Science of the Total Environment 541 (2016) 969–976

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Integrating hierarchical bioavailability and population distribution into potential eco-risk assessment of heavy metals in road dust: A case study in Xiandao District, Changsha city, China

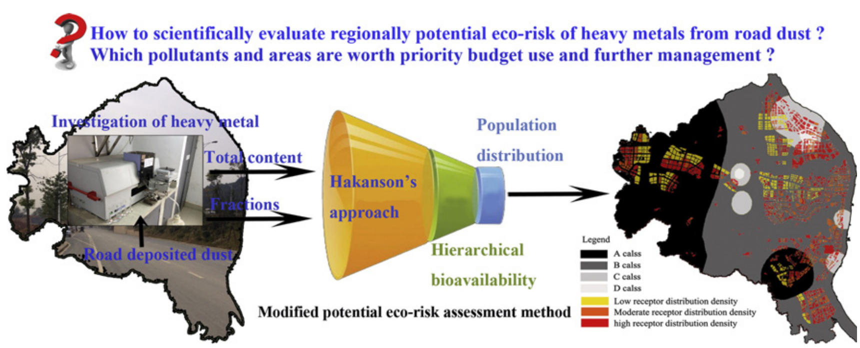
Jinhui Huang [a,b,*], Fei Li [a,b,c], Guangming Zeng [a,b,*], Wenchu Liu [a,b], Xiaolong Huang [a,b], Zhihua Xiao [a,b], Haipeng Wu [a,b], Yanling Gu [a,b], Xue Li [a,b], Xiaoxiao He [a,b], Yan He [a,b]

[a] College of Environmental Science and Engineering, Hunan University, Changsha 410082, China
[b] Key Laboratory of Environmental Biology and Pollution Control, Ministry of Education, Hunan University, Changsha 410082, China
[c] School of Information and Safety Engineering, Zhongnan University of Economics and Law, Wuhan 430073, China

HIGHLIGHTS

- Total content based assessment of dust metals may overestimate their eco-risks.
- Metal bioavailability was integrated into Hakanson's potential eco-risk index.
- Population distribution density map was made and incorporated by 3S technology.
- Cd was identified to be priority pollutant based on I_{geo}, E_r^i and bioavailability.
- Hierarchical risk map divided XDD into 4 area types under different priorities.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 11 October 2014
Received in revised form 22 September 2015
Accepted 27 September 2015
Available online 11 November 2015

Editor: Simon Pollard

Keywords:
Potential eco-risk assessment
Heavy metals
Road dust
Hierarchical bioavailability
Population distribution

ABSTRACT

Modified eco-risk assessment method (MEAM) integrated with the hierarchical bioavailability determined by the fraction detection of Cd, Pb, Zn, Cu, Cr in road dust samples and the local population distribution derived from the local land use map, was proposed to make the hierarchical eco-risk management strategy in Xiandao District (XDD), China. The geo-accumulation index (I_{geo}), the original potential eco-risk index (E_r^i) and the modified eco-risk assessment index (MEAI) were used to identify the priority pollutant. Compared with the Hunan soil background values, evaluated metal concentrations were found to different extent. The results of mean I_{geo}, E_r^i and bioavailability of studied metals revealed the following orders: Cd > Pb ≈ Zn > Cu ≈ Cr, Cd > Pb > Cu > Cr > Zn and Cd > Zn > Cu ≈ Pb > Cr, respectively. Therefore, Cd was regarded as the priority pollutant. To identify the priority areas taking into account cost consideration, the hierarchical risk map based on the results of the modified eco-risk assessment index with overlay of the population density map was needed and made. The west and partly south areas of XDD were under higher eco-risk generally. Moreover, the whole XDD area was divided into 4 area categories with different management priorities based on the possibility of occurrence of eco-risk, and the hierarchical risk management strategy associated with protecting local population was suggested to facilitate allocation of funds for risk management.

* Corresponding authors at: College of Environmental Science and Engineering, Hunan University, Changsha 410082, China.
E-mail addresses: huangjinhui_59@163.com (J. Huang), zgming@hnu.edu.cn (G. Zeng).

http://dx.doi.org/10.1016/j.scitotenv.2015.09.139
0048-9697/

1. Introduction

Industrial and economic activities are increasingly concentrated in urban areas, and cities have become the geographic focus of resource consumption and chemical emissions, which may cause a variety of problems including ecosystem degradation, public health risk, biodiversity decrease and so on (Lu et al., 2014; Tang et al., 2013). Road dust, which consists of soil, deposited airborne particulates, construction material, soot and fumes discharged from industry and vehicles, etc., is a sensitive indicator of urban ecosystem status than single compartmental monitoring of air, water and soil for it reflects pollutants from multi-media (H.M. Li et al., 2013; Z.G. Li et al., 2013; Lu et al., 2010). Moreover, road dust often contains high levels of heavy metals and organic contaminants such as polycyclic aromatic hydrocarbons (Jiang et al., 2014; Saeedi et al., 2012). Road dust contaminated by heavy metals has drawn particular attention due to their property of high toxicity, concealment, persistence and biological accumulation. Dust particles can migrate via saltation, creep (diameters > 100 μm), suspension (diameters < 100 μm), or can become incorporated in the urban aerosol (diameters < 10 μm) (De Miguel et al., 1999; Wang et al., 2006). Therefore, heavy metals associated with road dust may cause hazardous effects or potential risks to urban environment and endanger the ecosystem's health for a short or long time (Cook et al., 2005; H.M. Li et al., 2013; Z.G. Li et al., 2013; Tang et al., 2013).

