2.08	916
Background	28	13	12	63	NA	NA	0.73	45

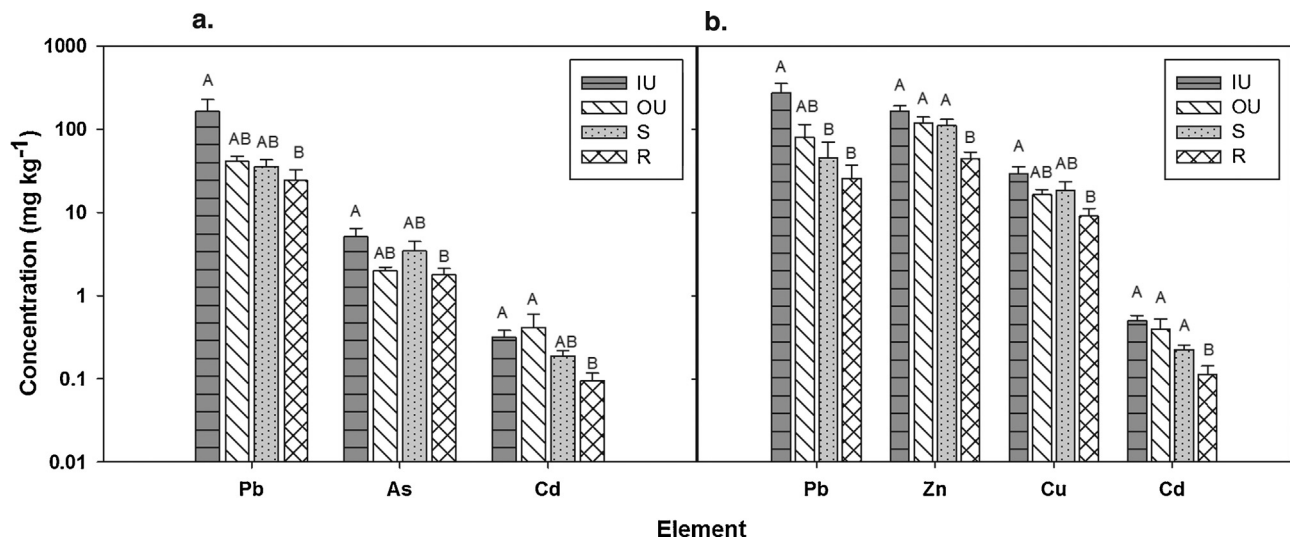


Fig. 4. Mean (S.E.) concentrations of soil metals in (a) residential lawns and (b) bed patches by land-use type: inner-urban (IU), outer-urban (OU), suburban (S) and rural (R). For lawn and bed patches, means were computed for each residential household. Different letters represent significant differences for a metal between land-use types at $\alpha=0.05$. Data were log transformed.

and lower concentrations of Ni and Se than the newer structures in the Baltimore–Washington metropolitan area.

3.2. Land-use type

Consistent with the PCA, a similar relationship for both lawn (front and back) and bed (road, foundation, and yard) soils was shown in statistical comparisons among land-use categories (Fig. 4). Lawn soil concentrations of As, Cd, and Pb varied significantly ($P < 0.05$) among the land-use categories (Fig. 4a), as did PC1 (results not shown). Differences between IU and R lawn soils varied almost eight-fold for Pb, three-fold for Cd, and more than two-fold for As. A post-hoc test showed that for all three metals, IU exhibited higher concentrations than in lawn soils of R residences.

Bed soil patches exhibited a slightly stronger relationship than lawn soils across the urban–rural gradient (Fig. 4b). In addition to Cd and Pb, the yard patch soil comparisons also differed statistically for Cu and Zn (Fig. 4b) as did PC1 (results not shown). Differences between IU and R bed patch soils varied almost ten-fold for Pb and

roughly five-fold for Cd, Cu, and Zn. A post-hoc test showed that for all 4 metals, the IU statistically differed from the R land-use type (Fig. 4b).

