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SPECIAL SECTION

SOIL IN THE CITY

Elemental Concentrations in Urban Green Stormwater Infrastructure Soils

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Abstract

Green stormwater infrastructure (GSI) is designed to capture stormwater for infiltration, detention, evapotranspiration, or reuse. Soils play a key role in stormwater interception at these facilities. It is important to assess whether contamination is occurring in GSI soils because urban stormwater drainage areas often accumulate elements of concern. Soil contamination could affect hydrologic and ecosystem functions. Maintenance workers and the public may also be exposed to GSI soils. We investigated soil elemental concentrations, categorized as macro- and micronutrients, heavy metals, and other elements, at 59 GSI sites in the city of Philadelphia. Non-GSI soil samples 3 to 5 m upland of GSI sites were used for comparison. We evaluated differences in elemental composition in GSI and non-GSI soils; the comparisons were corrected for the age of GSI facility, underlying soil type, street drainage, and surrounding land use. Concentrations of Ca and I were greater than background levels at GSI sites. Although GSI facilities appear to accumulate Ca and I, these elements do not pose a significant human health risk. Elements of concern to human health, including Cd, Hg, and Pb, were either no different or were lower in GSI soils compared with non-GSI soils. However, mean values found across GSI sites were up to four times greater than soil cleanup objectives for residential use.

Core Ideas

- A unique study of elemental concentrations in green stormwater infrastructure soils.
- Elements posing health risk were the same or lower in GSI compared with non-GSI soils.
- However, Cd, Hg, and Pb concentrations were greater than soil cleanup objectives.
- Calcium and iodine concentrations were greater than background levels at GSI sites.

The quality and characteristics of soils in urban areas vary over space and time. Although underlying geology is one influence (Pouyat et al., 2007), industrial and other human activities also influence the spatiotemporal patterns of soil elemental concentrations (Luo et al., 2012b; Schwarz et al., 2012; Thornton et al., 2008; Wong et al., 2006). Atmospheric deposition of pollutants from combustion (e.g., of coal, wood, fossil fuels, and solid waste) represents an indirect regional-scale influence on elemental concentrations in soil. Physical disturbances, such as infill, burial, or otherwise changing surface materials (e.g., paving or building); anthropogenic activities and management practices, such as transportation and pesticide and fertilizer application; and application of lead paint are also influential. Introduced elements can persist in soils over the span of a city's development (Semlali et al., 2004; Yesilonis et al., 2008).

Some toxic elements found in urban soils are a concern for public health managers. Heavy metals, especially Pb, are the top concern because of the human health consequences of exposure (e.g., cognitive impairment, especially for children) (Lanphear et al., 2005). Heavy metals tend to be most concentrated in soils in older developed areas (Filippelli et al., 2005; Laidlaw and Taylor, 2011). In particular, soil Pb concentrations are associated with proximity to roads and buildings (Schwarz et al., 2013) and with age of housing stock (Yesilonis et al., 2008). Ingestion and inhalation of dust and soils at both outdoor and indoor locations are critical pathways to human Pb exposure (Mielke and Reagan, 1998; Yiin et al., 2000). However, although total metal concentrations can indicate a potential presence of risk and soil Pb has been associated with blood Pb concentrations (Mielke et al., 1999), actual human health risks are not assured unless exposure pathways also exist (Luo et al., 2012a).

Urban stormwater runoff and drainage plays a role in the accumulation of pollutants in nearby soils. Runoff from impervious surfaces such as roadways can have relatively high pollutant loads (Zhao et al., 2011). Soils in close proximity to roadways

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Abbreviations: GSI, green stormwater infrastructure; NYSDEC SCO, New York State Department of Environmental Conservation Soil Cleanup Objectives; PC, principal components; XRF, X-ray fluorescence.

tend to be high in Cu, Cr, Pb, Ni, and Zn as a result of tire and break pad wear, vehicle emissions, road surface wear, and atmospheric deposition from other sources (Duong and Lee, 2009; Eriksson et al., 2007; Sutherland et al., 2012; Zhao et al., 2011). Elevated concentrations of Ca can be associated with dust from tire wear (Apeagyei et al., 2011) and concrete leaching (Harrison et al., 2004; Miguel et al., 1997). Brake pads can be a source of Ba, Cu, Fe, Mo, Ti, and Zr (Apeagyei et al., 2011), and vehicle emissions are or have historically been a source of Cd, Cu, Pb, and Zn in near-roadway soils (Zhao et al., 2011).

In recent decades, municipalities have been altering their approach to stormwater management, and this could have implications for urban soil characteristics in the United States and beyond (Valipour, 2014; Valipour, 2015). Public and private agencies are increasingly placing green stormwater infrastructure (GSI) or facilities designed to enhance evapotranspiration, filtration, detention, or reuse of stormwater runoff (Benedict and McMahon, 2002) throughout urban areas and often along roadways. Cities in the United States, such as Philadelphia, began investing in wide-scale GSI implementation after the USEPA endorsed GSI as an acceptable, often cost-effective, method to reduce combined storm sewer overflows (Jayasooriya and Ng, 2014; USEPA, 2007). Green stormwater infrastructure sites are designed to collect surface runoff often from upland areas that may include roadways, buildings, and contaminated sites and then retain the runoff and pollutants that runoff may carry (Liu et al., 2014). Green stormwater infrastructure tools typically involve above-ground trees or vegetation, and typologies can include tree trenches, stormwater swales, curb "bumpouts," planters, rain gardens, green roofs, and wetlands (see Fig. 1 for photographic illustrations of each GSI type). Runoff is sometimes directly routed into GSI sites via a curb cut or inlet. Almost all GSI projects include imported engineered sandy soils designed to promote infiltration, retention, or detention of runoff. This study primarily focuses on facilities designed for infiltration of stormwater runoff.

Green stormwater infrastructure represents a unique (though not homogeneous) type of land disturbance with ecological and public health implications. Green stormwater infrastructure projects are typically located on public or institutional lands, such as in street right-of-ways, parks, or school grounds. Due to their location in public spaces, there is reason to investigate GSI soil characteristics in relation to human exposure and to quantify the associated health risks. Concentration of elements in GSI might lead to human exposure due to proximity to areas of human use or contact. Chemical composition of soils could also affect GSI functions, such as infiltration or evapotranspiration rate (Dietz, 2007; Domenico and Schwartz, 1998; Woltemade, 2010). In addition, elements could affect biotic or aquatic species that have taken residence in GSI sites (Adriano, 2001), such as the balance between native and non-native species (Huebner et al., 2014; Kuhman et al., 2011).

