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RESEARCH ARTICLE

Risk assessment and interpretation of heavy metal contaminated soils on an urban brownfield site in New York metropolitan area

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Abstract In this study, soil samples were collected at 22 sites in Liberty State Park, New Jersey, in 2005, for metal enrichment and potential ecological risk assessment. The geoaccumulation index (I_{geo}) showed that enrichment levels of trace metals followed an order of Cu > Pb > Zn > As > Cr > Hg while the potential ecological risk factor (E_r^i) indicated that the potential ecological risk of the metals was in the order of Cu > Pb > As > Hg > Zn > Cr. Among these 22 sites, this investigation identified 9 sites at moderate ecological risk, 3 sites at considerable ecological risk, and 4 sites at high ecological risk according to the potential ecological risk index (RI). Hierarchical cluster analysis (CA) of soil metal concentrations separated the study sites into four groups, which are supported by the significant difference in RI values. Geographically, three regions in the Liberty State Park brownfield site were determined based on the CA results and RI values. Subarea 1 had low ecological risk while subareas 2 and 3 had a greater potential for ecological risk. Significant correlations of Pb with Cr and Zn were observed in subareas 2 and 3, respectively. This study shows that statistical approaches coupled with a risk assessment index provide a more comprehensive interpretation of land contamination than a single approach in support of planning land redevelopment.

Keywords Brownfield · Heavy metal · Soil risk index · Statistical analysis · Contamination assessment

Highlights • Metal enrichment level and ecological risk were investigated at a brownfield
• Risk indexes were used for evaluation of soil metal contamination
• Three subareas were identified based on risk assessment index and cluster analysis

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Introduction

Urban brownfields are abandoned lands previously contaminated by anthropogenic development. They are generated as the results of land use transition in the areas of rapid economic development within the urban context and most brownfield lands are contaminated with heavy metals and/or organics (Alker et al. 2000, Thornton et al. 2008). Heavy metals, in particular, are not degradable and tend to remain in soil and accumulated in organisms through uptake and transportation along food chain. Once metal present in an organism reaches a threshold concentration, many key lifecycle metabolic activities such as photosynthesis, respiration, and reproduction will be disturbed (Baker 1981, Gulten 2011).The ecotoxicological impact of heavy metals makes it necessary to consider potential ecological risk before brownfield redevelopment. Redevelopment of brownfields is critical to cities in the middle of transformation because it meets the urgent need for open space, provides employment opportunities, and reduces

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risk associated with abandon lands (De Sousa 2000, 2001, Greenberg et al. 2001, Nijkamp et al. 2002, Tedd et al. 2001, Thornton et al. 2007). However, regardless of the advantage of urban economy stimulation, brownfield redevelopment is often hampered due to the high potential risk of contaminants and the high cost of remediation projects (Garção 2015, Leger et al. 2016, Rudland et al. 2001, Thornton et al. 2007). Therefore, identification of source, transportation, and fate of contaminants will assist remediation project (Bolan et al. 2014, Suthersan 1999). However, the concentration of contaminants in soil could vary significantly within a small area due to the heterogeneous nature of soil (Garção 2015). Hence, a risk assessment of contamination must be done after a site has been abandoned for years.

Although several soil risk indexes have been developed for the evaluation of both soil metal enrichment level and possible ecological adverse effects, they provide neither the information about internal relationship between contaminants nor constructive suggestions on the choice of remediation alternatives (Alloway 2010, Gupta et al. 1996). Similarly, many studies apply statistical approaches to provide a better interpretation of the soil contaminated data and identify internal relationship between soil properties (Astel et al. 2011, Qian et al. 2014, Reimann et al. 2002, Stanimirova et al. 2009). However, statistical approaches were seldom used to assist the decision making of the contribution of each identified source to the total pollutant mass (Astel et al. 2011). Therefore, new alternatives that better interpret the potential risk and the internal relationship between contaminants are needed for the interpretation of metal contaminated soils.

The goal of this study is to evaluate brownfield soil contamination and identify the sites with different pollution source for future urban management and redevelopment by integrating both different evaluation indexes and statistical analysis approaches. It is expected that by a combination of the soil risk assessment indexes with classical statistical analysis methods can divide brownfields into different clusters that feature specific potential risk levels facilitating the remediation process. This study provides a new site assessment protocol, which integrates statistical approaches and evaluation indexes for a brownfield site.

Materials and methods

Study site

Liberty State Park is located in Jersey City, New Jersey, along the shore of New York Harbor (Fig. 1). It is part of the New York Metropolitan area, one of the most densely populated urban areas in the world. Liberty State Park was originally a coastal wetland and that was filled for use as a rail yard and station between the 1860s and 1930s by the Central Railroad of New Jersey (CRRNJ). The rail yard was under operation for over a century. The operation slowed down over the decades following World War II and eventually ceased in 1969 when CRRNJ declared bankruptcy. Due to its unique position and historical value, the land was purchased by the State of New Jersey and converted into a park which opened in 1976 (LSP 2008, NJDEP 1995). A soil survey conducted in 1990 by the NJDEP indicated the site had high levels of metals (NJDEP 1995). Although the State conducted several mitigation projects, such as clean soil capping and asphalt isolation on part of the site and reopened them to the public, a