3.3. Structure age

A similar relationship for both lawn and bed soils was shown in comparisons between pre- and post-1940 structures (Fig. 5). Lawn soil concentrations of Zn and Pb varied statistically ($P < 0.05$) between structure ages (Fig. 5a), as did PC2 (results not shown), which primarily related to positive loadings of Se and Ni (Fig. 3a). Differences between pre- and post-1940 structures for lawn soils were more than five-fold for Pb and two-fold for Zn. A post-hoc mean separation test showed that for Pb and Zn, the pre-1940 structures exhibited statistically higher concentrations than in lawn soils of post-1940 structures (Fig. 5a).

Bed soil patches exhibited a similar response as the lawn soils with respect to structure age (Fig. 5b). In the addition to Pb and Zn, the bed patch soil comparisons also differed statistically for Cd and

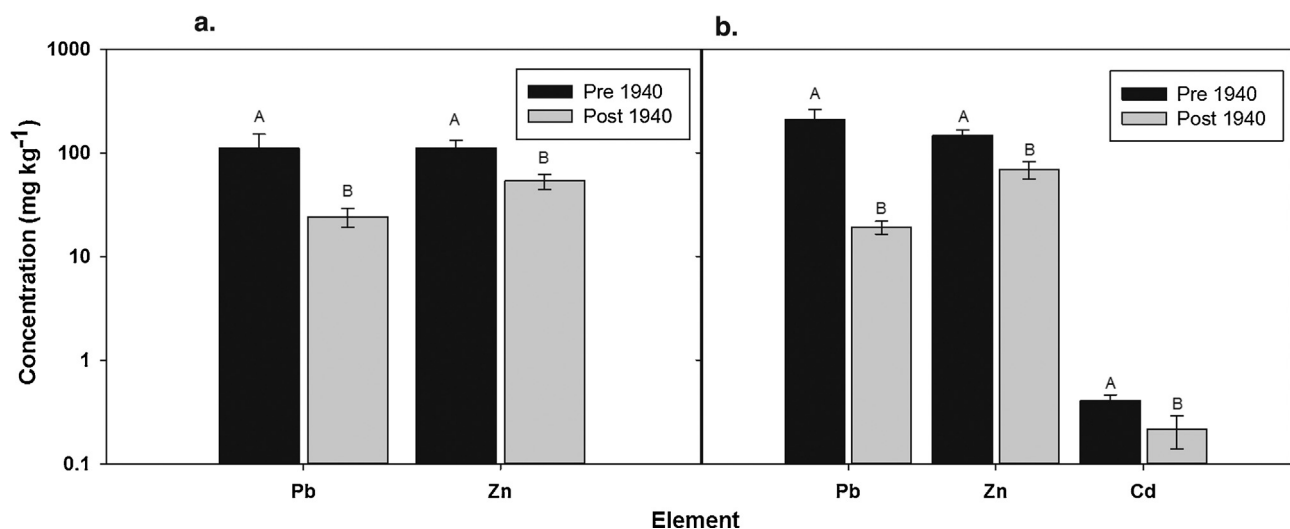


Fig. 5. Mean (S.E.) concentrations of soil metals in (a) residential lawns and (b) bed patches by age of structure (pre- and post-1940 structures). For lawns and bed patches, means were computed for each residential household. Different letters represent significant differences for a metal between structure age at $\alpha=0.05$. Data were log transformed.

Table 2

Mean concentrations (mg kg^{-1}) for nine metals (elements) of 0–10 cm soil in residential yards. Five patch types were distinguished. Standard error is given in parentheses. N = number of sampled patch types. There were no statistically significant differences among patch types at $P < 0.05$.