Although previous studies have characterized element loads in road-deposited sediments, stormwater ponds (Frost et al., 2014; Muthukrishnan, 2010), or waterbodies downstream from stormwater basins (Hale et al., 2015), published studies evaluating soil elemental concentrations in smaller, high-infiltration-rate GSI projects are rare. We investigated the effect of GSI project construction on elemental concentrations in surface soils compared with concentrations measured in control locations at each site. In addition, we tested for effects of ownership, type of GSI tool, distance from roadway, direct receipt of roadway runoff, project age, and background geology and soils on soil elemental concentrations. We also compared measured mean soil concentrations with background soil average concentrations and soil clean-up objectives to provide an initial, qualitative risk assessment.

Fig. 1. Photographs of city of Philadelphia green stormwater infrastructure tools (Kondo et al., 2015). Photographs taken by R. Schwartz.

Materials and Methods

Study Area

The study was conducted in the city of Philadelphia, PA, which is located at the confluence of the Schuylkill and Delaware Rivers. Philadelphia is located on the boundary between the Atlantic Coastal Plain and the Piedmont Plateau geologic provinces. Based on our calculations, 88% of the city's soils are classified as "urban" due to their modification over time by development. Heavy industry facilities in the city are located primarily along the Delaware River on the east side of the city (Fig. 2), with the exception of the Philadelphia Refinery, which is located in the southwest area of the city (built in 1850s and still operating in 2014). Elevated soil Pb content, as a result of historic leaded gasoline and paint use, has been a major public health concern in the city (Mielke, 1994).

Sampling Design

At the time of the study, the city water and parks departments had completed construction of approximately 75 GSI projects designed for infiltration, retention, reuse, detention, or evapotranspiration of stormwater, such as tree or storage trenches, pervious pavement, rain gardens, stormwater planters, bumpouts, swales, and basins (Fig. 1) (Philadelphia Water Department, 2011). With the exception of four wetlands in the study sample, most GSI facilities were not designed to retain water for significant periods of time. We sampled soils at all GSI facilities that could be located and that contained above-ground exposed soils. We excluded projects that did not involve exposed soils (e.g., projects involving pervious pavement) from the study. In addition, some sites were not found due to inaccurate coordinates (i.e., they were not found in the expected locations). Our final sample set included projects designed for stormwater infiltration or detention in seven classes, including tree or storage trenches, planters, bumpouts, rain gardens, swales (or grassed waterways, infiltration berms, or basins), wetlands, and gully repairs (see Table 1 for project type frequency).

During the months of May through October 2013, we collected a total of 396 soil samples at 59 GSI project sites, including 219 soil samples within GSI project sites (treatment samples; "GSI sites"), and 177 soil samples 3 to 5 m outside of GSI projects (control samples; "non-GSI sites"). Non-GSI control samples were not located in or near concentrated flowpaths or stormwater infiltration areas. At each GSI and non-GSI site, we collected three or more samples, aiming to sample the full variety of soil conditions at each site. Each sample consisted of three surface soil (0–5 cm depth) subsamples collected approximately 15 cm apart, which were then mixed thoroughly to obtain a composite sample. We collected all soils using a stainless steel trowel that was cleaned between each sampling. We stored samples in clean plastic containers. We then returned soils to the laboratory to air dry. Once dry, samples were returned to containers and hand-ground to homogenize soils. All field and laboratory equipment was cleaned between samples, and standard QA/QC protocols were followed.

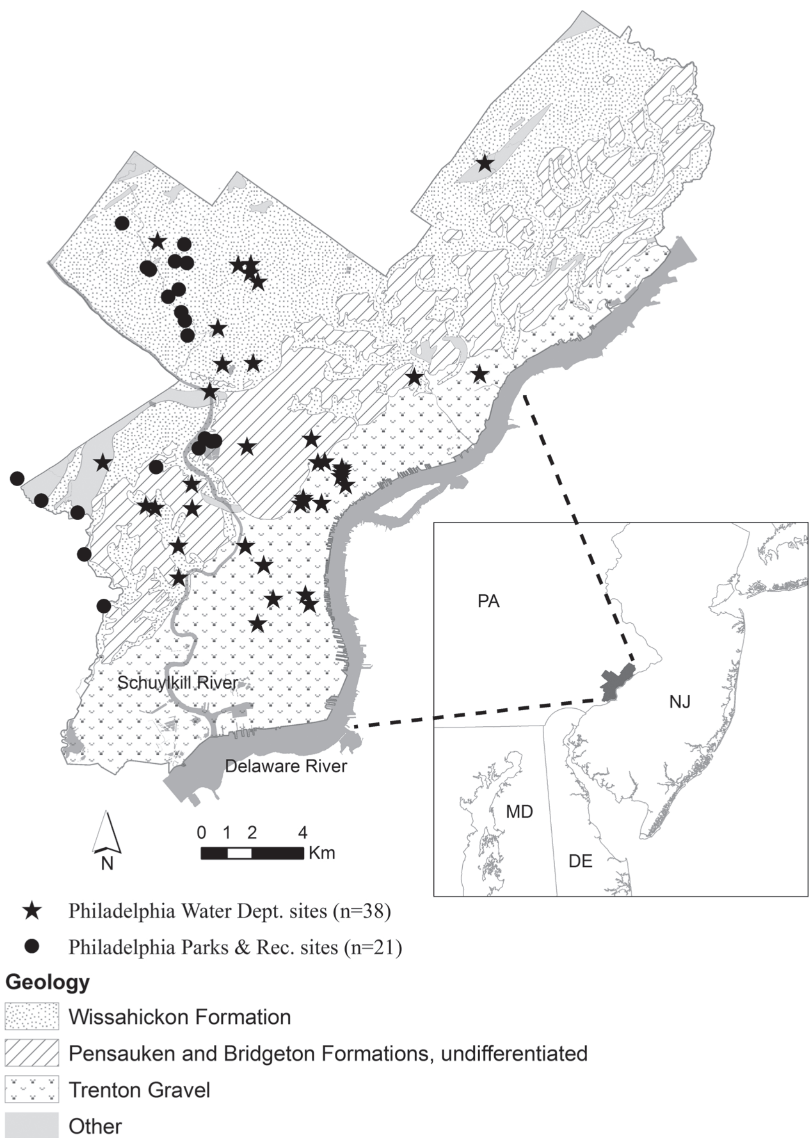

Fig. 2. Green stormwater infrastructure sites by agency ownership, with underlying geology.