In recent years, studies have increased in characterizing spatial pollution level of road dust metals, identifying the dominant anthropogenic sources of these metals, researching corresponding bioavailability by the fraction analysis of these metals, exploring the correlation among dust properties and these metal pollution features due to its important influence to urban ecological safety and human health (Acosta et al., 2014; Chen et al., 2014; H.M. Li et al., 2013; Z.G. Li et al., 2013; Liu et al., 2014; Lu et al., 2014). However, based on previous pollution assessment studies and the induced regional risk management strategies (H.M. Li et al., 2013; Z.G. Li et al., 2013; Tang et al., 2013), most studies are of less practical significance for: (i) possibly overestimating or underestimating the risk posed by heavy metals without considering of their specific chemical fractions and their binding state which are in close relationship with their bioavailability and mobility (Luo et al., 2012; Peijnenburg et al., 2007; Saleem et al., 2014). (ii) neglecting the principle of "no exposure, no risk" (Gay and Korre, 2006; Leeuwen and Vermeire, 2007) which may lead to meaningless management for dissevering the exposure risk with the local receptor distribution (since the source-pathway-receptor linkage is not intact) and (iii) not developing the elastic regional risk management strategies to distinguish different control areas with relatively different priorities which always makes decision making hard to determine the hierarchical strategies for efficient budget use.

The objectives of this study were: (i) to investigate spatial geo-accumulation degrees and hierarchical bioavailability of heavy metals in road dust of Xiandao District (XDD); (ii) to determine the possible population density map of XDD based on Landsat 7 remote sensing images; (iii) to develop a modified potential eco-risk assessment method (MEAM) integrated with heavy metals' bioavailability and possible population density for mapping the hierarchical risk map of XDD for use by decision makers.

2. Materials and methods

2.1. Study area

Xiandao District is a municipal district with a population of over 3 million which belongs to the capital of Hunan Province, Changsha city, Middle China. Changsha city is an important center of economy, culture, transportation, education and manufacturing. XDD belongs to subtropical monsoon climate and its annual average temperature is 17.2°°C. The urban average annual rainfall is 1361.6 mm. The total area of XDD is 1200 km^2 and the urban residents per capita disposable income reach 3449 dollars. From 2007 to 2013, the urbanization rate in Changsha city has increased from 56.5% to 70.6%. The local vehicular fleet has increased rapidly in recent years with an annual rate of over 17% and may reach 2 million vehicles in 2015. Outdoor air quality of Changsha city was not up to standard levels in 168 days out of the year in 2013. Rapid urbanization and economic development in XDD have resulted in a decline of environmental quality (Chen et al., 2011; Li et al., 2007; Yang et al., 2012).

2.2. Land use analysis

Present land use mapping was determined from a remote sensing image of the study area. Landsat 7 images acquired on January 15–23, 2014 were used in this work. The original images were successively processed by geometric correction (image to image), image fusion, radiometric calibration, atmosphere correction (Fast Line-of-sight Atmospheric Analysis of Hypercubes, FLAASH), and supervised classification using the remote sensing software ENVI 5.0 (ITT Visual Information Solutions Inc.). According to the land use map in 2011 and the land use planning map in 2013 of XDD by the Hunan government, the study area was divided into detailed land use types based on their remote sensing spectral characteristics (Fig. 1).

2.3. Samples collection, preparation and analytical methods

A total of 51 samples (3 parallel samples for each sampling site) were collected at the trunk road throughout XDD (Figs. 1 and S1). Samples at each sampling site were collected in October 2013 by gently sweeping an area of about 2 m^2 adjacent to the curb of the street with a clean plastic dustpan and brush, transferring about 300 g dust to a polyethylene bag for transport to the laboratory. In the laboratory, all samples were air-dried for 3 weeks, and then sieved through a 100 mesh nylon sieve to remove stones, dead organisms and coarse debris. As the road dust with diameters below 100 μm can be considered to mainly arise from atmospheric deposition and be transported by re-suspension (Charlesworth et al., 2011; Zhao and Li, 2013), therefore all the samples were sieved through 200 mesh nylon sieve (diameters below 73 μm) for analysis. For the total heavy metal content detection, 0.40 g samples were weighed on an electronic balance (Sartorius TE124S, Germany). Subsequently, the samples were placed in Teflon tubes and digested with HCl, HNO_3, HF, and $HClO_4$. Then the solutions were diluted with 2% (v/v) HNO_3 to a final volume of 50 ml, and analyzed for Cr, Cu, Zn, Pb, Cd by an atomic absorption spectrophotometer (AAnalyst700, Perkin-Elmer Inc., US). The metal fractions were studied using a sequential extraction scheme and the procedure proposed by Tessier et al. (1979) which is widely applied in various studies of heavy metal bioavailability (Acosta et al., 2014; Gao et al., 2015; Jiang et al., 2013) was used to study the partition of toxic metals. The selected Tessier method has been described in detail elsewhere (Shao et al., 2009).

Quality assurance and quality control were assessed using duplicates, method blanks, and State first-level standard materials (GBW GSS–5), with each batch of samples. The analysis results were reliable when repeat sample analysis error was below 5%, and the analytical precision for replicate samples was within ± 10%. The recovery of standard samples ranged from 95.7% to 103.6%. The results of five fractions were summed up and compared with total concentration to check the recovery percentage of the five-stage Tessier method. The obtained recovery percentages were Cr (85.32%), Cu (95%), Zn (93.5%), Pb (107.57%) and Cd (102.3%), respectively, indicating that the Tessier sequential extraction worked reasonably well. Throughout the experimental process, ultra-pure water was utilized for preparing the solutions, dilutions and blanks. All used chemical reagents was GR (guaranteed reagent). All the glassware and plastic vessels were treated with 10% (v/v) HNO_3 for at least 12 h and then washed with distilled and deionized