Label	Foundation bed (FB)			Yard bed (YB)			Road bed (RB)			Back lawn (BL)			Front lawn (FL)		
	N	Mean	S.E.	N	Mean	S.E.	N	Mean	S.E.	N	Mean	S.E.	N	Mean	S.E.
Cr	22	18	3.7	22	18	3.0	23	16	3.3	20	18	3.9	20	18	3.6
Ni	22	11	1.6	22	10	1.0	23	11	1.7	20	11	1.7	20	11	1.7
Cu	22	24	6.8	22	18	2.8	23	15	2.1	20	17	3.2	20	14	3.4
Zn	22	132	23	22	135	28	23	76	12	20	89	17	20	74	18
As	22	3.8	1.1	22	3.7	0.53	23	2.3	0.34	20	3.2	0.61	20	2.7	0.44
Se	22	0.61	0.074	22	0.65	0.08	23	0.59	0.073	20	0.58	0.063	20	0.54	0.064
Cd	22	0.41	0.091	22	0.32	0.065	23	0.24	0.046	20	0.27	0.061	20	0.20	0.037
Pb	22	162	47	22	148	53	23	81	30	20	85	33	20	44	12
Ca	21	3291	610	22	2855	585	23	3410	1010	19	1904	190	20	2584	850

Table 3

Pearson correlation coefficients (r) for earthworm and isopod body burdens and soil metal concentrations (0–10 cm) in residential yards. Soil and invertebrates were collected from the same patch. Significant differences are shown in bold face.

		Cr	Ni	Cu	Zn	As	Se	Cd	Pb
Worm	r	0.37	0.18	0.07	−0.22	0.08	0.33	0.05	0.52
	P	0.14	0.49	0.78	0.39	0.76	0.20	0.85	0.03
	N	17	17	17	17	17	17	17	17
Isopod	r	0.55	0.48	0.07	0.47	0.59	−0.11	0.09	0.79
	P	0.0004	0.003	0.67	0.003	<0.001	0.51	0.59	<0.0001
	N	38	38	38	38	38	38	38	38

Ca (Fig. 5b), as did PC1 and PC3 (results not shown). Differences between Pre and Post 1940 bed patch soils varied almost ten-fold for Pb and roughly five-fold for Cd and Zn. A post-hoc test showed that for all 3 metals the Pre and Post 1940 age types statistically differed from each other (Fig. 5b).

3.4. Yard patch type

There were no statistical differences ($P < 0.05$) among the five yard patch types for all land-use types and the land-use types that we expected to be the most polluted, i.e., OU and IU (Table 2). However, FB and YB Pb concentrations were up to 5-fold greater than FL and BL concentrations.

3.5. Earthworm and isopod body burdens

Earthworm adults were dominated by *Lumbricus rubellus* and *Lumbricus terrestris*. A few juvenile *Amyntas* sp. (Megascolecidae), and three *Diplocardia* sp. (Acanthodrilidae) individuals were caught across all sampling units. Most of the isopods caught were either *Trachelipus rathkii* or *Armadillidium nasatum*. Both are non-native, synanthropic, invasive species.

Body burden relationships between soil fauna and soil metal concentrations differed between earthworms and isopods (Table 3). Isopod burdens were, in general, directly related to soil metal concentrations (As, Cr, Ni, Pb, and Zn) compared to earthworm burdens, which were only statistically related to soil Pb concentrations at $P < 0.03$, $r = 0.52$. Of the metals associated with isopod body burdens, soil concentration of Pb explained more than 70% of the variation. All of the remaining metals that were statistically related at $P < 0.05$ explained between 47 and 59% of the variation in isopod body burdens for As, Cr, Ni, and Zn, respectively (Table 3).