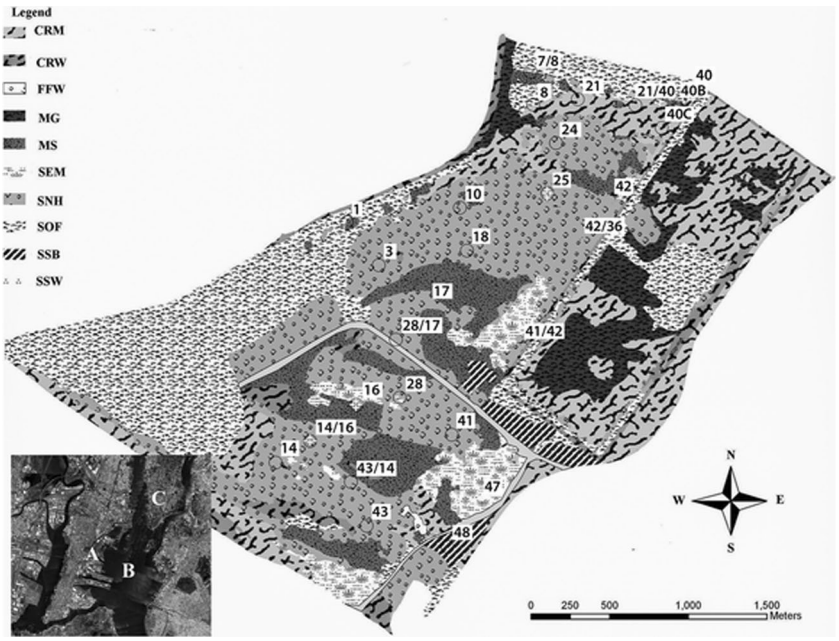

Fig. 1 Map of Liberty State Park in 2015 with study sites numbered. Key map of the New York City metropolitan area A) Liberty State Park, B) Upper New York Bay, C) Manhattan. Vegetative community key: CRM Phragmites australia's and Artimisia, CRW Pharagmites australia's wet, FFWFloodplain Forest Wetland, SEM Semi Emergent Marshlands, MS Maritime Shrublands, SNH Successional Northern Hardwood Forest, SOF Successional Old Field, SSB Successional Shrublands, SEM Shallow Emergant Marsh

251-acre interior section was still left contaminated and was isolated from public reach (Fig. 1) (Gallagher et al. 2008, LSP 2008, NJDEP 1995). However, as the location of this brownfield within the park is very close to a residential area and is accessible to many organisms, the ecological and human health risks have been questioned.

Field work and sample analysis

Based on vegetation assemblage boundaries map created from a vegetative inventory conducted in 2005 (Gallagher et al. 2008), 22 sites were selected for this study in 2005 (Fig. 1). On each site, three soil samples were collected in a 1-m triangular pattern at a depth of 10 to 25 cm, corresponding to a region of the greatest root densities. Their GPS coordinates were recorded at the same time with MC-GPS (Corvallis Microtechnology, Inc. Corvallis, OR, USA) at a 1-m accuracy (Gallagher et al. 2008). Soil samples for metal analysis were sieved through < 125-μm nylon mesh to remove organic detritus and gravels then thoroughly mixed. Then, the soil samples were oven-dried at 60 °C for approximately 48 h to a constant weight. 0.5 g soil was weighted for each sample and treated with 10 ml trace metal-grade HNO_3 in a Teflon bomb for microwave digestion using Mars-5 microwave digester (CEM Corp.). The digestion was conducted at 170 °C for 30 min. Once the digestion was finished, acid extracts were then placed on a hot plate and dried up to paste-like liquid and re-diluted with 1% HNO_3 to 10 ml for the AAS analysis. For the quality control purpose, the method blank and standard reference material (SRM) 1944 issued the National Institute of Standards and Technology (NIST) was digested along with the soil samples. Each set of analysis contained 12 soil samples. The analytical values of the metals of interest are within the range of the certified values. To overcome chemical and ionization interferences, standard solutions were prepared with the same matrix of the samples. Arsenic concentration was determined using Perkin ElmerZ5100 graphite furnace AAS with Zeeman background correction in the presence of $Mg(NO_3)_2$/$Pd(NO_3)_2$ as a matrix modifier. A cold-vapor AAS with a MAS-50D mercury analyzer (Bacharach, Inc.) was used for Hg concentration measurement (Gallagher et al. 2008).