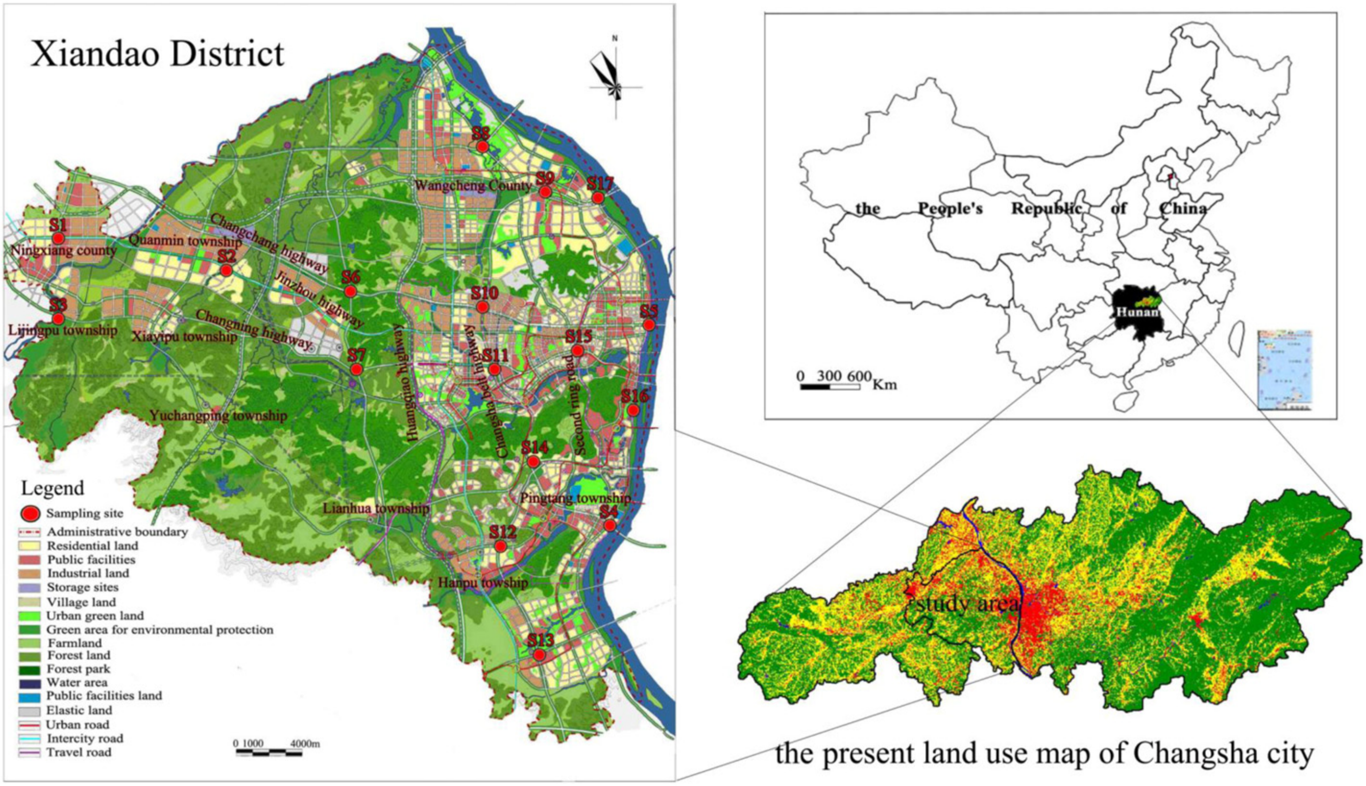


Fig. 1. Sampling sites in Xiandao District. The right images are maps of China and the present land use map of Changsha city respectively; the left image is a detailed land use map in which different functional zones and urban roads are labeled.

water respectively before use. To avoid potential cross-contamination of the samples, contact with metals was avoided during all procedures. The results met the accuracy demand of the Chinese Technical Specification for Soil Environmental Monitoring HJ/T 166-2004.

2.4. Pollution and bioavailability assessment methodology

2.4.1. Geo-accumulation index

The geo-accumulation index (I_{geo}) was originally defined by Muller (1969) and widely used to study level of metal enrichment in soils, sediments and dust (Tang et al., 2013; Zhang et al., 2012; Zhu et al., 2013). It is computed using the following equation:

$$I_{geo_i} = log_2[C_i/kB_i] \quad (1)$$

where C_i represents the measured concentration of the metal i in the samples. k, $k = 1.5$, is introduced to minimize the effect of possible variations in the background value or in this case reference, which may be attributed to anthropogenic influences or lithologic variations in the sediments. B_i is the geochemical background value or reference value of metal i in Hunan province (Pan and Yang, 1988; Zhou et al., 2008). I_{geo} is split into seven classes as follows: $I_{geo} \le 0$, class 0, uncontaminated (UC). $0 < I_{geo} \le 1$, class 1, uncontaminated to moderately contaminated (UMC). $1 < I_{geo} \le 2$, class 2, moderately contaminated (MC). $2 < I_{geo} \le 3$, class 3, moderately contaminated to heavily contaminated (MHC). $3 < I_{geo} \le 4$, class 4, heavily contaminated (HC). $4 < I_{geo} \le 5$, class 5, heavily to extremely contaminated (HEC). $5 < I_{geo}$, class 6 and extremely contaminated (EC).

