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Assessment of sources of heavy metals in soil and dust at children's playgrounds in Beijing using GIS and multivariate statistical analysis

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ABSTRACT

Potentially toxic elements such as heavy metals are ubiquitous in the environment. Risk-based environmental management relies upon identifying pollution sources, pathways, and the exposed population. In a Chinese urban setting, many residents live in high-rise buildings without private gardens. Therefore, the main residential risk of exposure to contaminated soils and dusts may be associated with public open spaces. As children are the most vulnerable receptor, playgrounds represent an important yet often overlooked exposure point. The present study assessed plausible sources of heavy metals at children's playgrounds in a representative metropolitan environment. Soil and equipment dust samples were collected from 71 playgrounds across Beijing, which were analyzed for 11 different heavy metals. Principal component analysis (PCA) was used to identify the latent constructs which control heavy metal variability and reflect potential sources. Cluster analysis (CA) was conducted to group sampled locations, which provided further insights on plausible sources. The main factors extracted from the PCA were then subject to geostatistical analysis. The systematic combination of GIS with multivariate statistical analysis proved valuable for elucidating anthropogenic and natural sources. Elevated Be, V, Cr, Mn, Co, Ni, As in playground soils were found to derive mainly from the natural background (spatial autocorrelation = 2 km), while elevated Cu and Pb was attributed to traffic activities (spatial autocorrelation = 17 km), especially along the routes of Beijing's inner ring-roads, the major roads toward the northwest and northeast, and the international airport. These results suggest that heavy metals in playground equipment dust may derive mainly from atmospheric deposition of air pollution of both natural and anthropogenic origin (spatial autocorrelation = 11–13 km). Among them, Be, V, Mn, Co, Cu, As, Pb were attributed to atmospheric pollution deriving from the north of Beijing, brought by the prevailing northern wind in the winter season; whereas, Cr and Ni may possibly be brought from the southeast by the summer season winds. Knowledge of anthropogenic vs. natural origins of heavy metals in playgrounds is critical in assessing health impact and designing policy instruments for metropolitan areas.

1. Introduction

Heavy metal contamination of the environment has become a serious issue globally (Järup, 2003), often requiring active cleanup (Hou and Al-Tabbaa, 2014; O'Connor et al., 2018a; Zhao et al., 2018). China's soil pollution is particularly serious. A 2014 national soil quality survey revealed that 16.1% of China's soil exceeded environmental quality standards (MEP, 2014), with heavy metals being the most widely distributed constituents of concern. In response to social and environmental pressure associated with soil pollution (Hou et al., 2018; Liu et al., 2018), the Chinese central government published a "Soil Pollution Prevention and Control Action Plan" (Hou and Li, 2017), which requires the national and local governments to conduct a detailed national scale soil pollution survey of mainly industrial sites and agricultural land. While such efforts are important for gaining an overall assessment of the level and extent of soil pollution across China, they overlook some unique and potentially important exposure sites such as children's playgrounds. As most Chinese urban residents live in high-rise buildings, the typical exposure scenario for residents is not like those living in single family houses in the US, where soils in front and backyards present typical exposure points. Instead, urban recreational spaces, especially children's playgrounds, represent an important

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source of exposure.

Heavy metals represent widely distributed environmental pollutants due to extensive usage in industrial and consumer products such as lead based paint, copper wire, nanomaterials, etc. (O'Connor et al., 2018b; Shen et al., 2018b; Wang et al., 2019; Zhang et al., 2018). A limited number of studies have examined heavy metal contamination at children's playgrounds. Wong and Mak (1997) measured Cd, Cu, Pb, and Zn concentrations in soil and equipment dust at seven playgrounds in Hong Kong. They found that these heavy metals were elevated in both soil and dust, especially in samples collected from playgrounds around commercial and industrial areas. A separate study in Hong Kong measured several heavy metals in ground dust at 89 playgrounds, finding high Zn (1883 mg/kg), Cu (143 mg/kg), and Cr (263 mg/kg) concentrations (Ng et al., 2003). De Miguel et al. (2007) studied heavy metal concentrations at 20 municipal playgrounds in Spain, and found that As concentrations represented the highest health risk.

In order to manage risks posed by heavy metals in playground soils and dusts, it is important to first identify potential sources. Existing studies indicate that the spatial distribution of heavy metals in soil may be attributed to either geogenic or anthropogenic sources (Hou et al., 2017). The distribution of naturally occurring heavy metals can be highly heterogeneous, with significantly elevated concentrations existing in certain types of soil. Human activities including mining, manufacturing, urban runoff, and atmospheric deposition have led to significantly elevated heavy metal concentrations in other areas (Romic and Romic, 2003; Zhang, 2006). The identification of potential sources of heavy metals at playgrounds has only be examined by a few studies to date. Most of these have relied upon empirical evidence, rather than distinguishing the different sources (see Table 1).

GIS and multivariate analysis are tools that can be used to assess heavy metal spatial distribution and identify sources. This is due to features such as visual display and geostatistic derivation with GIS, and the ability to establish latent constructs and identify non-trivial sources with multivariate analysis. To make better use of these tools, researchers have increasingly been using them in combination. However, as a recent critical review pointed out, most studies have only done this in a somewhat loose way (Hou et al., 2017), with only a small number systematically integrating GIS with multivariate analysis (Ha et al., 2014; Lu et al., 2012). To our knowledge, no existing studies have systematically integrated GIS and multivariate analysis for assessing the spatial distribution and potential sources of soil heavy metals at children's playgrounds.