Data analysis

Geoaccumulation index assessment

Geoaccumulation index (I_{geo}) compares the metal concentration in soil samples with its background concentration. The index was originally applied for sediment contamination assessment (Ghrefat et al. 2011, Müller 1969, Srinivasa Gowd et al. 2010), and then further used for soil contamination evaluation (Srinivasa Gowd et al. 2010). It is expressed as

$$I_{geo} = log_2\left(\frac{C_n}{1.5B_n}\right) \tag{1}$$

where C_n is metal concentration in the soil sample and B_n is background metal concentration of the study region. In this study, the New Jersey soil metal background concentrations, which are the average concentrations reported by EPA (EPA 2005a, 2005b, 2005c, 2007a, 2007b, 2008), were used as the metal background concentrations. Since no reference value of Hg is available in the soil metal background concentration developed by EPA, work by Sanders (2003), who collected 284 natural soil samples from urban Piedmont region, urban coastal plain and rural areas of New Jersey was used. As Liberty State Park locates in the urban Piedmont region, we selected the median Hg concentration (0.18 μg g^{-1}) in this region as the background concentration (Sanders 2003). Factor "1.5" was used to adjust potential variations of the background values caused by lithological variations. Seven accumulation categories are defined according to the I_{geo} values: $I_{geo} < 0$ indicates no contamination; $0 < I_{geo} < 1$, no contamination or moderate contamination; $1 < I_{geo} < 2$, moderate contamination; $2 < I_{geo} < 3$, moderate or strong contamination; $3 < I_{geo} < 4$, strong contamination; $4 < I_{geo} < 5$, strong or extreme contamination; and $5 < I_{geo}$, extreme contamination (Ghrefat et al. 2011).

Ecological risk assessment

Risk index (RI) is commonly used to assess the degree of metal ecological risk in soil and evaluates the overall toxicity when metals are combined together. The toxicity of one specific metal can be determined by toxic factor (T_r^i) (Hakanson 1980, Sun et al. 2010). According to Hakanson (1980),

$$C_f^i = \frac{C_D^i}{C_R^i} \tag{2}$$

$$E_r^i = T_r^i \times C_f^i \tag{3}$$

$$\mathrm{RI} = \sum_{i=1}^{m} E_r^i \tag{4}$$

where RI is the potential ecological risk index,E_r^i is the sum of potential risk ofindividual metal, T_r^i is the toxic-response factor for a given metal, C_f^i is the contamination coefficient, C_D^i is the metal concentration of soil sample, and C_R^i is the background concentration of the metal in soil. Based on Hakanson's approach, the toxic-response factor for As, Cr, Cu, Pb, Hg, and Zn are 10, 2, 5, 5, 40, and 1, respectively. This toxic-response factor is also widely accepted in many studies as a substitution for the real toxic-response factor

(Hakanson 1980, Xu et al. 2008). For each metal, Hakanson (1980) designed five classification categories, i.e., the low potential ecological risks (E_r^i <40), moderate potential ecological risks (40<E_r^i <80), considerable potential ecological risks (80<E_r^i <160), high potential ecological risks (160<E_r^i <320), and very high potential ecological risks (E_r^i >320). When considering the summation of potential ecological risk of a specific cite, the overall ecological risks RI can be divided into four categories based on the RI value, i.e., low ecological risks (RI < 150), moderate ecological risks (150 < RI < 300), considerable ecological risks (300 < RI < 600) and very high ecological risks (RI > 600) (Zhu et al. 2012).

Statistical analysis

The Shapiro-Wilk method for data normality distribution test indicated that the data was not normally distributed. Thus, the nonparametric method was applied to the data sets. Cluster analysis was used to merge the data into subgroups based on internal relationships between variables while maximizing the dissimilarity between those subgroups. In this study, hierarchical cluster analysis was applied to investigate the closeness of soil metal concentrations among the 22 sites. The type of joining algorithm used to amalgamate clusters was Ward's method and the metric for measuring distance for the raw and standardized data was Euclidean distance. In brief, Ward's method assesses the relationship between each cluster by calculating the total sum of squared deviations from the mean of a cluster, while Euclidean distance is determined by the actual arithmetic difference between two values (Burns and Burns 2008). The nonparametric Wilcoxon method was applied to test the significance of differences between the data groups. A correlation matrix was conducted to identify the relationship between soil metal concentrations in study sites. Least square linear regression was applied to test the linear correlation between metals in each subgroup. All the statistical data analysis was conducted using SYSTAT, while spatial analysis of metal distributions and site potential ecological risks was performed using ArcGIS.

Results

Soil contamination in the Liberty State Park

Among the 22 sites, most of them had a coefficient of variance for each metal greater than 20%, indicating uneven metal distributions at each site. In addition, a few sites had relatively higher metal concentrations than the rest of the sites (Fig. 2).