Elemental Concentrations

Bulk soil concentrations of metal (K, Ca, Ti, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Rb, Sr, Zr, Mo, Ag, Cd, Sn, Sb, Ba, Hg, and Pb) and nonmetal elements (S, Se, Cl, and I) were determined using X-ray fluorescence (XRF) (USEPA Method 6200). Concentrations were recorded as the mean of three measurements using a handheld XRF spectrometer (Olympus Inc.). We used the "three-beam soil" mode, which incorporates a correction for Compton scattering in nonsolid materials. We performed baseline correction and instrument check every 20 samples against a factory-supplied standard stainless steel disk. X-ray counts were converted to total concentrations using a matrix correlation method (Rollinson, 2014).

Although XRF is not strictly quantitative, its use for the determination of soil elemental concentrations is considered an acceptable standard screening method for large sample throughput applications (USEPA Method 6200). To support the XRF analyses, we randomly selected 5% of the study's soil samples and submitted them to an independent soil testing laboratory for conventional acid digestion analyses (USEPA Method 3050b) for Pb, Ni, Cr, Zn, and Cu concentrations. Analyses of other elements were not available or were not economically viable. We compared the XRF results with the laboratory results using scatter plots, paired *t* tests, and correlation analysis.

We also compared XRF elemental concentrations to mean concentrations found in eastern US soils (Shacklette and Boerngen, 1984). Shacklette and Boerngen (1984) performed analyses on strong acid-digested soils using several analytical detection methods to generate total elemental concentrations, which should be comparable to values generated by the XRF. In addition, we compared mean XRF elemental concentrations with the (total) concentrations recommended by the New York State Department of Environmental Conservation Soil Cleanup Objectives (NYSDEC SCO) for residential and ecological uses (New York State Department of Environmental Conservation, 2006). Toxicity values were calculated for long-term (chronic, including cancer) and short-term (acute) health effects based on exposure via multiple exposure pathways described in a Technical Support Document (New York State Department of Environmental Conservation and New York State Department of Health, 2006). The NYSDEC SCOs are based on nominal concentrations derived from the literature and are intended for comparison to extractable chemical measurements, which one would expect to be generally lower than the total concentrations determined by XRF.

Predictor Variables

We measured and recorded site and area characteristics that could potentially explain differences in soil elemental concentrations. These are shown in Table 1 and included (i) the GSI tool type; (ii) surrounding land use (school, farm, transportation, recreation, residential, or commercial); (iii) whether the site received drainage directly from a street; (iv) age of the installation classified as 1 to 5 yr old, 6 to 10 yr old, or ≥11 yr old (we determined classes based on frequency distributions and in an attempt to avoid problems associated with model overspecification); (v) underlying soils (Alluvial land or Hatboro silt loam, Manor loam, Urban land, or unknown); and (vi) underlying geology (Trenton Gravel, Pensauken and Bridgeton Formations, Granitic gneiss and granite, or Wissahickon Formation).

Table 1. Descriptive statistics of predictor variables.

Variable	Description	Number of samples	
		Non-GSI†	GSI
GSI/non-GSI site		177	219
Ownership	Philadelphia Water Department‡	114	155
	Philadelphia Parks & Recreation	63	64
GSI type	tree or storage trench‡	39	51
	planter	9	12
	bumpout	6	6
	rain garden	33	41
	Swale, grassed waterway, berm, basin	27	48
	wetland	4	4
	gully repair	59	57
Surrounding land use	school‡	18	21
	farm	15	18
	transportation	45	60
	recreational	55	73
	residential	41	42
	commercial	3	5
Receives street drainage	no‡	88	106
	yes	89	113
Age, yr	1–5‡	108	133
	6–10	45	65
	≥11	24	21
Soils	alluvial land or hatboro silt loam‡	12	12
	manor loam	27	33
	urban land	129	168
Geology	trenton gravel‡	51	66
	Pensauken and Bridgeton formations	27	30
	Granitic gneiss and granite	18	29
	Wissahickon formation	81	94

† GSI, green stormwater infrastructure.

‡ Reference class in mixed-effects mean regression models.

Statistical Analyses

Exploratory Analyses

We first conducted descriptive analyses of compiled data, testing for differences in mean elemental concentrations found in GSI and non-GSI soils using paired t tests, and two-sample Wilcoxon rank-sum tests. We calculated correlation coefficients among all elements to test for correlations between elemental concentrations. In addition, we conducted principal components analysis using all elements to assess for any patterns of clustering (latent structure) among soil elemental concentrations across sites. All statistical analyses were performed using Stata v13 (StataCorp LP).

Linear Mixed-Effects Mean Regression

We used linear mixed-effects models to conduct regression analyses to assess the potential impact of GSI project construction on soil elemental concentrations while controlling for predictor variables. Before regression analysis, we conducted tests for normality and multicollinearity in elemental concentrations. Skewness tests confirmed that all elemental concentration data were positively skewed. We therefore used a $\log_e(Y + s)$ transformation to address heteroscedasticity in the elemental concentrations, where s is half of the minimum of the observed nonzero concentration. We estimated the effect of predictor variables by comparing estimates of treatment in base models to full models for each element. The base model for each element was:

$$Y_{ij} = \beta_0 + \beta_1 R_1 + \xi_i + \varepsilon_{ij}, j = 1,...,n_i \quad [1]$$

The units of observation were constructed GSI projects or sites (i), each consisting of multiple observations or samples (j). Y_{ij} is the metal or element concentration, $\beta_1 R_1$ is a treatment-control term (variable of interest), ξ_i is the random-effect of the ith site, and ε_{ij} represents the residual error. Full regression models (see Eq. [2]) included a metal or element concentration Y_{ij}; a series of p covariates (listed in Table 1), $\beta_k X_{ijk}$; a random-effects parameter clustered by site, ξ_i; and residual error, ε_{ij}:

$$Y_{ij} = \beta_0 + \beta_1 R_1 + \sum_{k=4}^{p} \beta_k X_{ijk} + \xi_i + \varepsilon_{ij}, j = 1,...,n_i \quad [2]$$

In each model, the β_1 coefficient of the treatment/control term estimates the effect of the treatment on the outcome, with $e^{\beta 1}$ representing percent difference. Covariate effects for categorical predictors are represented as relative effect in comparison with a reference category.