The present study aimed to assess the concentrations of multiple heavy metals in playground soil and equipment surface dust in Beijing, China. Beijing was chosen as a major metropolitan area that epitomizes a Chinese urban environment and life style. A total of 71 playgrounds were selected to provide a good spatial coverage of Beijing. GIS and multivariate analysis methods were systematically integrated to assess the spatial distribution and potential sources of these heavy metals.

2. Materials and methods

2.1. Study location and field sampling

The study was conducted in Beijing, which covers an area of 164,100 km^2 with a population of 21.73 million people (BJStats, 2017). The climate in Beijing is continental monsoon (Beijing Weather, 2013). A total of 71 playgrounds were selected for the present study (see Fig. 1), based on a 25 km × 40 km grid pattern of the entire Beijing area, augmented with a 5 km × 6 km grid pattern in the central districts (main city), and a 8 km × 10 km grid in a newly developed district (Tongzhou). Samples were obtained from the playgrounds in April–May 2017. Soil was obtained from the top 10 cm of exposed soils. Dust samples were collected from the surfaces of playground equipment using paint brushes. The detailed sampling procedures were described in Peng et al. (2019).

2.2. Sample analysis

Both soil and dust samples were oven-dried at 60 °C, sieved to < 2 mm, homogenized, and microwave digested in accordance with USEPA Method 3051A (USEPA, 2007). The concentrations of 11 heavy metals (Be, V, Cr, Mn, Fe, Co, Ni, Cu, As, Se, Pb) were quantified by Inductively Coupled Plasma - Mass Spectrometry (ICP-MS) (XSERIES 2, Thermo-Fisher, USA). The analysis and quality assurance/quality control (QA/QC) was in accordance with USEPA Method 6020A (USEPA, 1998).

2.3. Data analysis

Statistical analysis was conducted using SPSS (IBM, USA). Pearson product-moment correlation analysis was conducted to estimate the linear dependence between variables. Principal component analysis (PCA) was conducted with quartimax rotation to identify latent factors (Bořůvka et al., 2005; Hou et al., 2014). The extracted factors were then used in geostatistical analysis. Cluster analysis (CA) was also used where set of data objects of similar characteristics were divided into different groups by algorithms that minimize intra-group variability and maximize inter-group variability (Young and Hammer, 2000). This approach is often used with soil heavy metals data as a confirmation of PCA (Hou et al., 2017). In the present study, we used CA to group sampled location points rather than heavy metal concentrations, which provides new information regarding the spatial distribution and potential sources of soil and dust contamination.

Geostatistic and GIS analysis were conducted using data obtained

Table 1
Source of heavy metals in children's playground in selected cities.

City	Year	Studied heavy metals	Attributed source(s) of heavy metals	References
Soil				
Hong Kong, China	1997	Cd, Zn, Cu, Pb	Traffic	(Wong and Mak, 1997)
Madrid, Spain	2007	33 trace elements	Sand substrate replacement	(De Miguel et al., 2007)
Athens, Greece	2010	Cr, Zn, Ni, Pb, Co, Mn, Cu, Fe	Atmospheric deposition	(Massas et al., 2010)
France	2012	As, Cd, Cr, Cu, Mn, Pb, Sb, Sr, V	Unknown	(Glorennec et al., 2012)
Port Harcourt, Nigeria	2017	Cd, As, Co, Pb, Hg, Cu, Zn, Fe, Ni, Cr, Mn	Zn & Mn: natural background Cu, Cr, Fe: anthropogenic	(Joy and Uchenna, 2017)
Beijing, China	2018	Be, V, Cr, Mn, Fe, Co, Ni, Cu, As, Se, Pb	Be, V, Cr, Mn, Co, Ni, As: natural background Cu, Pb: traffic	This study
Dust				
Hong Kong, China	1997	Cd, Zn, Cu, Pb	Traffic	(Wong and Mak, 1997)
Hong Kong, China	2002	Zn, Cu, Cd, Cr, Pb, Fe, Mn	Local traffic	(Ng et al., 2003)
France	2012	As, Cd, Cr, Cu, Mn, Pb, Sb, Sr, V	Unknown	(Glorennec et al., 2012)
Beijing, China	2018	Be, V, Cr, Mn, Fe, Co, Ni, Cu, As, Se, Pb	Be, V, Mn, Co, Cu, As, Pb: regional atmospheric deposition associated with northern wind Cr, Ni: atmospheric deposition possibly associated with southeastern wind	This study