2.4.2. Potential eco-risk index

To further study degree of eco-risk of dust pollution caused by heavy metals, the potential eco-risk index, which was developed based on sedimentary theory and abundance principle (Saeedi et al., 2014), was introduced. The potential eco-risk index was originally proposed by Hakanson (Hakanson, 1980) and widely used (Guo et al., 2010; F. Li et al., 2013; H.M. Li et al., 2013; Z.G. Li et al., 2013; Saeedi et al., 2012; Soltani et al., 2015). The potential eco-risk index states that the ecosystem risk is posed directly by heavy metals and further indicates the induced risk for corresponding local population indirectly. The original RI can be calculated by the following formulas:

$$RI = \sum_{i=1}^{m} E_r^i, E_r^i = T_r^i \times C_f^i, C_f^i = C_D^i/C_B^i \quad (2)$$

where RI is the sum of the potential risk of individual heavy metal, E_r^i is the potential risk of individual heavy metal, T_r^i is the toxic-response factor for a given heavy metal, C_f^i is the contamination factor, C_D^i is the present concentration of heavy metals in road dust, and C_B^i is the pre-industrial record of heavy metal concentration in road dust. Based on the Hakanson's approach, the toxic-response factors for Cr, Cu, Zn, Pb and Cd are 2, 5, 1, 5, and 30, respectively. In this study, the pre-industrial concentration records for Cr, Cu, Zn, Pb and Cd were replaced by their corresponding background values for Hunan province soil (Pan and Yang, 1988; Zhou et al., 2008). Hakanson defined five categories of E_r^i and four categories of RI, as shown in Table 1.

2.5. Modified potential eco-risk assessment method

It is widely recognized that the distribution, mobility, bioavailability and eco-toxicity of heavy metals in the environment depends not only on their total concentration but also on their fraction (Acosta et al., 2014; Zhao et al., 2009). Heavy metals with higher environmental bioavailability and mobility can migrate in different environmental media and be absorbed by the plant more easily. Afterwards they may enter the human food chain and thus pose a potential health risk to humans (Banerjee, 2003; He et al., 2005). Therefore, the modified potential

Table 1
Indices and corresponding degrees of potential eco-risk.

E_r^i value	Extent of eco-risk of single metal	*RI* value	Extent of potential eco-risk to the environmental
$E_r^i < 40$	Low risk	$RI < 150$	Low risk
$40 \le E_r^i < 80$	Moderate risk	$150 \le RI < 300$	Moderate risk
$80 \le E_r^i < 160$	Considerable risk	$300 \le RI < 600$	Considerable risk
$160 \le E_r^i < 320$	High risk	$RI \ge 600$	Very high risk
$E_r^i \ge 320$	Very high risk		

eco-risk assessment method (MEAM) was developed as the flow diagram in Fig. 2, namely: (i) developing the modified eco-risk assessment index (MEAI) and then mapping the corresponding interpolation maps of the hierarchical *RI*s; (ii) making the hierarchically potential eco-risk map based on the maps of the hierarchical *RI*s with overlay of the population density map (PDM).

Chen et al. (2008) established the relationship among heavy metal fractions, eco-toxicity, and bioavailability through the European Community Bureau of Reference sequential extraction (BCR). In this study, the fraction of heavy metals was measured using Tessier's method. Thus the hierarchical relationship among fractions of heavy metals, eco-toxicity, and bioavailability was modified (L. Li et al., 2012) and were calculated as follows:

$$C_{Bio} = C_i \times (f_{F1} + f_{F2}) \tag{3}$$

$$C_{Pbio} = C_i \times (f_{F3} + f_{F4}) \tag{4}$$

$$C_{Nbio} = C_i \times f_{F5} \tag{5}$$

where f_{F1}, f_{F2}, f_{F3}, f_{F4}, and f_{F5} are the percentages of the total metal concentration for exchangeable fraction (F1), carbonates associated fraction (F2), Fe–Mn oxide-associated fraction (F3), organic-associated fraction (F4), and residual fraction (F5). C_{Bio}, C_{Pbio} and C_{Nbio} are concentrations of the bioavailable, potentially bioavailable and non-bioavailable fractions in corresponding road dust samples, respectively. To integrate

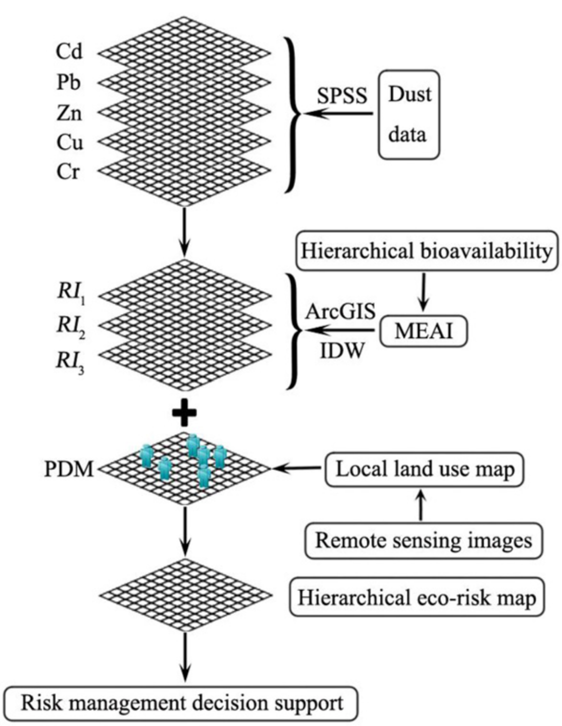


Fig. 2. Flowchart showing the modified potential eco-risk assessment method (MEAM) process for heavy metals.

bioavailability information into potential eco-risk assessment of heavy metals, the equations of the MEAI, based on Eqs. (2)–(5), were comprehensively defined as follows:

$$C_{f1}^i = C_{\text{Bio}}^i / C_B^i, C_{f2}^i = \left(C_{\text{Bio}}^i + C_{\text{Pbio}}^i\right)/C_B^i, C_{f3}^i = \left(C_{\text{Bio}}^i + C_{\text{Pbio}}^i + C_{\text{Nbio}}^i\right)/C_B^i \tag{6}$$

$$RI_m = \sum_{i=1}^{m} E_{rm}^i, E_{rm}^i = T_r^i \times C_{fm}^i \tag{7}$$

where C_{f1}^i, C_{f2}^i and C_{f3}^i are the hierarchical contamination factors for metal *i* based on their bioavailability analysis. E_{rm}^i is the hierarchically potential risk of heavy metal *i* at m (m = 1, 2 or 3) tier. RI_m is the sum of the potential risk of individual heavy metal hierarchically. Based on Eqs. (6)–(7), the three sets of modified potential eco-risk results were calculated and the calculated RI_1, RI_2 and RI_3 would be mapped by Inverse Distance Weighted interpolation (IDW), respectively.