Linear Mixed-Effect Quantile Regression

We used a linear mixed-effects quantile regression model to assess for the impact of GSI project construction on soil element concentration at different percentiles, such as 10, 25, 50, 75, and 90%, while controlling for other predictor variables. The quantile regression model allows the impact of GSI project construction to differ at different levels of soil element concentration, which is particularly suitable when heteroscedasticity is observed in the data. Compared with the linear mixed-effects mean model, which focuses on how the mean soil element concentration is affected by the GSI project construction, the quantile regression model provides information on how the entire distribution of soil element concentration is affected by the GSI project construction. As an analog to Eq. [2], we have the following equation:

$$Q_{i,\tau}(Y_{it}) = \beta_0(\tau) + \beta_1(\tau) R_1 + \sum_{k=2}^{p+1} \beta_k(\tau) X_{ijk} + \varepsilon_{ij}(\tau), 0 < \tau < 1 \quad [3]$$

where $Q_{i,\tau}(Y_{ij})$ represents the τ, which is the quantile of Y_{ij} (i.e., $P[Y_{ij} \le Q_{i,\tau}(Y_{ij})] = \tau$). In Eq. [2], ε_{ij} is assumed to be Gaussian distributed with mean zero and constant variance. The quantile regression model in Eq. [3] relaxes this assumption to allow heterogeneous and non-Gaussian errors. A special case of $\tau = 0.5$ leads to a median regression model, which is less sensitive to outliers compared with a mean regression model, which is important in light of the wide and highly skewed distributions of the concentrations of the elements. The parameter $\beta_1(\tau)$ represents the effect of GSI project construction, which can take different values at different τ. The parameter estimates and associated P values are obtained by using the method introduced in Wang and He (2007).

Results

Comparison analyses found that elemental concentrations measured by acid digestion and XRF were well correlated for Pb (Spearman's rank correlation coefficient, $\rho = 0.87$; $P < 0.00001$), Zn ($\rho = 0.86$; $P < 0.00001$), and Cu ($\rho = 0.69$; $P = 0.001$) but not for Ni ($\rho = 0.20$; $P = 0.4$) or Cr ($\rho = 0.16$; $P = 0.5$) (details shown in Supplemental Fig. S1 and Supplemental Table S4). Paired t tests also showed that differences in concentrations between the two methods were statistically different from zero (i.e., $P < 0.05$). However, the absolute differences in elemental concentrations between the two methods were generally on the order of a factor of 4 and therefore are within an order of magnitude. We are confident that any of the observed differences in elemental concentration measure by XRF between GSI and non-GSI samples are robust, although we interpret the absolute values of elemental concentrations with prudence. It is important to note that the error is in the dependent variables in our regressions; this does not create systematic bias but does dilute the power of our analyses.

Tables 2 and 3 shows mean and geometric mean elemental concentrations (mg kg^{-1}) as well as geometric standard deviation and minimum and maximum elemental concentrations by GSI and non-GSI site type. Elemental concentrations measured at GSI and non-GSI sites varied widely (geometric SD, 1.4–2.8). In general, statistical means-comparison tests indicated that mean concentrations at non-GSI sites were greater than mean concentrations at GSI sites. Concentrations of Ca and I at GSI sites were greater than concentrations at non-GSI sites (based on log-transformed values), and concentrations of Hg, Ag, Rb, Sn, Ti, and Zr at GSI sites were lower than at non-GSI sites ($p < 0.01$). The remaining elements showed no statistically significant difference in statistical means-comparison tests.

As shown in Tables 2 and 3, mean concentrations in GSI and non-GSI sites were generally greater than mean concentrations found within eastern US soils (Shacklette and Boerngen, 1984) and the concentrations recommended by the NYSDEC Soil Cleanup Objectives for residential and ecological uses (New York State Department of Environmental Conservation, 2006). In comparison with eastern US soil mean values, we found notably greater mean values of Co (~35 and 29% greater for non-GSI and GSI site samples, respectively), Sn (9 and 11%), Hg (30 and 26%), and I (15 and 52%).

Mean heavy metal concentrations in study samples were found to be uniformly greater than eastern soil mean values and NYSDEC soil cleanup objectives (Tables 2 and 3), even though Pb concentration was significantly lower in GSI soils than in non-GSI soils. Mean Hg and Cd soil concentrations were approximately 4 times greater and mean Pb concentration was 0.25 times greater than NYSDEC residential standards.

According to linear mixed-effects mean regression models, GSI type helped to explain this association. Compared with Pb concentrations found in tree trench sites, Pb concentrations found in wetlands, stormwater planters, bumpouts, and basins were more likely to be greater. Concentrations of Hg were more likely to be greater in gully repairs than in tree trenches. In addition, a greater Pb concentration was associated with sites that directly receive street drainage compared with sites that do not.

Tests for correlation found that multiple elemental concentrations were significantly intercorrelated, perhaps due to underlying geology and soil type (Supplemental Table S1). Principal components analyses assisted in revealing patterns of clustering of elemental concentrations across GSI sites. The first three principal components (PC) explained 51% of the variation in elemental concentrations: PC1 explained 27% of the total variance in the intercorrelation (Table 4 shows that the variables most strongly associated with PC1 were Fe, K, Co, and Rb [positive loadings]); PC2 explained 17% of the total variance, which was most strongly associated with Ca and Zn (positive) and with Ti and Zr (negative); and PC3 explained 7% of the total variance and was most strongly associated with Ag (positive) and Ba (negative) (Table 4).

Table 2. Macro- and micronutrient concentrations at green stormwater infrastructure project sites and control sites.

Element	GSI† (1) vs. non-GSI (0) soils	n < LOD‡	Mean	Geometric Mean	Geometric SD	Min.	Max.	Ratio of mean concentrations§ to: Eastern US soils mean concentration¶	NYSDEC SCO, eco.#	NYSDEC SCO, residential††
			— mg kg^{-1} —			— mg kg^{-1} —				
Macronutrients										
Ca	0	0	7,395	5,225	2.1	801	92,318	1.2		
	1	0	13,265	8,737	2.5	1,214	68,490	2.1		
K	0	0	11,199	10,239	1.5	3,595	31,238			
	1	0	9,983	8,561	1.8	749	28,185			
S	0	1	4,505	3,276	3.3	0	12,849	4.1		
	1	0	3,945	2,607	3.0	47	25,407	3.6		
Micronutrients										
Co	0	0	317	293	1.6	26	641	34.5		
	1	0	268	231	1.9	16	645	29.1		
Cr	0	0	77	74	1.4	8	231	1.5	1.9	2.1
	1	0	70	66	1.4	19	203	1.3	1.7	1.9
Cu	0	0	51	46	1.6	13	225	2.3	1.0	0.2
	1	0	51	44	1.8	4	198	2.3	1.0	0.2
I	0	138	18	1	5.6	0	578	15.0		
	1	145	62	3	12.8	0	875	51.7		
Fe	0	0	26,054	24,644	1.4	10,369	52,940	1.0		
	1	0	24,044	21,771	1.6	4,319	81,097	1.0		
Mn	0	0	588	544	1.5	185	2,870	0.9	0.4	0.3
	1	0	572	510	1.7	109	2,411	0.9	0.4	0.3
Mo	0	146	0	1	1.7	0	7	0.4		
	1	182	0	1	1.9	0	9	0.6		
Se	0	48	1	1	1.7	0	4	2.2	0.3	0.0
	1	80	1	1	1.7	0	6	1.8	0.2	0.0
Zn	0	0	165	129	1.8	50	1,934	3.2	1.5	0.1
	1	0	191	142	2.2	14	1,908	3.7	1.8	0.1

† GSI, green stormwater infrastructure.