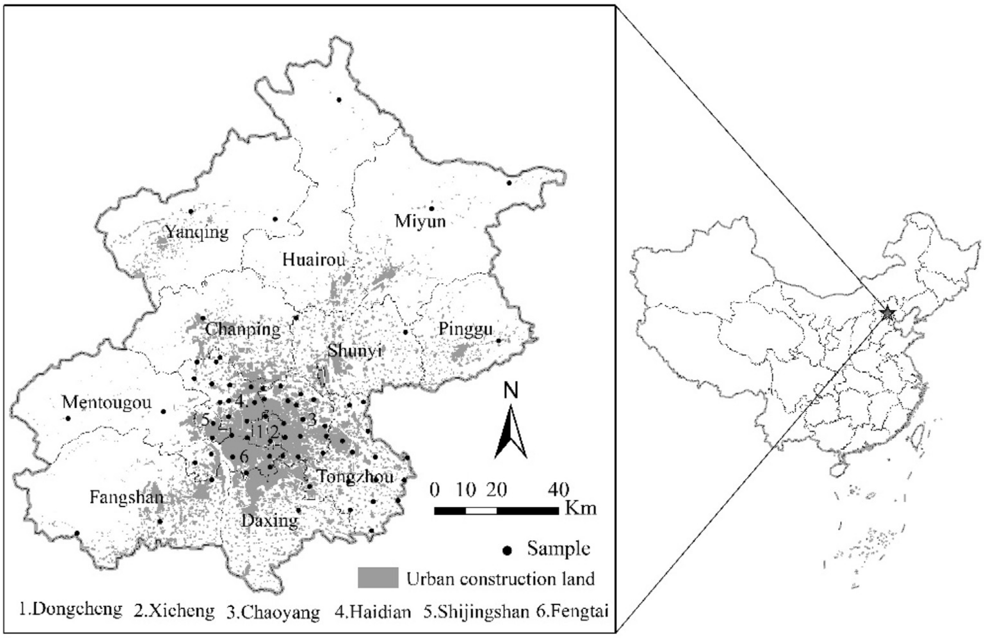

Fig. 1. Children's playground sampling locations in Beijing.

Table 2
Summary statistics of soil and dust heavy metal concentrations for 71 playgrounds in Beijing.

Element	Sample type	Minimum (mg/kg)	Maximum (mg/kg)	Median (mg/kg)	Mean ± SD (mg/kg)	Kurtosis	Skewness	C.V.(%)	Guide[1] value
Be	Soil	0.60	1.60	1.00	1.08 ± 0.23	0.34	0.10	21.24	15
	Dust	0.47	2.21	0.77	0.88 ± 0.27	7.68	1.79	30.16	
V	Soil	85.15	192.9	106.9	110.7 ± 16.30	8.30	2.03	14.73	165
	Dust	23.59	75.25	47.83	48.64 ± 10.61	−0.03	0.19	21.81	
Cr	Soil	32.18	105	53.38	54.69 ± 12.18	5.78	1.76	22.28	400
	Dust	0.00	383.	71.05	78.87 ± 64.32	6.64	1.94	81.55	
Mn	Soil	425.7	1084	588.7	600 ± 112	3.89	1.31	18.70	n/a
	Dust	319.5	779.8	512.1	521.7 ± 112.7	−0.54	0.23	21.61	
Fe	Soil	18,640	46,380	26,780	27,738 ± 4604	3.15	1.12	16.60	n/a
	Dust	17,646	65,690	29,480	30,746 ± 9771	2.51	1.42	31.78	
Co	Soil	7.60	21.60	12.20	12.61 ± 2.62	1.35	0.85	20.76	20
	Dust	4.81	24.63	8.47	8.78 ± 2.73	15.54	2.86	31.04	
Ni	Soil	17.89	47.49	33.79	33.70 ± 6.66	−0.59	−0.05	19.77	150
	Dust	0.00	95.40	18.34	22.25 ± 19.89	2.14	1.36	89.39	
Cu	Soil	21.29	155.3	40.59	43.41 ± 17.61	23.27	3.98	40.57	2000
	Dust	0.00	271.7	46.23	52.06 ± 38.74	14.27	2.85	74.43	
As	Soil	10.73	20.73	15.53	15.55 ± 2.24	−0.17	0.12	14.38	20
	Dust	3.51	26.09	9.13	10.02 ± 4.31	2.65	1.40	43.04	
Se	Soil	10.80	28.00	16.90	17.35 ± 3.46	0.42	0.75	19.96	n/a
	Dust	0.00	12.82	3.33	4.04 ± 3.72	−0.75	0.58	91.94	
Pb	Soil	12.28	326.3	27.48	36.58 ± 40.39	39.50	5.78	110.4	400
	Dust	9.55	269.6	64.78	80.27 ± 55.97	1.32	1.22	69.73	

[1] Guide values are based on GB36600—2018 Soil environmental quality: risk control standard for soil contamination of development land.

from the multivariate statistical analysis. A semivariogram was first used to analyze the spatial structure of heavy metal concentrations in soil and dust at playgrounds. The semivariogram, as defined below, is one of the most commonly used functions in assessing spatial autocorrelation (Goovaerts, 1999).

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i + h) - Z(x_i)]^2 \tag{1}$$

where γ(h) is the semivariogram that measures the dissimilarity between locations with h distance and direction, N(h) is the number of data pairs with h distance and direction, $Z(x_i)$ and $Z(x_i + h)$ are the heavy metal concentrations at location x_i and another location with h distance and direction from x_i.

Spatial autocorrelation, i.e. the statistical dependence of intermediate values on spatial proximity, was tested with variograms and covariance functions. The semivariogram analysis was conducted using GS + 9.0 (Robertson, 2008). To evaluate whether heavy metal concentrations had strong spatial autocorrelation or were randomly distributed, the sill, nugget variances, and range were estimated. The principal components identified in PCA were used in the geostatistical analysis. For this, the data were tested for normality, with Box-cox transformation applied to non-normally distributed variables.

For variables with spatial structure, kriging was performed using ArcGIS10.5 (Keranen and Malone, 2018) as the interpolator to derive intermediate values at spatial proximity (Kravchenko and Bullock, 1999). Kriging is the most widely used interpolation approach in studying the spatial distribution of soil heavy metals, originating from the Regionalized Variable Theory (Oliver and Webster, 1990). The generic equation for kriging is described by the following equation

(Oliver and Webster, 1990):

$$z(B) = \sum_{i=1}^{n} \lambda_1 z(x_i) \quad (2)$$

where z(B) is the estimate over an area of land and λ_i are the weights, which sum to one to ensure that there is no bias and, subject to this, are chosen to minimize the estimation variance.