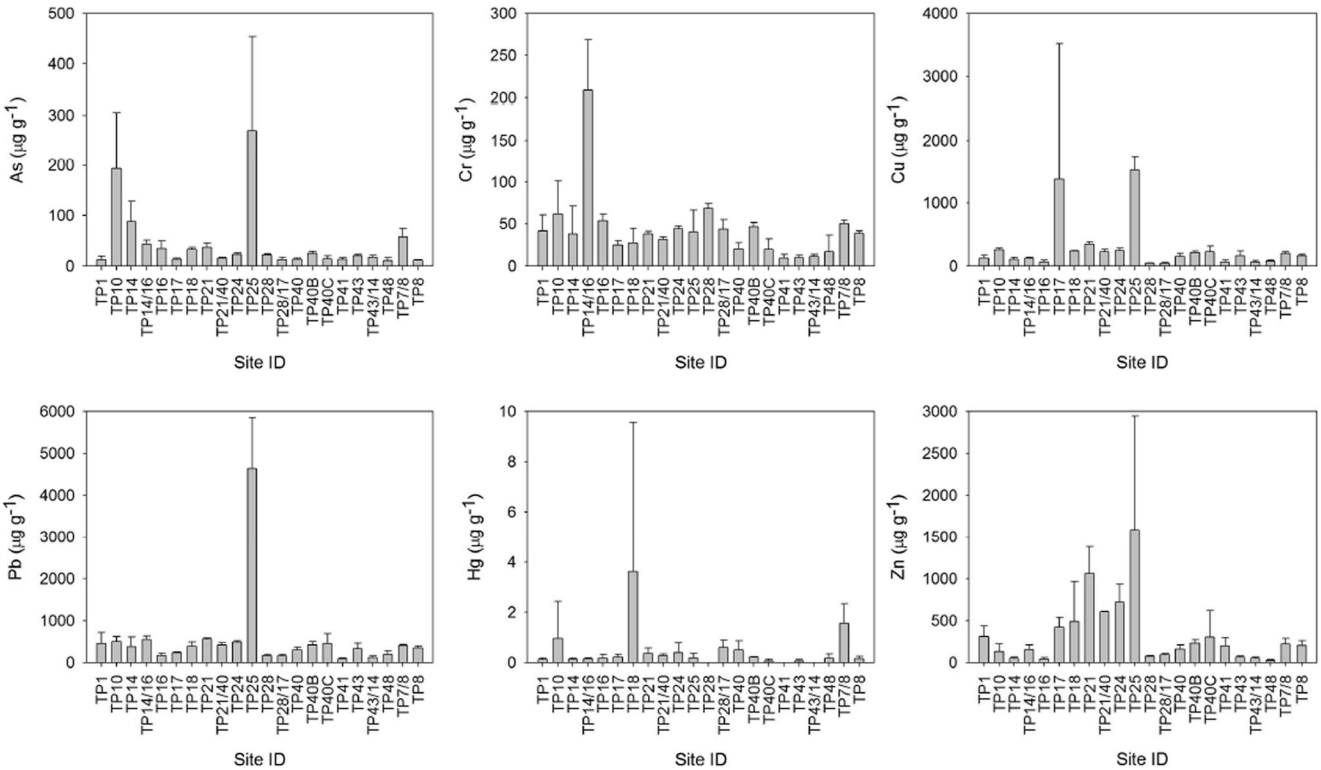

Fig. 2 Comparisons of metal (As, Cr, Cu, Pb, Hg, and Zn) concentration in the 22 study sites. The results indicate that the soil metal concentrations have significant spatial variation

For example, sites TP10, TP14, and TP25 had relatively high As concentration, whereas site TP14/16had high Cr concentration, and sites TP17 and TP25 had a high Cu concentration. Lead concentration was extremely high at site TP25 (4640 ± 1210 μg g^{-1}), while sites TP10, TP18, and TP7/8 showed a higher level of Hg (0.96 ± 1.48 μg g^{-1}, 3.62 ± 5.95 μg g^{-1} and1.57 ± 0.77 μg g^{-1}, respectively) than the other sites. Zinc concentration at site TP25 (1586 ± 1358 μg g^{-1}) was higher than the rest of the sites. In addition, the range of each metal concentration varied widely. The concentrations ranged from 10.7–270 μg g^{-1} for As, 10.9–335 μg g^{-1} for Cr, 53.6–2200 μg g^{-1} for Cu, 96.6–6670 μg g^{-1} for Pb, n.d.– 3.62 μg g^{-1} for Hg, and 10.9–2330 μg g^{-1} for Zn (Fig. 2). It is obvious that the distributions of the six metals (As, Cr, Cu, Hg, Pb, and Zn) at the 22 sites are heterogeneous with considerable spatial variations.

Soil metal accumulation evaluation

To further evaluate the levels of metal enrichment, the metal geoaccumulation indexes (I_{geo}) were estimated. The magnitude of I_{geo} in Liberty State Park followed the following pattern: Cu > Pb > Zn > As > Cr > Hg. The geoaccumulation index of Cu (I_{geo}(Cu)) at all sites was greater than 1.0, indicating that this metal species had been enhanced throughout the site. Forty-six percent of the sites were strongly to extremely contaminated and 54% sites were contaminated by Cu moderately to strongly. The I_{geo} of Pb at most fell in the categories between moderate contamination and strong contamination: 23% sites were moderately contaminated by Pb, 32% of the sites were moderately or strongly contaminated, and 36% of the sites were strongly contaminated. Geoaccumulation index of Zn (I_{geo} (Zn)) ranged from −0.41 to 5.59, spreading over all the seven categories of from no contamination to extreme contamination. Moderate to strong Zn contamination in the soils were found on 59% of the sites, while strong to moderate Zn contamination were found at 18% of the sites. According to the geoaccumulation index of As (I_{geo} (As)), 77% of the total sites were classified as uncontaminated to moderately contaminated; only three sites were considered strongly or extremely contaminated. For both Cr and Hg, more than 60% sites had negative I_{geo} values, indicating no contamination, and 18% of the sites showed no contamination to moderate contamination, indicating that these two metals were not major contaminants in this urban brownfield. The results show that the most enriched contaminants in the study area were Cu, Pb, and Zn.