‡ Number of samples below limits of detection.

§ Mean values and objectives are not available for all metals and elements.

¶ Source: Shacklette and Boerngen (1984).

Source: NY State Department of Environmental Conservation, soil cleanup objectives, protection of ecological resources. Title 6 of the Official Compilation of New York Codes, Rules & Regulations, Part 375.

†† Source: NY State Dept of Environmental Conservation, soil cleanup objectives, residential use. Title 6 of the Official Compilation of New York Codes, Rules and Regulations, Part 375.

There were few discrepancies between base regression (Eq. [1]) and full regression (Eq. [2], which included covariates) analyses, which indicates that the GSI versus non-GSI site status had a strong effect on concentrations compared with other predictors. Regression estimates of the treatment effect showed that many concentrations within GSI sites were on average less than concentrations in non-GSI sites (Tables 5 and 6). The strongest associations in this direction were for K (−13%), S (−22%), Co (−17%), Pb (−30%), Sb (−22%), and Ti (−7%). On the other hand, the full models showed that Ca and I concentrations were on average greater in GSI than in non-GSI sites by 65 and 148%, respectively (Supplemental Tables S2 and S3).

According the mean-regression models, GSI and non-GSI sites that receive direct street runoff had significantly lower concentrations of Co, Cu, Fe, Pb, and As compared with those that do not. Project type was also associated with elemental concentrations. Stormwater bumpouts (Fig. 1) were associated with elevated concentrations of K, Co, Cr, Fe, Mn, Pb, Ba, Ni, and Rb (compared with soils in tree trenches as a reference class). Older GSI sites had soils with significantly greater Cl and had lower Ni concentrations. In addition, an underlying urban land soil classification had a negative effect on soil K, Se, and Ti concentrations.

Quantile regression models demonstrated treatment effects on the entire distribution of concentrations within GSI sites compared with background levels (Fig. 3). Figures 3a and 3b

Table 3. Heavy metal and other elemental concentrations at green stormwater infrastructure project sites (n = 219) and control sites (n = 177).

Element	GSI† (1) vs. non-GSI (0) soils	n < LOD‡	Mean	Geometric Mean	Geometric SD	Min.	Max.	Ratio of mean concentrations§ to: Eastern US soils mean concentration¶	NYSDEC SCO, eco.#	NYSDEC SCO, residential††
			—mg kg^{-1}—			—mg kg^{-1}—				
Heavy metals										
Cd	0	10	11	9	2.6	0	27		2.8	4.4
	1	19	12	8	2.8	0	28		3.0	4.8
Hg	0	20	4	3	2.3	0	12	29.6	19.8	4.4
	1	39	3	2	2.6	0	53	25.8	17.2	3.8
Pb	0	0	126	89	2.2	20	846	7.4	2.0	0.3
	1	0	91	61	2.6	3	1,387	5.4	1.4	0.2
Other elements										
Ag	0	22	11	7	3.5	0	37		5.6	0.3
	1	62	9	4	4.6	0	65		4.4	0.2
As	0	6	9	7	2.0	0	19	1.2	0.7	0.5
	1	4	9	7	2.1	0	36	1.2	0.7	0.5
Ba	0	3	446	397	2.2	0	808	1.1	1.0	1.3
	1	0	395	360	1.8	3	994	0.9	0.9	1.1
Cl	0	36	217	40	11.5	0	5,147			
	1	47	149	36	11.4	0	3,463			
Ni	0	49	8	4	4.3	0	49	0.4	0.3	0.1
	1	48	9	4	4.1	0	119	0.5	0.3	0.1
Rb	0	0	71	66	1.5	18	164	1.3		
	1	0	58	49	2.0	2	131	1.1		
Sb	0	51	4	2	3.6	0	23	5.3		
	1	86	3	2	3.6	0	20	4.2		
Sn	0	9	16	12	2.4	0	82	11.0		
	1	16	13	8	3.2	0	79	8.5		
Sr	0	0	78	74	1.4	30	174	0.6		
	1	0	77	69	1.7	7	231	0.6		
Ti	0	0	4330	4189	1.3	1477	6,492	1.2		
	1	0	4037	3815	1.4	1629	16,518	1.2		
Zr	0	0	593	561	1.4	208	1,362	2.0		
	1	0	461	424	1.5	149	1,747	1.6		

† Green stormwater infrastructure.

‡ Number of samples below limits of detection.

§ Mean values and objectives are not available for all metals and elements.

¶ Source: Shacklette and Boerngen (1984).

Source: NY State Dept of Environmental Conservation, Soil Cleanup Objectives, Protection of Ecological Resources. Title 6 of the Official Compilation of New York Codes, Rules & Regulations, Part 375.

†† Source: NY State Dept of Environmental Conservation, Soil Cleanup Objectives, Residential Use. Title 6 of the Official Compilation of New York Codes, Rules and Regulations, Part 375.

show treatment effects at all quantile levels for elements where treatment substantially and negatively affects low concentration but barely influences high concentration, Fig. 3c shows treatment effects for elements where treatment substantially and positively affects high concentration but barely influences low concentration, and Fig. 3d shows elements where a somewhat even effect is observed at different quantile levels. The obvious trend in treatment effects at different quantile levels indicates that there is still substantial heterogeneity in the data even after the log transformation; thus, GSI installment affects not only the center of the distribution of concentration but also the shape of the distribution. Treatment had especially negative effects at low concentrations of Ag, Rb, and Sn. We also noted evidence in support of differences in Ca, I, and Zn concentrations at GSI sites, and this positive effect is stronger within high quantile levels, especially for I.

Discussion

Green stormwater infrastructure facilities are designed to collect street runoff and retain any pollutants it carries to improve water quality in receiving waters. Street runoff or street dust has been found to contain multiple metals (primarily from building materials and traffic) in urban areas (Duong and Lee, 2009; Eriksson et al., 2007; Miguel et al., 1997; Sutherland et al., 2012; Zhao et al., 2011). Yet, we found elements of concern, such as Hg, Cd, and Pb, within GSI sites to be not different or lower than in soils at non-GSI sites. We also found lower concentrations of As, Co, Cu, Fe, Pb, and Rb in sites that receive direct street runoff compared with those that do not. This finding is potentially a result of accumulation over time of metals in non-GSI background sites from historical industrial, land-based activities and atmospheric sources in Philadelphia.