To further identify the priority control area with the aim of protecting the safety of citizens, their possible exposure time and the exposure frequency were considered, and the PDM derived from the detailed land use types of XDD (Fig. 1) would be made based on the close relationship between the population distribution and the local land use pattern. Furthermore, the hierarchically potential eco-risk map would be obtained based on the spatial MEAI results combined with the PDM in order to make scientific reference for efficient regional risk identification and management.

2.6. Data mining and spatial analysis methods

To explore the spatial pollution characteristics of the studied metals in road dust samples, the software package ArcGIS 9.3 and SPSS version 16.0 for Windows were utilized. Under consideration of the study requirement and number of sampling sites, the spatial analysis method (including Inverse Distance Weighted interpolation (IDW), Local Polynomial interpolation (LP) and Radial Basis Functions interpolation (RBF)) and the geostatistical method (including Simple Kriging interpolation (SK)) were tested simultaneously to find optimal interpolation method with better parameter uncertainty control. IDW method was finally applied to map the spatial characteristics of regional pollutants. IDW employs a specific number of nearest points that are then weighted according to their distance from the point being interpolated. In this study, the power of 2 and the number of neighboring samples of 15 were chosen to clearly show both spatial variation and spatial patterns of the pollutants.

3. Results and discussion

3.1. Heavy metal enrichment in road dust

Statistical information about the original analysis results of three samples from each site and the total metal concentrations (Cu, Zn, Pb, Cd and Cr) in road dust in XDD are shown in Tables S1 and 2 respectively. It can be seen that the mean concentrations of all studied metals exceeded corresponding soil background values which indicated anthropogenic input. This was especially true for Cd and Zn which were on average 130.1 and 2.2 times their background values. Compared with the secondary class values of the Chinese Environmental Quality Standard for Soils, the mean concentrations of Cu, Zn, Pb and Cr were below their secondary standard values except for Cd was 15.2 times higher than its secondary standard value. Higher coefficients of variation (CV = 100SD/Mean) were found for Zn and Cd indicating their higher spatial variability which are probably due to human activities.

To study enrichment levels of heavy metals in road dust, the calculated results of I_{geo} are presented in Fig. 3. The order of pollution

Table 2
Descriptive statistics of metal concentrations in road dust from XDD mg/kg.

Elements	Cu	Zn	Pb	Cd	Cr
Mean	43.90	214.92	66.58	9.11	80.66
SD	21.69	179.68	30.17	6.28	47.86
CV(%)	49.41	83.60	45.31	68.94	59.34
BV_{Hunan}[a]	25	96	30	0.07	68
Class I	35	100	35	0.2	90
Class II[b]	100	300	250	0.6	250
Class III	400	500	500	1.0	300

Mean: arithmetic-mean; SD: standard deviation; CV: coefficient of variation.
[a] Soil background value of Hunan province, China (Pan and Yang, 1988; Zhou et al., 2008).
[b] Environmental quality standard secondary grade for soils, soil limitations to ensure agricultural production and human health (National Environmental Protection Agency of China, 1995).

levels based on their mean I_{geo} values was Cd (6.15) > Pb (0.40) > Zn (0.25) > Cu (−0.17) > Cr (−0.51), and mean I_{geo} values of Cu, Zn, Pb, Cd and Cr belonged to classes UC, UMC, UMC, EC and UC, respectively. Specifically it shows that 88.2% of the I_{geo} values for Cd belonged to EC, and the values of other sites belonged to HEC, which suggested that the road dust in XDD was significantly contaminated by Cd.

3.2. Original potential eco-risk assessment

To further study the original potential eco-risk of heavy metals in road dust, based on Eq. (2), the E_r^i and *RI* of the studied heavy metals were calculated and shown in Fig. 4. The mean E_r^i of Cu, Zn, Pb, Cd and Cr decrease in the order: Cd (3689.8) > Pb (10.76) > Cu (8.57) > Cr (2.35) > Zn (2.17), which is different from the result of I_{geo} to some extent. Spatially, E_r^i of Pb, Cu, Cr, and Zn at each sampling sites belongs to low risk, while E_r^i of Cd at each sampling site belongs to very high risk. Further, the calculated *RI* values of every sampling site were in the descending order: S2, S12, S1, S15, S13, S14, S3, S6, S11, S5, S10, S16, S7, S4, S9 and S8. Fig. 4 shows that the *RI* of every sampling site belonged to very high risk (*RI* > 600), and the E_r^i for Cd made average contribution of 93.9%. Cd was preliminarily identified to be the priority pollutant in road dust of XDD, and decision makers should take immediate measures to handle its levels.

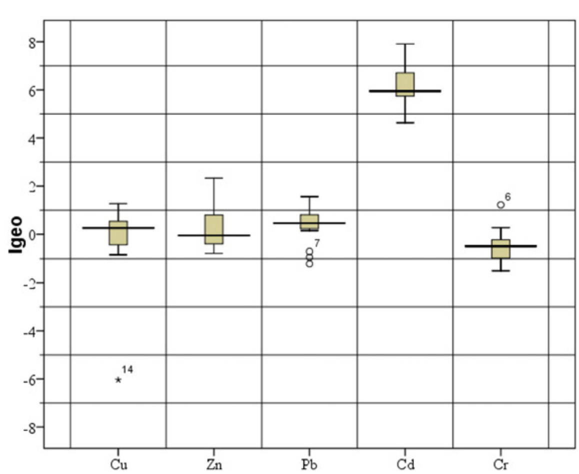


Fig. 3. Box-plots of the geo-accumulation index (I_{geo}) for studied metals in road dust from XDD.