3. Results and discussion

3.1. Heavy metal concentrations in soil and dust

Descriptive statistics for Be, V, Cr, Mn, Fe, Co, Ni, Cu, As, Se, and Pb in soil and dust samples from the children's playgrounds are listed in Table 2, including minimum, maximum, median, mean, standard deviation, kurtosis, skewness, and coefficient of variation (CV). The heavy metal concentrations are compared to national guide values (MEE, 2018). The maximum concentrations of all heavy metals except V, Co, and As, were below the relevant soil quality standard. The kurtosis of the Cu and Pb distributions were relatively high. This indicates the presence of unusually high values, which may have been caused by distinct anthropogenic sources of Cu and Pb.

Table 3 shows the results of the Pearson correlation analysis. For soil heavy metals, strong correlations (coefficients > 0.6) were observed between many of the metals. For instance, Be concentrations were strongly correlated to 70% of the other metals, especially Mn, Co, Ni, and As. Strong correlations suggest a common source, and these metal types may be indicative of soil parent materials of lithogenic origin (Ali et al., 2016; Facchinelli et al., 2001). On the other hand, correlations among the heavy metals in dust were generally weaker. This may suggest that the dust heavy metals were more likely to have been influenced by different unrelated sources, possibly including anthropogenic sources.

3.2. Multivariate statistical and geostatistical analysis

3.2.1. Principal component analysis (PCA)

Principal component analysis (PCA) was conducted for four series of variables: soil heavy metal concentrations, dust heavy metal concentrations, the difference of soil and dust heavy metal concentrations (S-D), and the ratio of soil and dust heavy metal concentrations (S/D). The KaisereMeyereOlkin (KMO) measures of sampling adequacy for the above variables were 0.73, 0.82, 0.77, and 0.76, respectively. The KMO values were all well above a recommended cut-off threshold of 0.5, suggesting there are compact correlations and the PCA should yield distinct and reliable factors (Field, 2009).

Table 4
Results of principal component analysis.

Metals	Soil		Dust		S-D		S/D	
	PC1	PC2	PC1	PC2	PC1	PC2	PC1	PC2
% variance	*44%*	*16%*	*40%*	*13%*	*31%*	*15%*	*33%*	*18%*
Be	0.85	0.02	0.84	−0.21	0.83	−0.01	0.84	0.10
V	0.63	−0.20	0.82	−0.12	0.70	0.23	0.74	0.05
Cr	0.78	0.37	0.41	0.76	0.08	0.81	0.14	0.85
Mn	0.83	0.07	0.82	−0.10	0.82	0.11	0.82	0.12
Fe	0.48	−0.30	0.34	0.05	−0.14	−0.26	−0.32	−0.64
Co	0.94	−0.02	0.69	0.36	0.63	0.53	0.65	0.42
Ni	0.89	0.12	0.49	0.67	0.42	0.70	0.37	0.64
Cu	0.19	0.82	0.60	0.20	0.46	0.19	0.51	0.36
As	0.66	0.16	0.70	−0.24	0.83	−0.18	0.81	−0.07
Se	0.00	−0.02	0.42	0.12	0.08	0.02	0.17	0.14
Pb	0.08	0.88	0.59	0.22	0.17	−0.02	0.34	0.38

Note: Loading stronger than 0.5 are in red font.

Table 4 shows the component matrix post rotation, with two Factors shown for each set of variables. Based on the component matrix, Factor 1 for soil heavy metals captures Be, V, Cr, Mn, Co, Ni, and As, with these metals together indicative of soil parent materials/geogenic sources (Ali et al., 2016; Facchinelli et al., 2001). Factor 2 for soil captures Cu and Pb, which is attributed to anthropogenic pollution. In particular, Cu and Pb pollutants in soil are known to often originate from leaded gasoline, braking, engine wear and other traffic related activities (Zhang, 2006). For dust heavy metals, Factor 1 was loaded on Be, V, Mn, Co, Cu, As, and Pb. Some of these heavy metals can be associated with soil parent materials, while others are associated with anthropogenic pollution, but both would likely have been transported in air and re-deposited as dust. Therefore, dust Factor 1 is attributed to wide-ranging atmospheric deposition. Factor 2 was loaded on Cr and Ni which may be attributed to either local soil parent materials (Hou et al., 2017), or to certain industrial sources such as metal plating. Further discussion on the natural and anthropogenic sources of these heavy metals is provided in Section

Table 3
Correlation analysis of heavy metal concentrations in playground soil and dust, Beijing.

Type		Be	V	Cr	Mn	Fe	Co	Ni	Cu	As	Se	Pb
Soil	Be	1.00										
	V	0.40**	1.00									
	Cr	0.59**	0.43**	1.00								
	Mn	**0.72****	0.43**	**0.60****	1.00							
	Fe	0.35**	0.23	0.27*	0.20	1.00						
	Co	**0.72****	**0.70****	**0.73****	**0.77****	0.34**	1.00					
	Ni	**0.70****	0.46**	**0.71****	**0.68****	0.33**	**0.86****	1.00				
	Cu	0.17	0.03	0.31**	0.19	−0.06	0.17	0.30*	1.00			
	As	**0.64****	0.46**	0.38**	**0.60****	0.19	0.56**	**0.62****	0.32**	1.00		
	Se	0.04	0.39**	−0.05	0.01	−0.03	−0.02	−0.04	0.04	0.46**	1.00	
	Pb	0.07	−0.13	0.48**	0.09	−0.07	0.05	0.11	0.56**	0.08	−0.09	1.00
Dust	Be	1.00										
	V	**0.64****	1.00									
	Cr	0.25*	0.22	1.00								
	Mn	**0.71****	**0.66****	0.33**	1.00							
	Fe	0.17	0.25*	0.12	0.23	1.00						
	Co	0.48**	0.55**	0.49**	0.47**	0.24*	1.00					
	Ni	0.29*	0.40**	0.56**	0.31**	0.10	0.51**	1.00				
	Cu	0.40**	0.36**	0.34**	0.48**	0.18	0.35**	0.36**	1.00			
	As	0.55**	0.49**	0.16	0.43**	0.09	0.39**	0.23	0.36**	1.00		
	Se	0.28*	0.36**	0.16	0.19	0.16	0.14	0.33**	0.30*	0.23	1.00	
	Pb	0.40**	0.30*	0.34**	0.43**	0.25*	0.50**	0.30*	0.34**	0.36**	0.18	1.00

*$p < 0.05$; **$p < 0.01$; coefficients above 0.6 are in bold.