Table 4. Principal components (eigenvectors) for study elements.

Element	PC1†	PC2	PC3
S	0.11	−0.08	−0.18
Cl	−0.09	0.10	0.20
K	0.32	−0.02	0.10
Ca	−0.04	0.38	0.21
Ti	0.18	−0.33	0.08
Cr	0.23	0.08	−0.23
Mn	0.21	0.18	−0.02
Fe	0.34	0.04	−0.01
Co	0.31	0.09	0.00
Ni	0.07	0.04	0.03
Cu	0.20	0.27	0.14
Zn	0.09	0.36	−0.03
As	0.19	0.12	−0.03
Se	0.24	−0.10	0.18
Rb	0.33	0.03	−0.05
Sr	0.24	0.25	−0.01
Zr	0.06	−0.35	−0.09
Mo	−0.04	0.11	0.23
Ag	0.21	−0.25	0.33
Cd	0.05	−0.11	0.51
Sn	0.19	−0.03	−0.12
Sb	−0.05	0.08	−0.18
I	−0.11	0.32	0.26
Ba	0.15	−0.08	−0.33
Hg	0.23	−0.20	0.26
Pb	0.19	0.17	−0.19

† PC1, PC2, and PC3 indicate the first three principal components.

As a pervious medium, GSI soils promote the interception, percolation, and infiltration of runoff. It is important to assess whether metal accumulation is occurring in GSI soils because other studies (Sutherland et al., 2012; Zhao et al., 2011) have found that urban stormwater drainage areas, such as roadway drainage swales, are often host to soils with accumulated elements of concern. This is important because many types of GSI facilities require that accumulated soils be dredged, vacuumed, or removed, which may expose workers performing these tasks. Furthermore, because GSI sites are often in places where the public could also be exposed, it could be advisable to evaluate exposure to the public at large. Lastly, metal concentrations are of concern because they could affect the designed functions of GSI facilities through potential phytotoxicity. In addition, the potential concentration of elements of concern in GSI sites could represent novel mechanisms for increased environmental contamination and a new route of exposure that could degrade ecosystem function.

Although we found that GSI sites were less contaminated than non-GSI sites, mean heavy metal concentrations were generally greater than regional background concentrations and the NYSDEC objectives for residential soils, and this could be a health concern for the general public in Philadelphia. It should be noted that the XRF method is a screening method that measures total concentrations without consideration of environmental mobility or availability. Although this study did not directly test for mobility, availability, or modifiers of heavy metal concentrations, regression models indicated that project type and street drainage potentially influence especially Pb concentrations in GSI and nearby soils.

Constructed facilities receive maintenance (e.g., weeding, trash, and sediment removal) at least once per year and as frequently as once per month, and these area-wide high heavy metal concentrations could be further examined to determine whether they pose concern for maintenance workers or to the general public due to the disposal of contaminated soils. However, this study did not assess environmental mobility, bioavailability of contaminants, or estimated exposure; therefore, there is no basis to assume a meaningful health risk to maintenance workers from any of the studied elements. Future estimation of environmental mobility, bioavailability, exposure, and health risk is warranted.

Soil Ca concentrations were 65% greater in GSI sites compared with non-GSI sites. The average soil Ca concentration in GSI sites was 13,265 mg kg^{-1}, which is over two times greater than mean values found in eastern US soils. Other studies have demonstrated increasing soil Ca concentrations along urban to rural gradients (Pouyat et al., 1991; Lovett et al., 2000). We hypothesize that the greater soil Ca concentrations could be due to concrete leaching or to accumulation of road sand applied during winter storm events. High Ca soil concentrations are not a human health concern, and certain forms of Ca are important for plant nutrition (Hirschi, 2004), although high concentrations have also been found to facilitate growth of invasive species over the growth of native species (Huebner et al., 2014; Kuhman et al., 2011). Soil Ca concentrations were on the order of 110 times greater than those found by Huebner et al. (2014) to support domination of *Rosa multiflora* Thunb. in Pennsylvania forests. Further assessment of ecological impacts would need to be assessed using extractable concentrations.

The average I concentrations in GSI sites were 148% greater than concentrations in non-GSI sites. The average I concentration was 62 mg kg^{-1}, which is 52% greater than the mean values found in eastern US soils. Iodine concentrations in GSI soils could be greater than concentrations in background areas due to road salt drainage.

Table 5. Regression coefficients of micro- and macronutrient concentrations from multiple linear mixed-effects regressions of log(concentrations)† of macro- and micronutrients.

Predictor	Macronutrients				Micronutrients							
	Ca	K	S	Co	Cr	Cu	I	Fe	Mo	Mn	Se	Zn
GSI‡ in place (yes/no)	0.50¶ (0.1)§	−0.14¶ (0.06)	−0.25¶ (0.13)	−0.19¶ (0.09)	−0.09 (0.05)	−0.01 (0.09)	0.9 (0.3)	−0.08 (0.05)	0.03 (0.07)	<0.01 (0.06)	−0.08 (0.04)	0.1 (0.1)
GSI type												
Planter	−0.02 (0.2)	0.1 (0.2)	−0.4 (0.7)	0.48¶ (0.15)	0.08 (0.1)	0.4 (0.2)	−0.1 (0.8)	0.22¶ (0.09)	−0.06 (0.2)	0.07 (0.1)	0.2 (0.1)	0.61¶ (0.25)
Bumpout	−0.4 (0.2)	0.32¶ (0.12)	−1.35 (0.6)	1.01¶ (0.24)	0.38¶ (0.13)	0.3 (0.2)	0.3 (0.8)	0.48¶ (0.13)	−0.32¶ (0.16)	0.62¶ (0.18)	0.1 (0.1)	0.1 (0.2)
Rain garden	0.4 (0.2)	0.28¶ (0.13)	0.04 (0.6)	0.40¶ (0.15)	0.23¶ (0.08)	0.50¶ (0.16)	0.5 (0.6)	0.29¶ (0.11)	0.1 (0.2)	0.51¶ (0.16)	0.08 (0.1)	0.67¶ (0.21)
Other various#	−0.2 (0.3)	0.35¶ (0.11)	0.9 (0.6)	0.29 (0.14)	0.01 (0.1)	0.3 (0.2)	−1 (0.6)	0.2 (0.1)	−0.25 (0.12)	0.1 (0.1)	0.25¶ (0.11)	0.1 (0.2)
Wetland	−0.2 (0.3)	0.01 (0.2)	1.3¶ (0.5)	0.2 (0.2)	−0.2 (0.1)	0.1 (0.3)	−1.3 (0.9)	−0.04 (0.1)	−0.4 (0.2)	−0.07 (0.2)	0.43¶ (0.14)	<0.1 (0.4)
Gully repair	−0.2 (0.3)	0.40¶ (0.1)	0.2 (0.5)	0.23¶ (0.11)	0.1 (0.1)	0.2 (0.2)	−0. 5 (0.7)	0.25¶ (0.07)	−0.1 (0.1)	0.1 (0.1)	0.1 (0.1)	0.1 (0.2)
Receives street drainage (yes/no)	0.28¶ (0.12)	−0.1 (0.1)	−0.2 (0.2)	−0.22¶ (0.07)	−0.1 (0.1)	−0.24¶ (0.1)	0.91 (0.37)	−0.16¶ (0.05)	0.16 (0.07)	−0.1 (0.1)	−0.02 (0.1)	−0.2 (0.1)
Sample size	377	377	380	381	380	381	380	380	381	378	381	381