Relationships between soil, body burden, and bird blood metal concentrations at the commonly sampled locations are mixed, probably due to low sample sizes; still, the results suggest that Pb soil contamination can potentially lead to both higher body burden and bird blood concentrations (Fig. 6a and b). For earthworm burden and bird blood levels of Pb, 78% of the variation is explained at

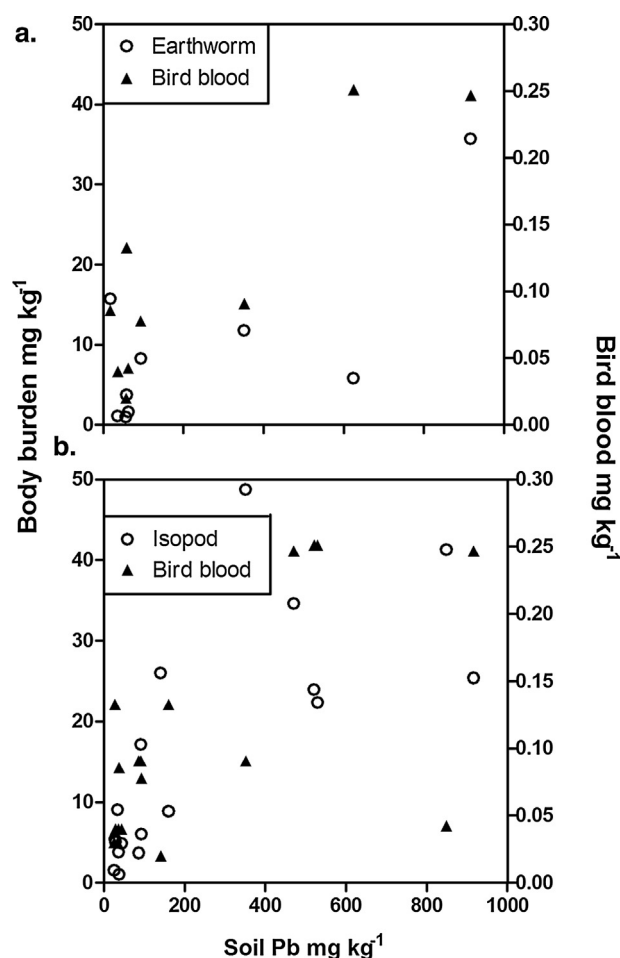


Fig. 6. Scatter plot of Pb concentrations found in bird blood, soil, and body burdens in residential yards in which bird blood data were available. Statistical results (Pearson correlation) for relationships between bird blood and soil Pb concentrations, and bird blood and worm body burden concentrations are shown in (a) and bird blood and soil Pb concentrations and bird blood and isopod body burden concentrations are shown in (b).

$P=0.013$ (Fig. 6a). By contrast, earthworm burden to soil Pb concentrations were not significant for this subset of residences ($r=0.50$, $P=0.17$). However, for the same residences, bird blood and soil Pb concentrations were significantly related ($r=0.66$, $P=0.05$). Similar relationships were found with the isopod body burden, bird blood, and soil Pb concentrations at a subset of residences where bird blood and isopod burdens of Pb were measured (Fig. 6b). For isopod burden and bird blood levels of Pb, the Pearson correlation was not statistically significant ($P=0.19$). However, soil Pb concentrations were statistically related to isopod body burdens ($r=0.83$, $P<0.0001$) as well as bird blood and soil Pb concentrations ($r=0.58$, $P=0.01$).

4. Discussions

4.1. Soil metal concentrations

Overall OU, IU, S land-use types had higher soil concentrations of Pb, Cu, Zn, As, and Cd, than rural residential soils (Fig. 4). The accumulation of Pb, Cu, Zn, Cd, and Cr in particular corresponded to urban environments, which we expect is primarily the result of vehicle pollutant sources (Van Bohemen & Janssen Van de Laak, 2003; Councell, Duckenfield, Landa, & Callender 2004; Hjortenkrans, Bergback, & Hagerud, 2006). Our expectation that Cu, Pb, and Zn would be higher in IU than rural land-use types was thus supported; however, the differences found with Cd and Cr was not expected for the residential soils included in this study. Despite our expectations, studies have found soils contaminated with Cd and Cr in urban areas, and in both cases, contamination was associated with vehicular sources at a city scale (Steiner, Boller, Schulz, & Pronk, 2007; Xia et al., 2011). Indeed, Cd has been associated with tire wear while Cr is used for a number of parts found on vehicles (Van Bohemen & Janssen Van de Laak, 2003).