Ecological risk evaluation

The potential ecological risk of the six metals decreases in the order of Cu > Pb > As > Hg > Zn > Cr. The ecological risk of each individual metal is summarized as follows: Nine percent of the sites were under extremely high potential ecological risk of Cu, 59% sites were at moderate to considerable potential ecological risk, and only 32% sites were immune from ecological risk caused by Cu. The majority (59%) of the sites were considered as moderate potential ecological risk for Pb. However, only 10% of the sites were under considerable to extremely high potential ecological risk of Pb. More than 60% of the sites fell into the class of low ecological risk caused by As. Only a few sites were under considerable (9%), high (5%) and extreme (5%) potential ecological risk of As. Although 64% sites were not considered as contaminated by Hg according to the geoaccumulation index, the risk analysis based on E_r^i showed that 55% of the sites were still considered at low Hg potential ecological risk, 14% of the sites at moderate potential ecological risk, 18% of the sites at considerable potential ecological risk, 5% of the sites at high potential ecological risk, and 5% of the sites at extremely high potential ecological risk. Although Hg has relative low metal enrichment as suggested by I_{geo}, it can have a high potential ecological risk as indicated by E_r^i. Zinc fell into two categories, i.e., low potential ecological risk and moderate potential ecological risk. Although 59% of the sites were moderately or strongly contaminated by Zn, only 9% of the sites were considered as moderate potential ecological risk, implying that high Zn enrichment was not equivalent to high potential ecological risk; the ecological risk caused by Cr was considered low at all the sites, implying that Cr did not cause significant potential ecological risk in this brownfield ecosystem.

Risk Index (RI), represented by the summation of the individual of As, Cr, Cu, Pb, Hg, and Zn, indicates to what extent the urban brownfield ecosystem was suffering from the soil contaminated from the above six metals (Sun et al. 2010). The RI at the 22sites in this study ranged from 65.3 to 1693, suggesting that the ecological risk in the investigated sites varies widely. Among the 22 sites, six sites were classified as low ecological risk, nine sites as moderate ecological risk, and three sites as considerable ecological risk. It should be noted that sites TP10, TP17, TP18, and TP25 were at very high ecological risk (Fig. 3). At the sites of moderate ecological risk, the E_r^i of Cu yielded the highest weight of the RI in each site, followed by Pb and As. At the sites of considerable risk, the E_r^i of Cu contributed most to the RI, followed by As and Hg. Finally, the E_r^i of Pb followed by that of Cu and Hg contributed the most to the RI of the greatest risk. Therefore, the potential risk caused by Cu, Pb, As, and Hg should be considered first during the soil remediation.

Cluster analysis

Hierarchical cluster analysis was performed on metal concentration data from the 22 urban brownfield sites. As shown in Figure 4, the sampling sites are divided into four clustering

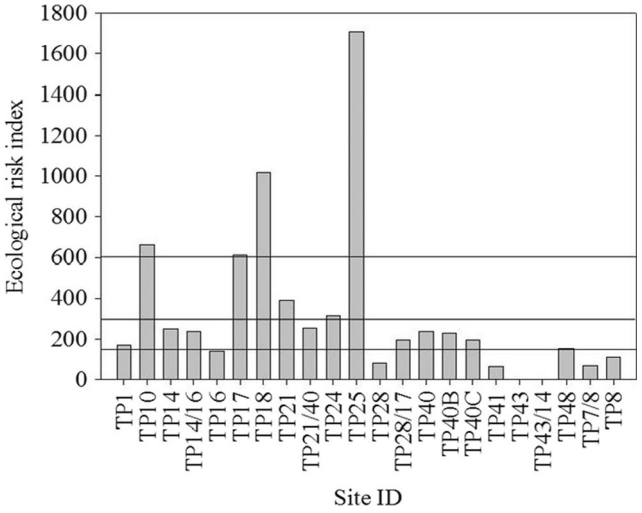

Fig. 3 Risk Index (RI) of As, Cr, Cu, Pb, Hg, and Zn in 22 sites. The lines represent different level of ecological risk: low ecological risk (RI < 150); moderate ecological risk (150 < RI < 300); considerable ecological risk (300 < RI < 600);very high ecological risk (RI > 600)

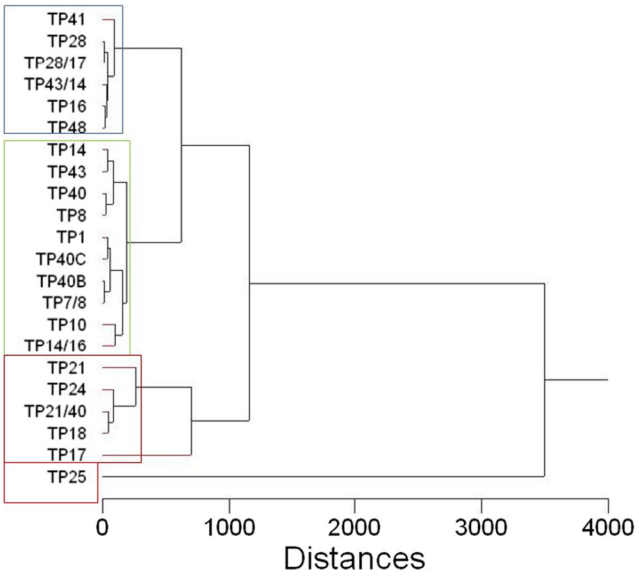

Fig. 4 Hierarchical dendogram for 22 sites obtained from hierarchical cluster analysis (Linkage: Ward; Distance: Euclidean)