† The dependent variable is represented by log($Y + s$), where s is half of the minimum non-zero value of each metal concentration.

‡ Values in parentheses are SE.

§ GSI, green stormwater infrastructure. Tree trench serves as reference class.

¶ Effect estimates that had $p < 0.05$ (adjusted for geology, soils, and land use) in addition to covariates shown. Full model details are given in Supplemental Fig. S2.

Basin, swale, grassed waterway, berm. Refer to Fig. 1 for photographs of each GSI type.

Table 6. Regression coefficients of heavy metals and other elemental concentrations from multiple linear mixed-effects regressions of log(concentrations)† of macro- and micronutrients.

Predictor	Heavy metals			Other elements										
	Cd	Hg	Pb	Ag	As	Ba	Cl	Ni	Rb	Sb	Sn	Sr	Ti	Zr
GSI‡ in place (yes/no)	−0.06 (0.08)§	−0.30¶ (0.09)	−0.36¶ (0.18)	−0.50 (0.13)	−0.03 (0.09)	−0.09 (0.06)	−0.01 (0.2)	0.1 (0.2)	−0.25¶ (0.08)	−0.25¶ (0.10)	−0.42¶ (0.13)	−0.02 (0.07)	−0.07¶ (0.03)	−0.25¶ (0.04)
GSI type														
Planter	−0.07 (0.3)	−0.1 (0.2)	1.09¶ (0.28)	<0.01 (0.4)	0.59¶ (0.22)	0.2 (0.1)	1 (0.8)	−0.05 (0.4)	0.4 (0.2)	−0.48¶ (0.20)	0.1 (0.3)	0.2 (0.2)	0.03 (0.1)	0.1 (0.1)
Bumpout	−0.4 (0.4)	−0.1 (0.3)	0.91¶ (0.3)	0.03 (0.5)	0.50¶ (0.25)	0.52¶ (0.17)	2 (1)	−1.68¶ (0.47)	1.00¶ (0.17)	0.6 (0.6)	−0.1 (0.4)	0.3 (0.2)	0.01 (0.10)	−0.1 (0.1)
Rain garden	−0.1 (0.3)	0.1 (0.2)	0.60¶ (0.27)	0.2 (0.5)	0.3 (0.2)	0.26¶ (0.11)	−0.5 (0.7)	0.83 (0.36)	0.40¶ (0.17)	−0.1 (0.3)	0.3 (0.3)	0.2 (0.2)	−0.1 (0.1)	−0.33¶ (0.11)
Other various#	0.4 (0.3)	0.48¶ (0.23)	0.73¶ (0.26)	1.09¶ (0.5)	0.44¶ (0.19)	0.23¶ (0.11)	−1.47¶ (0.73)	0.3 (0.3)	0.44¶ (0.15)	−0.5 (0.3)	0.56¶ (0.28)	0.1 (0.1)	0.1 (0.1)	0.1 (0.1)
Wetland	−0.3 (0.4)	0.1 (0.2)	1.15¶ (0.41)	−0.1 (0.5)	0.3 (0.3)	0.3 (0.2)	2.16¶ (1)	0.3 (0.4)	0.27¶ (0.14)	−0.4 (0.3)	0.6 (0.4)	0.2 (0.2)	−0.01 (0.1)	−0.02 (0.1)
Gully repair	0.4 (0.2)	0.63¶ (0.23)	0.4 (0.3)	1.28¶ (0.53)	0.3 (0.2)	0.1 (0.1)	−1.18¶ (0.51)	0.98¶ (0.41)	0.45¶ (0.12)	−0.61¶ (0.31)	0.4 (0.3)	0.01 (0.2)	0.1 (0.1)	0.02 (0.1)
Receives street drainage (yes/no)	−0.1 (0.2)	−0.2 (0.1)	−0.49¶ (0.13)	−0.2 (0.2)	−0.57¶ (0.15)	−0.3 (0.3)	−0.26¶ (0.08)	−0.2 (0.1)	−0.2 (0.6)	−0.1 (0.2)	−0.20¶ (0.08)	0.03 (0.1)	−0.2 (0.2)	−0.03 (0.04)
Sample size	381	381	381	381	381	381	381	381	381	381	381	381	337	381

† The dependent variable is represented by log($Y + s$), where s is half of the minimum nonzero value of each metal concentration.

‡ GSI, green stormwater infrastructure. Tree trench serves as reference class.

§ Values in parentheses are SE.

¶ Effect estimates that had $p < 0.05$ (adjusted for geology, soils, and land use) in addition to covariates shown.

Basin, swale, grassed waterway, berm. Refer to Fig. 1 for photographs of each GSI type.