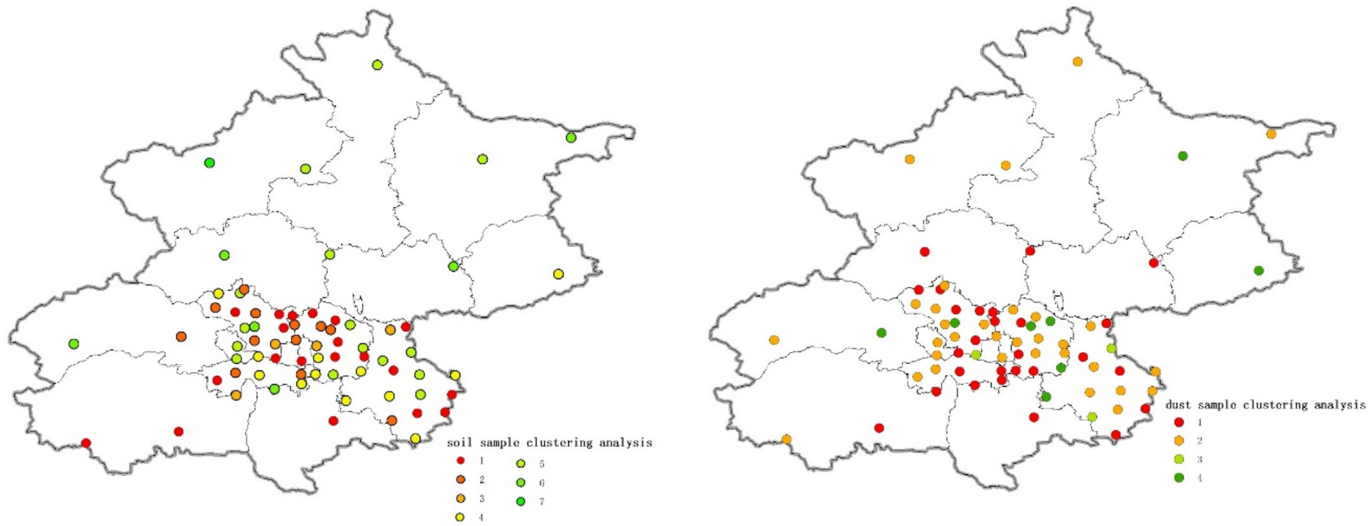

Fig. 2. Cluster Analysis (CA) of soil and dust heavy metal concentrations.

3.3.

The PCA results for both S-D and S/D are similar to those of dust heavy metals. Some existing studies suggest that the atmospheric deposition of dusts and aerosols are a source of soil heavy metal contamination (Davis et al., 2009; Lee et al., 2006). Nicholson et al. (2003) attributed a large portion of heavy metals in agricultural land to atmospheric deposition. Atmospheric heavy metals may derive from variety of natural sources, including forest fires, emission from volcanos, and degassing in the Earth's crust, or from anthropogenic sources, like metal production, mining, oil combustion, coal combustion, and waste incineration (Lado et al., 2008; Nriagu, 1979). The PCA suggests that heavy metals in equipment dust at children's playground in urban area mainly derive from atmospheric deposition (i.e. dust PCA Factor 1), accounting for 40% of the total variance.

3.2.2. Cluster analysis (CA)

Unlike most reported studies which used cluster analysis (CA) to confirm PCA results (Hou et al., 2017), here we also used CA to assess the grouping of spatial locations by transforming the data matrix, with the results plotted in GIS. As Fig. 2 shows, the groupings identified by CA displayed spatial clustering. Sampling locations in the northern part of Beijing, a mountainous area that has little industry, tended to be within the same group of sites less impacted by anthropogenic activities. Southern Beijing and central Beijing had many points in another group indicative of anthropogenic activities, because many industrial facilities are located in southern Beijing, and many commercial and transportation activities occur in central Beijing. In comparison, the CA results for dust heavy metals have a less distinct spatial pattern. This may reflect the fact that dust heavy metals are attributed to long-distance transport and deposition of atmospheric particles, which results in widespread regional pollution, rather than local acute pollution.

3.2.3. Spatial autocorrelation and variogram modeling

The optimal semivariogram models were Gaussian or exponential in most cases (Table 5). Quotient values, derived by the diving nugget variance by the sill, were used to classify spatial autocorrelation (Cambardella et al., 1994). A quotient of < 0.25 suggests strong spatial autocorrelation; 0.25–0.75 suggests moderate spatial autocorrelation; and, > 0.75 suggests weak or no spatial autocorrelation. As Table 5 shows, both principal components of soil heavy metals have quotients < 0.25, suggesting strong spatial autocorrelation. In comparison, principal components of dust heavy metals only have moderate spatial autocorrelation. The relationship between soil and dust heavy metal concentrations also had strong spatial autocorrelation.