Overall, the patterns for metal concentrations found in residential soils in the Washington–Baltimore area (Table 1) were comparable to patterns found in forest patches more remote from roads along urban–rural gradients in Baltimore and New York (Pouyat et al., 2008), but with residential soils measured in this study generally having higher concentrations than those found in forest soils. In comparison to Pouyat, Yesilonis, Russell-Anelli, and Neerchal (2007), where in Baltimore only IU residential land use types were sampled (0–10 cm depth), Cu ($43 \pm 3.3 \text{ mg kg}^{-1}$) and Zn ($145 \pm 21.0 \text{ mg kg}^{-1}$) concentrations (mg kg^{-1}) generally fell within the range of values measured in this study, while Pb ($340 \pm 118 \text{ mg kg}^{-1}$) concentrations appeared to be slightly higher.

Concentrations for Pb found in this study are noticeably lower than values (mean 363 mg kg^{-1}) reported in Baltimore by Schwarz et al. (2012). The lower concentrations may be due to differences in sample depth, as Schwarz et al. (2012) reported X-ray fluorescence (XRF) measurements directly at the soil surface vs. samples in this study, which were collected from 0 to 10 cm depth. In addition, the use of XRF technology enabled individual point measurements, whereas our samples were composited per individual patch (ranged 9–300 m² in area), which may have diluted core samples that had higher concentrations. Mielke et al. (2008) compared soils sampled from 0 to 2.5 cm depth in the inner core area of New Orleans with residences in “outer core” areas (approximately corresponding to this studies IU and OU land-use types) and found median concentrations of Pb between 96–152 and 132–707 mg kg^{-1} for outer and inner core areas, respectively. These values, while more comparable to this study, still had much higher maximum values (over 700 mg kg^{-1}), which were measured for individual (and not composited) samples taken directly near a road or building foundation. By contrast, Mao et al., 2014 compared soil

Pb concentrations among varying urban land uses along an urbanization gradient in Beijing China and found much lower urban soil concentrations (maximum of approximately 55 mg kg^{-1}) than our (Table 1) or Mielke et al. (2008) results.

The source of As in residential soils of the more urbanized areas is uncertain. The concentrations of As, while elevated in the IU and S vs. the R land-use types, were not high when compared to other studies. For example, Diawara et al. (2006) found maximum concentrations of As as high as 65 mg kg^{-1} in residential communities in Pueblo, CO while Zhang (2006) found maximum concentrations of As at 30 mg kg^{-1} in Galway, Ireland, which in both cases were associated with either coal combustion or industrial sources that were prevalent in both study areas. Alternatively, the relatively high concentrations found in these studies may be the result of the method used to digest soils (HF acid), which is likely to strip more metal ions from soil exchange sites and minerals than the acid digest method (HCl and nitric acid) used in our study. Arsenic is also used in wood preservatives, which have been implicated in contaminating soils (up to 30 mg kg^{-1}) in residences having wood decks (Townsend et al., 2003). High spatial resolution sampling (meters), such as that employed in Schwarz et al. (2012) for measuring surface soil Pb, would be needed to spatially relate As contamination with wood decks occurring in the residential yards of this study.