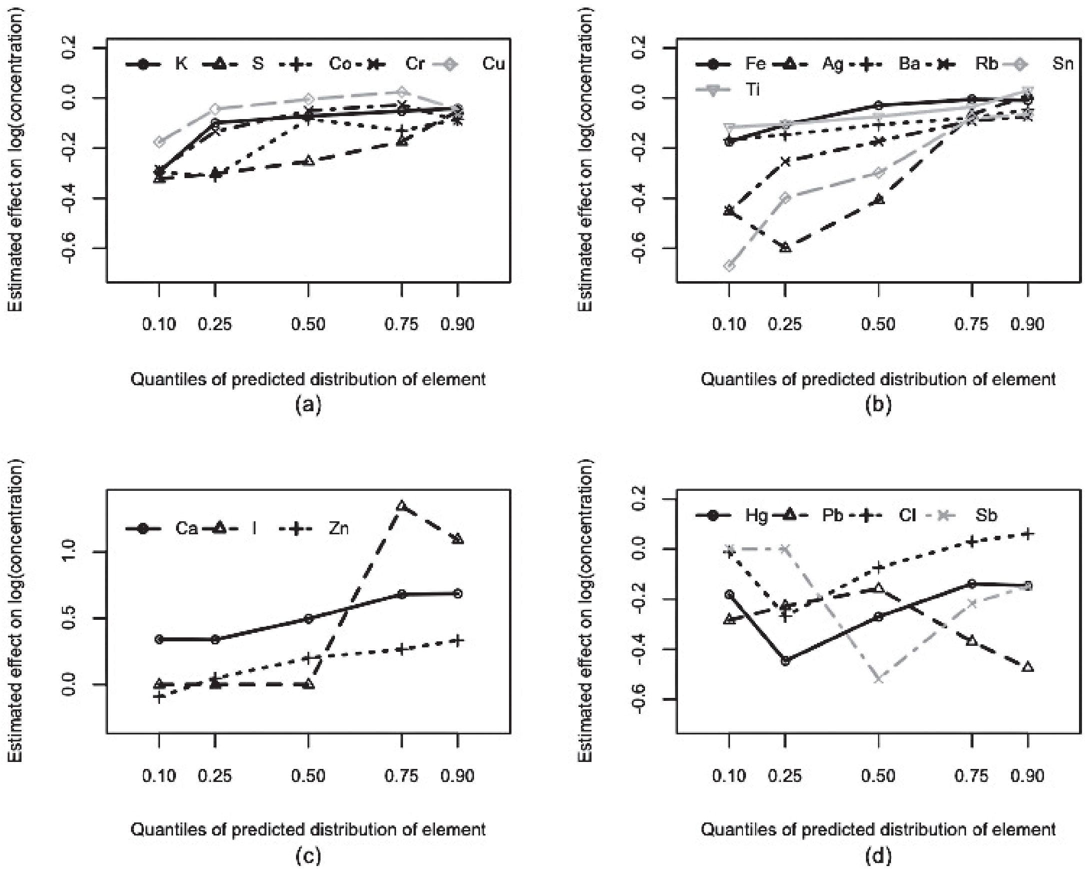

Fig. 3. Treatment effects on elemental concentrations at quantile levels.

Quantile regression helped illustrate the fact that GSI installment has an effect not only on mean values but that the effect is especially strong at greater concentrations for certain elements, including Ca and I. In other words, in GSI sites with relatively high elemental concentrations (Ca, I, and others), surrounding concentrations at non-GSI sites are also relatively low; there is more of a stark contrast at these sites between soils within GSI sites and the soils immediately surrounding. In addition, analyses showed substantial negative treatment effects at low concentrations: when GSI soil concentrations, especially of Ag, Rb, Sn, and Zn, are low, surrounding soil concentrations for these elements are high. Such stark differences between GSI and the surrounding soils could have to do with underlying elemental concentrations in fill used to construct the GSI or with interactions between GSI and surrounding soils via plant matter or surface water runoff. The importance of quantile regression in these analyses is that, whereas change in mean levels, estimated in "traditional analyses," may pose no concern to risk, there may still be significant effect on extremes of the distributions.

In our case, whereas mean regression indicates 65 and 148% increases in Ca and I concentrations, respectively, this underestimates the impact of GSI on the sites with very high background concentrations, where the effects are on the order of 99 and 199% (at the 0.9 quantile level), respectively. This is important if we are concerned about exceedance of guidelines in terms of contamination at specific sites with already "high/concerning" concentration (such that remediation can be targeted efficiently) rather than "on average" (wasting resources on sites where effects are small and not giving due attention to sites where the hazard of exceeding guidelines is more likely). More generally, there is a concern among public health scholars and practitioners about the importance of a shift in distribution of exposure/hazard that may appear as a small (i.e., not clinically meaningful) shift in mean but may actually be important if the entire distribution shifts and tails of the distribution affect a substantial number of people. This argument holds if we assume that "treatment" does not alter the shape of distribution of exposure/hazard, but quantile regression allows us to examine this directly and, in our case, demonstrates that inferences on population impact based on shift in mean alone can be misleading and can both overstate and understate the impact.

Mixed-effects mean regression models found no statistically significant associations with underlying geology. However, principal component analyses found that Fe, K, Co, and Rb were positively intercorrelated, which could signal the presence of soils derived from the Piedmont Plateau geologic formation, which is rich in Fe and Co. In addition, there were only spurious associations with underlying urban soil type (30% lower K, 40% lower Se and Ba, and 16% lower Ti at GSI sites compared with non-GSI sites). We might expect underlying geology and soils to have a more prominent effect on soil elemental concentrations. However, most of the project area is underlain by urban soils that are "fill" and were possibly moved from one location to the other, which represents an unpredictable effect. The differences in pollutant concentrations between GSI and non-GSI sites could therefore be due to the effect of soil disturbance during GSI installation and insufficient time for accumulation of these metals within GSI facilities compared with undisturbed control locations. However, we did not find significant interaction effects with age of GSI in any elements except Cl and Ni.

The portable XRF is widely used to detect metals in environmental soil samples and, compared with other methods (e.g., atomic absorption spectrophotometry), is more accessible and portable (Kalnicky and Singhvi, 2001; Radu and Diamond, 2009). However, the limits of detection can be greater for this unit compared with more costly equipment, and the method is frequently considered only a screening tool rather than a fully quantitative approach. Measurement standard deviations might reflect this limitation. Although our comparison analyses indicated no statistical differences between acid digestion and XRF measurements in some metals, not all elements analyzed by XRF were confirmed by the conventional method. Values of XRF-measured concentrations of elements other than Pb, Zn, and Cu should not be interpreted as absolute values but rather as indicators of potential risk. In addition, we measured total elemental concentration and did not measure potential environmental availability or mobility of these elements. These values provide an initial screening, which should be followed by studies of mobility of target elements out of the GSI soils and into the stormwater itself.

Conclusions

The City of Philadelphia initiated an ambitious program to construct GSI throughout the city to promote infiltration, evapotranspiration, and capture and use or reuse of stormwater runoff and thereby reduce combined sewer overflows. This study provides a unique evaluation of soil elemental concentrations in GSI projects constructed over a decade in a legacy city of the United States. Our study represents a unique assessment of whether contamination is occurring in GSI soils because urban stormwater drainage areas often accumulate elements of concern. This study found that elements of concern to human health were either no different or were lower in GSI soils compared with non-GSI soils. Urban soils surrounding new GSI installments in legacy cities such as Philadelphia could be of greater concern for chemical contamination despite the role of GSI in stormwater drainage. Nevertheless, heavy metal concentrations in all study samples were on average greater than recommended values for ecological and residential uses.

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