groups (Cluster 1, 2, 3, and 4) based on soil metal (As, Cr, Cu, Hg, Pb, and Zn) concentrations. Cluster 1 includes six sites (TP41, TP28, TP28/17, TP43/14, TP16, and TP48) located at the southern part of the study area. Cluster 2 consists of ten sites (TP14, TP43, TP40, TP8, TP1, TP40C, TP40B, TP7/8, TP10, and TP14/16) spreading at the periphery of the park. Cluster 3 contains five sites (TP21, TP24, TP21/40, TP18, and TP17) located at the northeast and center of the brownfield. Cluster 4 has only one site, i.e., TP25 (Fig. 4).

The mean concentrations of all metals except Cr at the sites in Cluster 1 were lower than those in the other three clusters (Table 1). However, it was reported that the mean concentrations of As and Pb in the soil for plant growth were above the ecological soil screen level (the threshold concentration of contaminants in soil that potentially pose ecological risk to living organism, SSL) set by USEPA (Gallagher et al. 2008), suggesting that these two elements may have adverse effect on the terrestrial plants growing at these sites in cluster 1 (EPA 2005a, 2005b). In cluster 2, the mean concentrations of metals in soils were higher than that in cluster 1 (Table 1). Except Cr, the mean concentrations of all other metals in the soils at the cluster 2 sites were above plant SSL, suggesting possible toxic effect on vegetation. Therefore, the sites in cluster 2 had potential toxic effects on the plants. The sites in cluster 3 had higher concentrations of Cu, Pb, Hg, and Zn than that in the cluster 1 and cluster 2 sites (Table 1). As shown in Table 1, not only mean concentrations of these metals exceeded the SSL limit but also the concentrations of As and Cu at several sites in cluster 3 were above the NJ non-residential criteria. Therefore, the potential risk of As and Cu was especially high at the sites in cluster 3. Finally, the mean concentrations of As, Cu, Pb, and Zn were the highest at site TP25, the only site in cluster4. Table 1 shows the mean concentrations of As, Cu, Pb, Hg, and Zn at this site exceeded both plant SSL level and non-residential NJ limit, suggesting that site TP25 is not favorable for either terrestrial plant growth, or public non-residential land use.

To further investigate the difference in potential ecological risk among the four clusters, a nonparametric paired test was performed to compare RI values between each cluster. Cluster 4 was excluded from this exercise because there was only one site (TP25), making it not appropriate for statistical analysis. According to the results from the Wilcoxon paired test, significant differences were found between cluster 1 and cluster 2 ($p = 0.0014 < 0.05$), cluster 2 and cluster 3 ($p = 0.04 < 0.05$), and cluster 1 and cluster 3 ($p = 0.01 < 0.05$) (Fig. 5).

Discussion

The accumulation of metals in urban soils has a chronic impact on urban ecosystem through either direct exposure or indirect trophic transfer. In order to minimize the potential risk of metals from the contaminated soil, land use management and remediation are required. The four cluster groups correlated well with the RI levels. All the low ecological risk sites were found in the cluster 1 region, which is the southeast corner of the study site. This area was identified as subarea 1 to distinguish it from the whole study area. Most sites in cluster 2 (subarea 2) have moderate ecological risk and several with considerable ecological risk. These sites are distributed at the rim of the study area (Fig. 1). Finally, sites with considerable or extreme ecological risk belonged to cluster 3 (subarea

Table 1 Descriptive statistics of metal concentrations of urban brownfield soils in the four group sites in Liberty State Park

Sites	As	Cr	Cu	Pb	Hg	Zn
Group 1	$n = 6$					
Mean	18.6	33.9	62.4	149	0.16	79.9
Standard deviation	9.13	24.7	12.9	40.4	0.23	63.2
Minimum	10.7	9.70	44.3	86.1	0.00	24.9
Maximum	35.1	68.5	76.4	196	0.60	198
Group 2	$n = 10$					
Mean	47.8	53.6	174	415	0.39	183
Standard deviation	56.6	56.8	49.5	77.0	0.50	88.6
Minimum	11.9	10.2	103	303	0.06	50.0
Maximum	193	209	257	552	0.57	309
Group 3	$n = 5$					
Mean	24.6	33.4	487	420	0.98	658
Standard deviation	10.7	7.77	500	122	1.48	250
Minimum	12.8	25.0	224	236	0.21	421
Maximum	37.4	44.0	1377	563	3.62	1058
Group 4	$n = 1$					
Mean	270	40.4	1527	4640	0.18	1586
Standard deviation	184	26.1	219	1210	0.19	1358
Minimum	69.8	23.1	1277	3249	0.02	739
Maximum	440	70.5	1685	5442	0.39	3153
Plant soil scree level[a]	18.0	N/A	70.0	120	0.30[d]	160
New Jersey background concentration[b]	7.00	13.9	14.0	35.0	0.18[e]	22.0
Non-residential clean up[c]	20.0	6100	600	600	270	1500