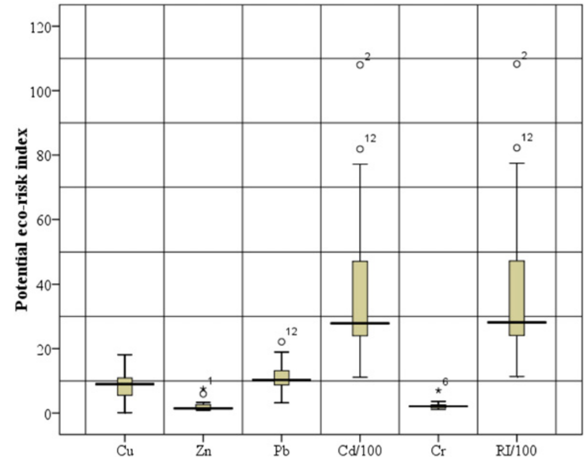


Fig. 4. Box-plots of the potential eco-risk index for metals for the studied metals in road dust of XDD.

3.3. Modified potential eco-risk method based on the hierarchical bioavailability and the local population distribution

Based on the total contents of the studied heavy metals and the commonly used evaluation methods, Cd was identified to be the priority pollutant. However, there is abundant evidence showing that total contents of heavy metals in road dust do not suffice for an evaluation of their mobility, which is critically dependent on their chemical fraction alongside with their bioavailability and toxicity (Acosta et al., 2014; Ma et al., 2016). With consideration of metals' bioavailability, may the result of the priority pollutant be changed? Where are priority control areas? Therefore, the developed MEAM (Fig. 2) was introduced to handle these questions.

Firstly, the percentages of heavy metals in different fractions in road dust from XDD are calculated and shown in Fig. 5 based on Eqs. (3)–(5). Results indicated that the percentage of Cd and Zn were higher in the bioavailable fraction with mean value of 18.29% and 10.38% respectively. Furthermore, the mean percentages of Cu, Pb and Cr in the bioavailable fraction were 7.87%, 8.50% and 9.71% respectively. Cu and Pb were found in the higher potentially bioavailable fraction with respective mean values of 56.49% and 43.73%. In addition, the mean percentage of Cd, Zn and Cr in the potentially bioavailable fraction were 37.16%, 37.47% and 31.46%, respectively. Spatially, for Pb the sites S2, S3, S8, S9, S13 and S14 had higher bioavailability. For Cd the sites S2, S3, S5, S8, S10, S11, S12, S14 and S17 had higher bioavailability (Fig. 5). In short, bioavailability of the studied metals were ranked in the order: Cd > Zn > Cu ≈ Pb > Cr. Spatially, sampling sites S2, S3, S5, S10 and S12 generally had higher bioavailability of all the studied metals. Therefore, combined with the previous results and Eq. (6), Cd was finally identified as the priority pollutant owing to its highest enrichment level and bioavailability than the remaining 4 metals.

Secondly, when to further identify priority control areas, some questions were posed like “with the calculated *RI* values, the whole XDD should be regarded as the control area (map of RI_3 in Fig. 6) which needs environmental management, but is it the optimal decision with the aim of protecting civil health and related eco-system ?” and “With limited human and material resources, how to utilize the budget efficiently with the different extents of management measures to the different priority classes of the areas”. Therefore, to make the original *RI* integrate with the information of metals' bioavailability and local population distribution is significant for producing a more realistic evaluation (Gay and Korre, 2006; Saleem et al., 2014). According to Fig. 2, the interpolation maps based on calculated hierarchical data of the MEAI (Fig. 5 and Eqs. (6)–(7)) were obtained as shown in Fig. 6 assisted by the IDW interpolation. Maps of RI_1, RI_2 and RI_3 as shown in Fig. 6 show the different bioavailabilities of metals divided the area of XDD into three different eco-risk distributions. Areas at moderate risk, considerable risk and every high risk account for around 3%, 41% and 56% of the area in RI_1 map. For RI_2 map, areas at moderate risk, considerable risk and every high risk account for around 1%, 3% and 94%. The total area was under very high eco-risk based on RI_3 (RI_3 equal to the original *RI*). Spatially, the west part of XDD was under higher eco-risk than the east part, and generally the north part was under lower eco-risk than the south part.

To further identify the priority control area with the aim of protecting safety of urban eco-system and citizens, the local population distribution derived from the detailed land use map of XDD (Fig. 1) and their possible exposure time, the exposure frequency were considered. Afterwards, the areas of XDD were preliminary divided into four parts including areas of high population density (Residential land, Public facilities land included), moderate population density (Village land and Urban green land included), low population density (Industrial land included) and the rest area with scarce exposure possibility. The processed PDM is shown in detail in Fig. 6. The PDM indicates areas at high population density, moderate population density and low population density account for around 20%, 10% and 15% of the XDD area. Further, there was an obvious spatial distribution that the west corner and east parts of XDD had higher population density than that of other areas.