Surprisingly, no consistent differences in concentrations were found among the five yard patch types even if the uncontaminated residences in the R and S land uses were not included in the statistical test (Table 2; statistical results not shown). There are two possible reasons for the lack of spatial differences within the residential yards. First, the lack of a relationship with the FB patch type suggests a dispersion pattern that overtops the 2–3 story building structures found in the residential areas included in this study. This finding is consistent with the “aerosol hypothesis” by Cook and Ni (2007) that indicates that relatively large aerosols will be deposited near roads while relatively light particles are carried around structures by air currents. However, an overall trend in soil concentrations was found between lawn and planting beds with the bed soils accumulating more Cd, Cu, Pb, and Zn than in the lawn soils (Figs. 4 and 5). Closer inspection of the data suggests that the planting beds near roads and foundations drove this overall result although small sample sizes and a large amount of variability associated with estimates of soil metal concentrations among yard patch types kept these comparisons from being significant (Table 3). Other studies have found similar relationships with spatial deposition patterns in urban residential yards (Mielke, 1999; Schwarz et al., 2012). Second, the yard patch types themselves represent differently “treated” soils from the perspective of landscape maintenance practices. The patch types related to planting beds (FB, YB, RB) are typically disturbed on an annual basis and may have additions of commercially available garden soil or compost, which due to governmental regulations will not be contaminated with metals. The addition of uncontaminated organic matter or mineral soil would thus dilute any existing contamination by metals.

Older pre-1940 structures had statistically higher concentrations of Zn, Cd, and Pb than younger post-1940 structures, with Cu and Cr exhibiting a similar trend that was not statistically significant at $P<0.05$. These metals also exhibited differences along the urbanization gradient, which was partly driven by the fact that the OU and IU residences were generally older than the S and R (Figs. 2 and 3). A MIXED approach with land use and age did not show significant interactions for either the lawn or yard patch data (results not shown). These results are consistent with Yesilonis et al. (2008) and Schwarz et al. (2012) who showed that Pb concentrations in residential soils within Baltimore were higher in yards with older structures. It is interesting that PC2, which loaded positively with Se and Ni and negatively with As, appeared to separate IU (elevated Se and Ni, lowered As) from R residences, while the pattern

for age of structure showed negative loadings (lowered Se and Ni, and elevated As) for pre-1940 structures (Figs. 2 and 3). This relationship between Ni and age may be one reason Ni did not differ statistically along the urbanization gradient in this study, when Ni has differed in other urbanization gradient studies (e.g., Pouyat & McDonnell, 1991 in the New York City metropolitan area). Moreover, Ni may be confounded by mafic parent materials occurring in the Piedmont province just north and west of the metropolitan area (Pouyat et al., 2007; Pouyat et al., 2008). The apparent tendency for newer lots to have elevated levels of As is consistent with the use of wood preservatives in wood decks, which are typically found in residential areas of relatively low population densities (Townsend et al., 2003).

4.2. Earthworm and isopod body burdens

The stronger relationship between isopod metal burdens and soil concentrations than that found for worms was unexpected (Table 3). We hypothesized that isopods would exhibit less of a relationship with soil concentrations than earthworms due to their feeding preferences (detritus) and higher mobility and thus range in which they can forage. However, earthworm burdens were only statistically related to soil concentrations of Pb, while isopod burdens were related to Pb in addition to Cr, Ni, Zn, and As, which were all associated with IU and OU residences (Table 3). In addition, we correlated soil pH and metal body burdens to see if metal bioavailability may be related to soil acidity and found only a significant, but weak, relationship with isopod body burdens of Pb and Cr ($r = -0.432, P = 0.017$ and $r = -0.528, P = 0.0027$, respectively). Otherwise, we do not have any definitive explanations for these results. Perhaps the extended range that isopods can forage led to a greater integration of coarse and fine detritus, including contaminated particulate matter, found in residential yards and thus led to higher concentration levels. Isopods are epigeic (surface dwellers) and thus were more likely than earthworms to forage outside the individual patches in which they were sampled. Isopods have been shown to be good indicators of heavy metal contamination both in urban (Dallinger, Berger, & Birkel 1992) and industrial (Hopkin, Hardisty, & Martin 1986; Donker, Van Capelleveen, & Van Straalen 1993; Scharenberg & Ebeling, 1996) areas. A handful of isopod species, including those in our study area, are synanthropic, occurring in every continent both in anthropogenic and natural landscapes. Hopkin, Jones, and Dietrich (1993) suggested using such cosmopolitan species as “universal indicators” in pollution monitoring studies. Our results showed good agreement between isopod tissue concentration and the environment, thus supporting this assertion.