[a] EPA Ecological soil screen level (EPA 2005a, 2005b, 2005c, 2007a, 2007b, 2008)

[b] EPA Guidance for developing ecological soil screen levels (2005)

[c] N.J.A.C 7.26D Remediation standards (2012)

[d] Swartjes, F.A. (1999), Dutch target soil screening value of Hg, which have negligible risk for ecosystems

[e] Sanders (2003), ambient levels of metals in New Jersey soils. Median concentration of Hg for soil samples collected in urban piedmont

3).These sites are located in the northern part of the study area. Cluster 4 (site TP25) is also located in the northern part and it featured extreme ecological risk, hence cluster 4 is classified with subarea 3 in the following discussion.

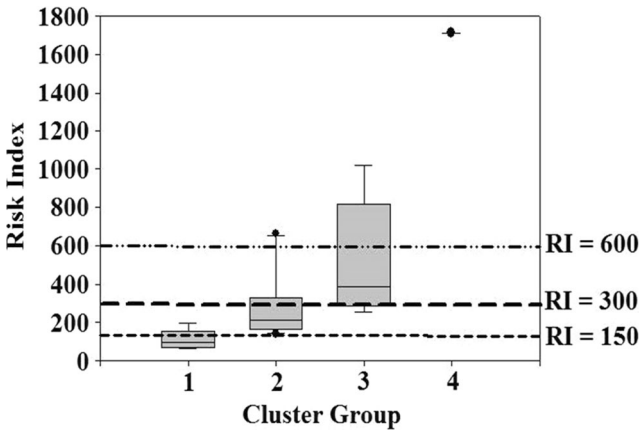

Fig. 5 Nonparametric paired test of risk index values between the sites in the four clusters

The patchy distribution of metals (As, Cr, Cu, Hg, Pb, and Zn) within the subareas indicates the extreme heterogeneous metal accumulation. According to an 8-m sampling survey decades before, there were little or no metal contamination in deep soil samples compared to the surficial soil samples (NJDEP 1995, EPA 2005c), indicating soil contamination is the result of the historical transportation operation. Pearson multiple linear correlation analysis was conducted to further test the relationship among the metals within each subarea.

Subarea 1 had a relatively low ecological risk (Table 1 and Fig. 1). In addition, no significant correlation between the six metals was observed in third subarea (Table 2), indicating that the metals in these sites probably did not share the same sources. Subarea 1 sites were originally low-lying areas between the rail road tracks (Fig. 1); while still constructed of fill material, the disturbance as a result of the rail operation could have been less than the rest of the freight yard.

Table 2 Pearson correlation matrix among different metal concentrations in urban soils in three group sites in Liberty Satate Park

	As	Cr	Cu	Pb	Hg	Zn
Group 1						
As	1					
Cr	0.608	1				
Cu	−0.099	−0.767	1			
Pb	0.194	0.541	−0.155	1		
Hg	−0.21	0.213	−0.351	0.321	1	
Zn	−0.337	−0.272	−0.142	−0.773	−0.089	1
Group 2						
As	1					
Cr	0.149	1				
Cu	0.392	−0.222	1			
Pb	0.412	0.741*	0.193	1		
Hg	0.484	−0.01	0.437	0.073	1	
Zn	−0.417	−0.061	0.271	0.263	0.005	1
Group 3						
As	1					
Cr	0.333	1				
Cu	−0.557	−0.573	1			
Pb	0.707	0.815	−0.793	1		
Hg	0.491	−0.371	−0.317	−0.077	1	
Zn	0.641	0.718	−0.451	0.895*	−0.337	1

*Correlation is significant at 0.05 level

Subarea 2 exhibited strong spatial variation in As, Cr, and Hg concentrations (Table 1). This is possibly due to the industrial activities over the years in subarea 2 (Table 2). For example, the mean concentrations of Cu, Pb, and Zn in this subarea was greater than that in subarea 1, suggesting subarea 2 received and accumulated extra Cu, Pb, and Zn from a specific source. According to Fig. 1, subarea 2 was originally covered by railroad tracks, which railroad might be the source of the three metals. Previous studies on soil contamination showed significant elevated concentrations of Cu, Zn, Cd, and Mn in the soil near railroads in Sichuan, China, and high concentrations of Zn, Cd, and Pb in the soils near railroad transport in the Qinghai-Tibet railroad (Liu et al. 2009, Zhang et al. 2012). Hence, the historic railroad transportation might contribute to the elevated concentrations of Cu, Pb, and Zn in this area. However, there were no significant correlations observed between these three metals (Table 2). One possible reason is that the operation of the rail yard for one century had introduced metals from various cargo or maintenance activities, which diluted the relationship between Zn, Pb, and Cu. Another possible reason is that plant uptake of Zn and Cu as nutrients at different rates has been demonstrated (Gallagher et al. 2008). Such extraction of the mobile fraction of metals from the soil to biomass could change the original metal concentrations (French et al. 2006, Imperato 2003, Qian et al. 2012, Smolders et al. 2009). Finally, significant ($p = 0.018 < 0.05$) correlation between Cr and Pb was observed in subarea 2. It is known that major industrial sources of Cr in soil include tannery, wood preservation, chromium chemical production, and stainless steel. With regard to Pb, anthropogenic activities such as lead mining and smelting, shooting ranges, automobile exhausts, and pesticides are well-known sources (Alloway 2010). While there is no record of the industrial activities above on the study area according to the published documentations (EPA 2005b, 2008, LSP 2008), there is a record indicating that the site had received filling material from an electroplating plant in Jersey City (NJDEP 1995). It is possible that the filling material from the electroplating plant is the source of Cr and Pb in the area.