Table 5 shows that for soil concentrations, the second principal component varied further in special distance than the first. This is illustrated by the smaller nugget effect and greater range (Goovaerts, 1999). Relating this to the PCA loading results, the 'soil parent material' source is considered to have an autocorrelation distance of ~2 km. This is consistent with the findings of Goovaerts (1999), who reported a range of ~1.6 km for rock type represented by Ni concentrations. In contrast, the 'traffic' source has much larger range, which may be due to the longitudinal extension of traffic routes, rather than cross gradient dispersion of traffic related emissions. A previous study on heavy metals in Beijing suggested that Pb concentrations had a similar autocorrelation distance of nearly 30 km (Hu et al., 2006). The low R2 value of the second soil principal component reveals that it did not fit the model well. This is likely due to the traffic only having spatial autocorrelation along the traffic route itself, but not perpendicularly. This is consistent with existing studies that show Pb and Cu concentrations attenuating quickly in the direction perpendicular to roads (Pagotto et al., 2001).

For dust concentrations, Table 5 shows that the first principal component range was ~10.8 km, and the second was ~12.8 km. Factor 2 of the PCA was loaded on Cr and Ni, which could be attributed to either local soil parent materials or industrial sources (Section 3.2.1). The relatively long range suggests that atmospheric dispersal and deposition of industrial pollution is likely. It should be noted that > 150 data points are usually required for reliable estimates from a semivariogram analysis, and more for direction dependent estimates (Goovaerts, 1999). Therefore, the results reported here are considered indicative.

3.2.4. Kriging of latent construct and GIS analysis

Figs. 3 and 4 show the results derived from kriging the principal components of soil and dust heavy metals, respectively. Fig. 3(a) shows that the soil first principal component has a general decreasing trend from north to south. The higher values overlap well with the northern mountainous area of Beijing. This is consistent with our interpretation that the first principal component is attributed to soil parent materials. The soil in the mountainous region of Beijing is less sandy, and subjected to less severe acidic leaching due to podsolization, and thus contains higher levels of heavy metals of lithogenic origin (Alloway, 2013). Fig. 3(b) shows that the second principal component of heavy metals overlaps with the major traffic routes of Beijing. The roads in the metropolitan area of Beijing are well-known for heavy traffic volume, especially the inner ring roads, the major road to Northwestern China (G6), and the major from Beijing to Northeastern China (S11). In addition, heavy traffic is associated with an international airport in the

Table 5
Results of semivariogram analysis.

Types	Component	Model	Nugget	Sill	Quotient	Range (m)	R2	RSS
Soil	PCA1	Gaussian	0.55	3.11	0.18	2066	0.62	1.43
	PCA2	Spherical	0.01	0.23	0.05	16,630	0.00	0.01
Dust	PCA1	Gaussian	0.75	1.93	0.39	10,770	0.68	0.39
	PCA2	Exponential	0.25	0.66	0.38	12,810	0.70	0.01
S-D	PCA1	Gaussian	0.74	1.58	0.47	1555	0.75	0.21
	PCA2	Linear	0.97	0.97	1.00	16,630	0.00	0.202
S/D	PCA1	Exponential	0.18	1.06	0.17	15,330	0.46	0.23
	PCA2	Exponential	0.15	1.02	0.14	7920	0.01	0.26

northeastern part of central Beijing. As the second principal factor overlaps with the main traffic corridors, it strongly indicates that that Pb and Cu concentrations in Beijing's playground soils are associated with vehicle traffic.

The kriging results for the principal components for playground dust show that the first principal component has a general decreasing trend from northern Beijing to southern Beijing (Fig. 4a), and the second principal component has higher scores along a northwest-southeast corridor (Fig. 4b). The PCA and semivariogram analysis results indicated that both principal components were associated with atmospheric deposition. These two principal components may, therefore, correspond to different sources of dust carried by the different prevalent wind directions in Beijing observed in the winter and summer seasons. Existing studies on the inter-city transport of particulate matter shows that in January, ground level wind and particulate matter influx is mainly from a northern direction, but in the upper level (e.g. 450–600 m), both wind and particulate matter influx is mainly from the northwest. In contrast, in July, wind and particulate matter influx at ground level is from the southeast, while the upper level is from south (Chang et al., 2018). Comparing the meteorological data and atmospheric particulate matter influx with the dust kriging results, it suggests that perhaps the first principal component of playground dust heavy metals is associated with atmospheric pollution from the ground level in the winter, while the second principal component may possibly be associated with atmospheric pollution at ground level in the summer. The above plausible explanation fits not only from the perspective of wind direction, but also from the perspective of heavy metal composition. The first principal component includes Be, V, Mn, Co, Cu, As, Pb, which feasibly originate from soil parent materials blown from the mountainous area in northern Beijing, or from polluting activities that include heavy traffic in the more developed region of northern Beijing. The second principal includes Cr and Ni, which may derive from the direction of Tangshan to the southeast of Beijing, which is associated with Cr and Ni pollution as the largest steel manufacturing area in China. The geographic corridor of higher values across central Beijing shown in Fig. 4b may be associated with the terrain of Beijing, with mountainous areas to the north and south-west.

3.3. Natural and anthropogenic sources of heavy metals

Heavy metal may originate from either natural or anthropogenic sources. Elevated natural background levels require regional management of potential risks. Heavy metals from natural source may have lower bioavailability, because natural soils may undergo weathering and rainfall leaching for thousands to millions of years, leaving only the metals that exist in a highly stable form. On the other hand, anthropogenic sources of heavy metals are more manageable and often involves penalties for polluting behavior, and liability for cleanup operations (polluter pays principal). Therefore, the management of these two types of sources of heavy metals could be dramatically different.