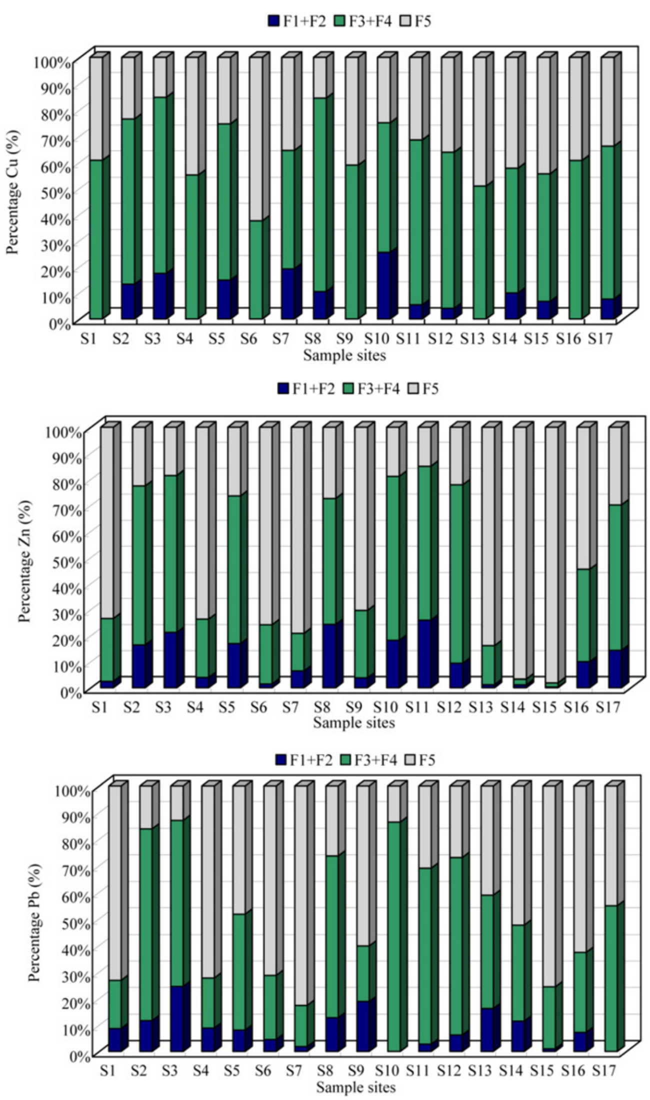


Fig. 5. Percents of heavy metals in bioavailable fraction, potentially bioavailable fraction and non-bioavailable fraction in road dust from XDD.

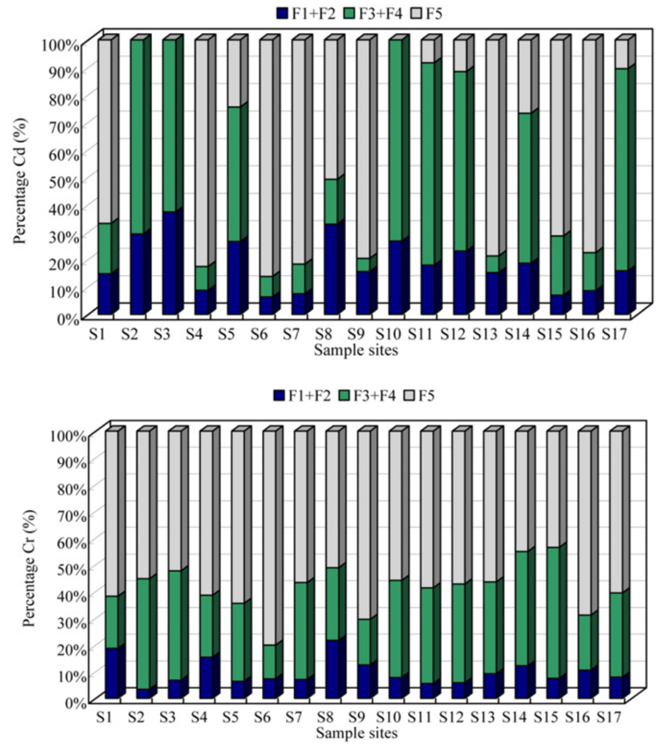


Fig. 5 (*continued*).

Finally, based on the processed maps of RI_1, RI_2 and RI_3, XDD was divided into A, B, C and D class areas with decreasing modified eco-risk index values (Table 1). The hierarchical eco-risk map for the studied metals based on the modified eco-risk assessment index with overlay of the PDM is shown in Fig. 6. Fig. 6 indicates that above 35% and 50% areas of XDD are in A and B class respectively. Generally, the west and partly south areas of XDD are under higher eco-risk. For local population safety and efficient budget utilization, the whole area of XDD can be divided into 4 area types with different management priorities based on the possibility of occurrence of a risk (Fig. 6). Namely, the first priority areas included the areas in A class with high or moderate population density and in B class with high population density. The secondary priority areas contained the areas in B class with moderate population density and in C class with high population density. The third priority areas included the areas in C class with moderate population density. At last the fourth priority areas contained the areas in A and B with low population density and D class with all kinds of population density. Furthermore, for different area types, risk management suggestions differ. For the first priority areas, preferentially environmental management and remediation was required and worth preferred budget support not only for its highest eco-risk and bioavailability of dust heavy metals but also its higher population density. As shown in Fig. 6, it was found that the secondary priority areas were in close relationship with the first priority areas geographically. Therefore, it was suggested to make a plan for 2 stages to be an extension of the management plan for the first priority areas, and the corresponding population protective measures and risk prevention propaganda were required as the transitional measures. At last, it was suggested that the third and fourth priority areas should take conservative measures temporarily for their relatively low eco-risk and population density, and perform regular monitoring for any new contamination changes. Fig. 6 also shows that the spatial modified eco-risk index for the studied metals was in close relationship with the industrial land use of XDD, and the farm land and forest land were under higher eco-risk. Therefore, the very high eco-risk of XDD mainly caused by Cd is probably derived from local industry type and industry distribution, and the local food safety is of concern.