In subarea 3, As and Cr concentrations are close to that in subarea 1, suggesting limited additional sources of these two elements. The average concentrations of Hg were the highest among all the three subareas (Table 1). As shown in the historical map of the study area (Fig. 1), subarea 3 sites were previously covered by the railroads for transportation. This area was where the trains loaded and unloaded cargo. Compared to other area in the park, this area appears to have the greatest contaminant loading. The major anthropogenic activities that release Hg to the environment include mining and smelting of ores, combustion of coal fuels, cement production, and gold production (Alloway 2010). In view of the intense transportation activities on sites in subarea 3, and the fact that the rail yard served as a major coal transfer hub and the early steam train engines were coal fired, combustion of coal is likely the source of Hg in this area. Although the mean concentration of Pb at the sites in subarea 3 were close to that in subarea 2, the concentrations of Cu and Zn were much higher at the sites in subarea 3 (Table 1). In addition, a significant ($p = 0.04 < 0.05$) linear correlation between Pb and Zn was found, suggesting that Pb and Zn may share the same pollution sources. It was mentioned early that Pb and Zn were two featured contaminants from railroad operations (Liu et al. 2009, Zhang et al. 2012). Therefore, coal and diesel combustion from the trains was likely the major contaminant source in subarea 3.

After combined classical site ecological risk assessment indexes with statistical analysis approaches, the study site was divided into three different subareas. Remediation alternatives could be decided based on the ecological risk level and contamination source of each subarea. For subarea 1, considering the relatively low contamination risk, monitored natural attenuation should be seriously considered. In subarea 2, because the contamination level is moderate and the major contaminants, Cu, Cr, Pb, and Zn are either nutrients of plant or could be fixed in soil as immobilized form, phytoremediation combined with soil amendment will possibly control the bioavailable contaminant concentration level under the regulatory threshold. Our previous research had indicated that

modification of soil pH and organic matter content could effectively alternate the bioavailability of Zn and Cu in the study site (Qian et al. 2012). Finally, the subarea 3 has the highest metal contamination level and some of the metals, such as Hg, will put the ecosystem and the public under high risk. Therefore, remediation alternatives for subarea 3 should be more progressive. Soil capping and groundwater contamination monitor, soil washing after excavation, and/or soil amendments are some of the options that might help to control the contamination in subarea 3 (Suthersan 1999). However, it should be noted that if more detailed information such as groundwater contamination level and soil texture is available, subareas with more specific remediation approaches could be identified.

Conclusion

Metal (As, Cr, Cu, Hg, Pb, and Zn) concentrations in this brownfield site show heterogeneous distributions. The enrichment level of these metals, as indicated by I_{geo} follows an order of Cu > Pb > Zn > As > Cr > Hg, while the contribution of each metal to the potential ecological risk is in an order of Cu > Pb > As > Hg > Zn > Cr. In this study, three subareas with different contamination level and pollution source are identified based on RI and cluster analysis of the 22 sites. Subarea 1 has a low contamination level with no distinct contamination source. Subareas 2 and 3, located in an area where the railroad track was laid a century ago, have relatively high concentrations of Cu, Pb, and Zn. Pearson linear correlation analysis showed significant correlations between Cr and Pb in subarea 2 and between Pb and Zn in subarea 3, indicating that electroplating plant material and railroad transportation could be the major contamination sources in these two areas, respectively. The elevated Hg concentration in subarea 3 is possibly due to combustion of coal in the past. Management plans for the restoration of this brownfield can be developed based on potential ecological risks and metal contamination source, with different emphases on each subarea indicated in this study. While regular monitoring on metal concentrations in the soils and organisms might be the best option for Subarea 1, further assessment of metal bioavailability and groundwater contamination in subareas 2 and 3 is necessary before making the remediation plan.

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Compliance with ethical standards

Ethical statement The manuscript submitted to ESRP has not been submitted to more than one journal for simultaneous consideration. The manuscript is based on our original work and has not been published previously. No data in this manuscript were fabricated or manipulated (including images) to support the conclusions. All the references were properly cited to acknowledge other work. Consent to submit this manuscript has been received explicitly from all co-authors. Authors whose names appear on the submission have contributed sufficiently to the scientific work and therefore share collective responsibility and accountability for the results. The research did not involve any human participants and/or animals.

Conflict of interest The authors declare that they have no conflicts of interest.

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