To interpret earthworm body burden data is more problematic due to relatively low sample size, and several potentially confounding factors that affect tissue metal concentration. The earthworm diet consists of a mixture of soil and detritus and varies depending on species, age, and season. Earthworms also can take up metals dermally through pore water (Vijver, Vink, Miermans, & Van Gestel 2003). Metal bioavailability depends on soil pH, organic matter content, iron oxide and phosphate concentrations, and their interaction with each other (Ma, Edelman, Van Beersum, & Jans 1983; Pižl & Josens, 1995; Spurgeon & Hopkin, 1996; Park, Bolan, Chung, Naidu, & Megharaj 2011). Metals, such as Cu and Zn are essential micronutrients at low concentrations, but can be toxic at high concentrations. Tissue concentrations of these elements are actively regulated by earthworms and physiological mechanisms exist to quickly eliminate excess levels. This has been demonstrated both in artificial (Lev, Matthies, Snodgrass, Casey, & Ownby, 2010) and field soils (Spurgeon & Hopkin, 1999). Such complex interactions of environmental, biogeochemical, physiological, and ecological factors do not necessarily result in a simple relationship

between the environment and earthworm tissue, and explain the wide range of published data not only in field correlational studies (Beyer & Cromartie, 1987; Spurgeon & Hopkin, 1996; see review in Hobbelen, Koolhaas, & van Gestel 2004), but also under highly controlled laboratory experiments (see review by Nahmani et al., 2007).

The fact that soil organisms sampled in the IU and OU land-use types in the Baltimore–Washington metropolitan area accumulated trace metals suggests the potential for trophic responses, such as the bioaccumulation in birds inhabiting these areas. Roux and Marra (2007) found higher blood levels of Pb for adult and nestling birds in a number of the IU and OU residences we sampled. This was the case not only for resident but also migratory bird species. In addition, the author's found that ground foraging birds had higher blood Pb concentrations than shrub-canopy foragers suggesting soil invertebrates were a primary means of bird blood Pb contamination. In our comparisons, relationships between body burdens and bird blood Pb levels were mixed. In the case of earthworms, the relationship was statistically significant, with 78% of the variance explained in the Pearson correlation (Fig. 6a). However, isopod burdens were not statistically related to bird blood Pb due primarily to one residence where bird blood concentrations were much lower than what would be expected based on soil Pb concentrations found for the same site (Fig. 6b). Obviously, study designs with more observations, and thus statistical power, than the limited observations available in this study are needed to more fully assess the relationship between body burdens of soil invertebrates and bird blood, for Pb and other metals.

5. Conclusions

This study demonstrates that differences exist in soil metal concentrations at multiple scales; from both the scale of a residential yard to a metropolitan region among urban (IU, OU), suburban (S) and rural (R) land-use types. In all cases the concentrations for Cu, Pb, and Zn, were higher in the more urbanized areas than the R or “background” concentrations expected for the soils in the study area (Coastal Plain and Piedmont physiographic provinces). Elevated concentrations of Cu, Pb, and Zn and to a lesser extent As and Cd in residences located in older and more urbanized areas suggest that people living in these areas have a greater risk of exposure to these metals. However, while older structures exhibited higher concentrations across the urbanization gradient, in only the IU and OU residences did the concentrations exceed EPA and The Netherlands standards for Pb (400 ppm and 90 ppm, respectively). Obviously, the overall risk of human exposure to trace metals will depend on characteristics of the soil itself (e.g., pH, P, organic matter content, and the concentration of iron oxides) and the juxtaposition of human activities with soil accumulations of metals. Whatever the case, the fact that Pb contamination persists in residential soils after regulatory measures to reduce Pb exposure took effect decades ago is alarming. Moreover, the risk to wildlife, especially ground foraging birds, appears to also be great given that soil invertebrates constitute a large proportion of their diet.

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