Naturally occurring heavy metals are affected by various factors, such as mineral composition, particle size, and soil organic carbon (Dung et al., 2013). Heavy metals in natural soil can be redistributed by surface runoff (Herngren et al., 2005), weathering, erosion, etc.

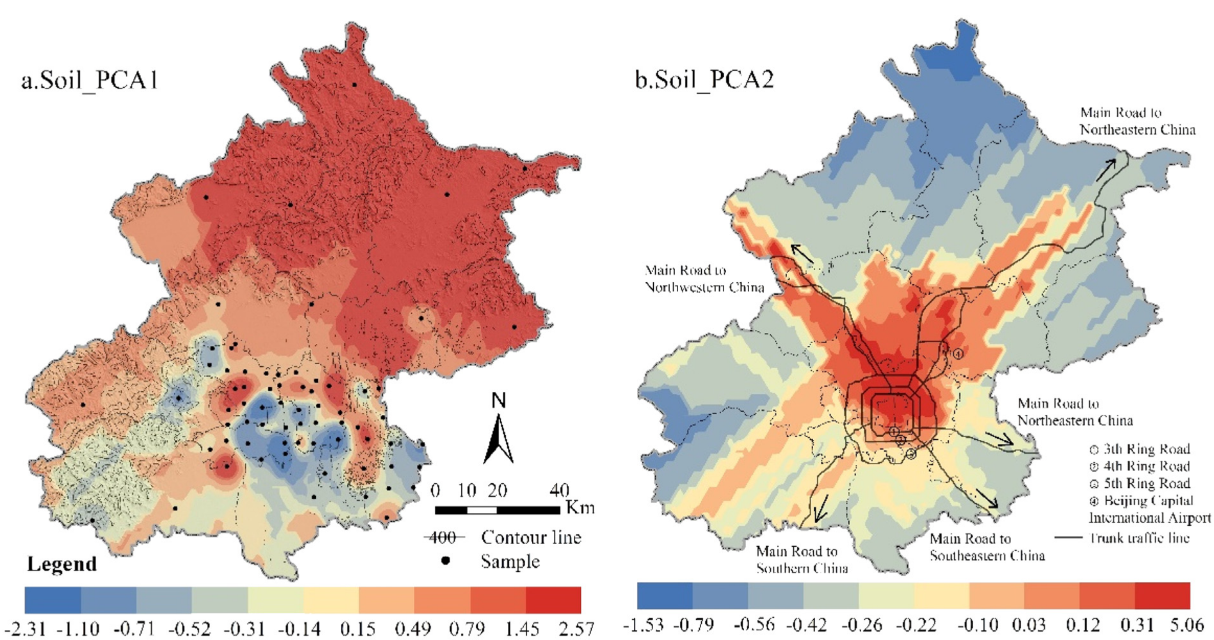

Fig. 3. Principal component distribution (Beijing playground soil): a) First principal component - the distribution strongly correlates with mountainous area of Beijing; and, b) Second principal component - the distribution strongly correlates with major roads and international airport of Beijing.

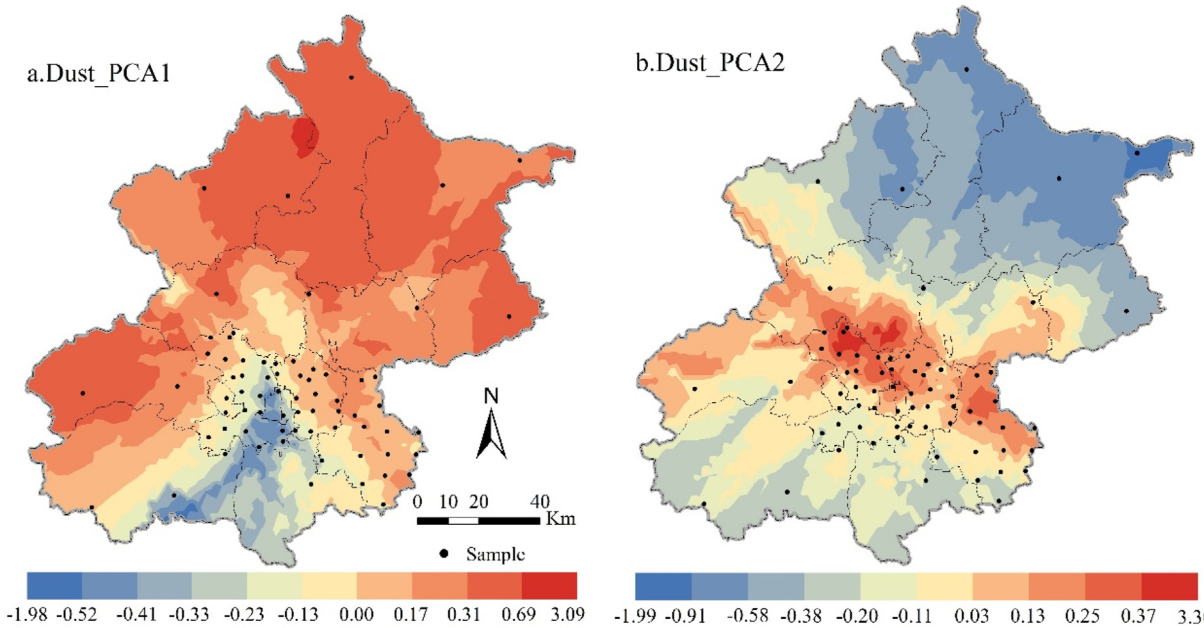

Fig. 4. Principle component distribution (Beijing playground dust): a) First principal component - the distribution is consistent with northern wind in the winter season and atmospheric particulate matter influx from the north; and, b) Second principal component - the distribution matches with southeastern wind in the summer season and atmospheric particulate matter influx from the southeast.

(Nriagu, 1989). Most existing studies have used PCA and CA to confirm natural sources of heavy metals. Different studies have attributed different groups of heavy metals to natural sources. For instance, Ali et al. (2016) attributed Ni, Mn, and Co concentrations to natural sources; Romic and Romic (2003) attributed Fe, Mn, and Ni to natural origin; and Kelepertzis (2014) attributed Ni, Cr, Co and Fe to natural origin. In the present study, Elevated Be, V, Cr, Mn, Co, Ni and As in playground soils were found to derive mainly from the natural background.

Anthropogenic sources of heavy metal may include vehicle exhausts, waste disposal, sewage, industrial emissions (Chen et al., 2005; Hou et al., 2012; Kelly et al., 1996; Li et al., 2001; Wei and Yang, 2010), as well as pesticide and fertilizer applications, livestock manures, and atmospheric deposition (Hu et al., 2006; Micó et al., 2006; Nicholson et al., 2003; Pagotto et al., 2001). Some studies have compared heavy metals concentrations within different types of land use to make conclusions regarding the anthropogenic sources of heavy metals. Recently, researchers have also increasingly used a combination of GIS and multivariate analysis for soil quality assessment. Nearly all of these studies have distinguished natural sources from anthropogenic sources. However, many studies failed to recognize the role of spatial autocorrelation. Unlike deterministic interpolation methods, such as the inverse distance weighted (IDW) method, which are directly based on the surrounding measured values, the kriging method is based on spatial autocorrelation among the measured values. Existing studies have rarely attempted to identify the type of anthropogenic activities which contribute specific heavy metal contamination in soil. Moreover, most existing studies integrating GIS and multivariate statistical analysis that examine regional soil heavy metals have used the default settings. In the present study, we showed that the tighter integration of GIS and multivariate analysis with the use of refined analysis, can provide valuable insights into the identification of anthropogenic sources of heavy metals. Elevated Cu and Pb was attributed to traffic activities, especially along the routes of Beijing's inner ring-roads, the major roads toward the northwest and northeast, and the international airport. For equipment dust, Be, V, Mn, Co, Cu, As, Pb were attributed to atmospheric pollution from the north of Beijing, while Cr and Ni were attributed to atmospheric pollution from the southeast.

3.4. Implications for health risk management and policy making

It is important to identify sources of heavy metal pollution in order to enable sound assessment of human health risks, and to implement necessary mitigation actions where necessary (Maas et al., 2010). Children's playgrounds represent an important exposure scenario for urban residents; therefore, the sources of heavy metals in the soil and dust of these playground have important implications for health risk management and policy making.

For soil at playgrounds, the present study suggests that heavy metals are mostly from natural sources, and traffic activities are probably the biggest anthropogenic source. While most playgrounds in Beijing were found to contain relatively low heavy metals concentrations, with only a few exceedances, there are probably more serious heavy metal contamination at playgrounds in other industrial oriented cities across China. The latest Chinese regulations require management of risk when heavy metal concentrations exceed corresponding screening values (MEE, 2018). Heavy metal contaminated soils maybe managed by a variety of techniques, including soil washing (Shen et al., 2018c; Song et al., 2018), solidification/stabilization (O'Connor et al., 2018c; Shen et al., 2019; Shen et al., 2018a), thermal treatment (e.g. for Hg) (Hou et al., 2016; Ma et al., 2015), chemical reduction (e.g. Cr(VI)) (Liu et al., 2018), and phytoremediation (O'Connor et al., 2018d). For children's playgrounds and their surrounding areas, soil heavy metals should be tested, and risk-based management performed on a site-by-site basis.

For playground dust, the present studies suggest that heavy metals mostly derive from atmospheric deposition. Therefore, it is important to mitigate the influx of particulate matter pollution originating from traffic activities or from industrial activities such as steel manufacturing. Because the source of heavy metals is regional, it is difficult to manage the sources of dust heavy metals on a site-by-site basis. The reduction of heavy metal concentrations and corresponding health risk will require air pollution control measures, including measures to reduce transportation related emissions, industrial emissions, etc.

4. Conclusion

The present study analyzed heavy metal concentrations in soil and dust at 71 children's playgrounds in Beijing. Multivariate statistical analysis, including principal component analysis and cluster analysis, were conducted to identify the pattern of heavy metal concentrations, the latent constructs that control heavy metal variability, and potential sources. Spatial autocorrelation was examined by using semivariogram analysis. Subsequently, kriging of the principal components was conducted. The geostatistical analysis combined with the multivariate statistical analysis results provided a more refined analysis of the sources of heavy metals at these playgrounds. Elevated Be, V, Cr, Mn, Co, Ni, As in Beijing's playground soils are reported to be mainly associated with natural background, which are higher in the mountainous regions due to the nature of the soils. Elevated Cu and Pb levels in playground soil are attributed to vehicle traffic activities, especially those associated with the inner ring roads, the major roads toward northwestern and northeastern Beijing, or to the Beijing international airport. Heavy metals on playground equipment dust were found to mainly derive from atmospheric deposition of particulate matter of both natural and anthropogenic origin. Among them, Be, V, Mn, Co, Cu, As, Pb may originate from atmospheric pollution to the north of Beijing brought about the prevailing northern wind in the winter season; and, Cr and Ni may originate from atmospheric pollution from the southeast of Beijing, possibly brought by winds in the summer season.

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