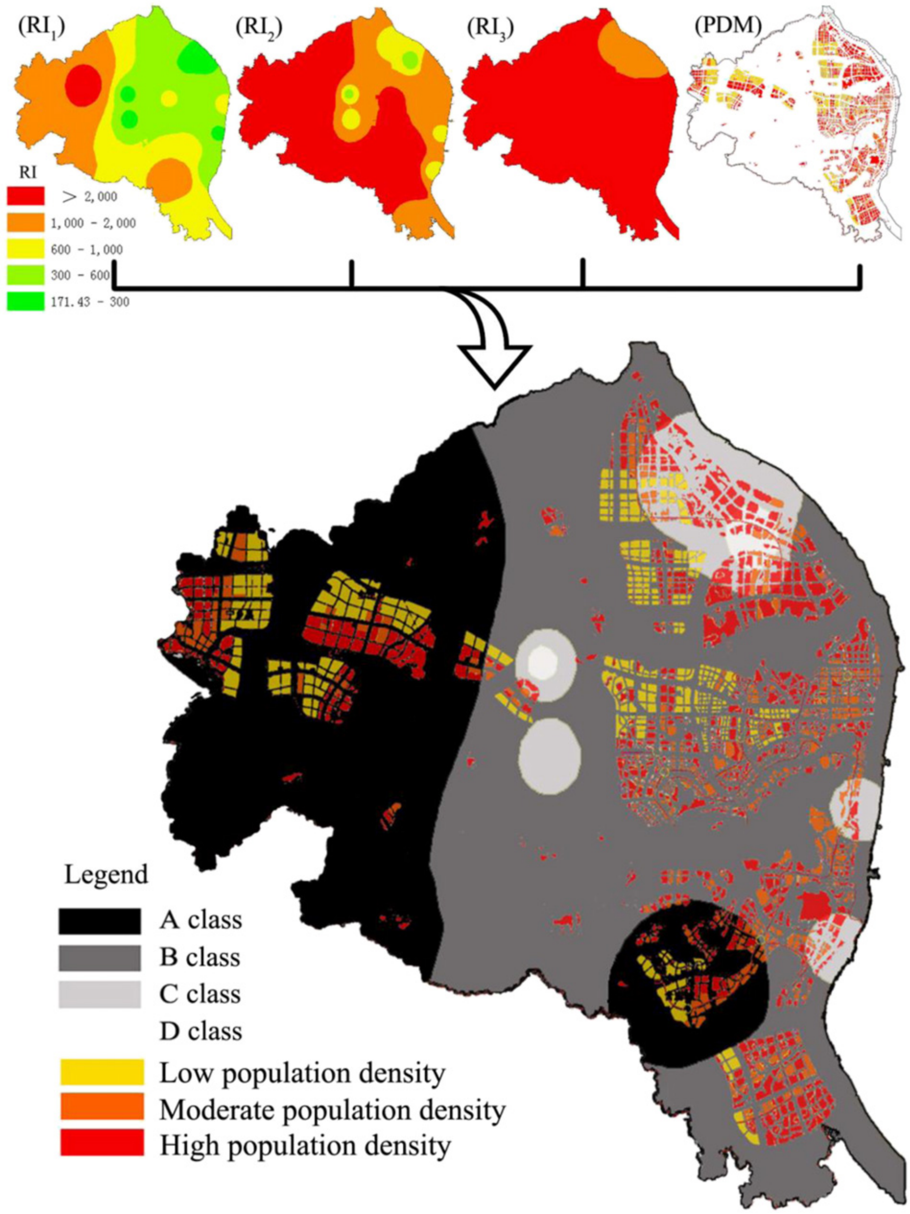


Fig. 6. Hierarchical eco-risk map for heavy metals using modified potential eco-risk assessment with overlay of the processed population density map.

The evaluation of uncertainty is an important step accompanying risk assessment. In the present study, it is recognized that the developed methodology has its limitations and is at the screening level. It is clear, for example, that the assumption that population never moves and is at same risk tolerance. The dust properties (pH and Dust Organic Matter) variance and the climate variance are likely to influence the total concentration and bioavailability of heavy metals. Therefore further research concerning the correlation among these factors is in progress in order to establish relevant regression prediction equation. The seasonal contamination changes of the studied metals were not considered and heavy metals pollution in one environment medium was considered. Some sources of uncertainty are well emphasized in literature (F. Li et al., 2012). However, boundaries for any study must be set, and seen in the light of the original eco-risk assessment method, the developed MEAM does provide an improvement and a useful tool, especially for making efficient environment research and management policy in condition of limited human and material resources.

4. Conclusions

The concentrations of Cu, Zn, Pb, Cd and Cr in road dust in XDD were higher than their local soil background values to different extent, indicating an anthropogenic input. Mean concentration of Cd was 15.2 times higher than its secondary standard value of the Chinese Environmental Quality Standard for Soils. The ranking of mean I_{geo} of the studied metals follows the order: Cd (EC) > Pb (UMC) ≈ Zn (UMC) > Cu (UC) ≈ Cr (UC). The mean E_r^i showed in decreasing order: Cd > Pb > Cu > Cr > Zn. Bioavailability of the studied metals were ranked in order: Cd > Zn > Cu ≈ Pb > Cr. Therefore, Cd was identified as the priority pollutant based on MEAI.

To further identify the hierarchical priority areas of XDD, the modified potential eco-risk assessment method (MEAM) was developed and performed. Based on the hierarchical eco-risk map, XDD was divided into 4 area categories (A, B, C and D) with the different management priorities based on the possibility of occurrence of a risk, and the hierarchical risk management strategy associated with local population distribution was recommended for maximizing social and environmental benefits from limited administrative resources. Overall, MEAM proved to be an improvement on previous methods in as so far as it spatially incorporated the hierarchical bioavailability and the local population density for providing a more realistic level of conservatism which thus enhanced the accuracy of predicting risk to receptors of concern, and supplying decision-makers with the valuable references to efficiently formulate or improve the regional risk management policy, the regional environmental research plan and land use planning.

Conflict of interest

The authors declare no conflict of interest.

Acknowledgments

This study was financially supported by the National Natural Science Foundation of China (51178172, 51039001, 51308076 and 51378190), the Project of Chinese Ministry of Education (113049A) and the Research Fund for the Program for Changjiang Scholars and Innovative Research Team in University (IRT-13R17).

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2015.09.139.

References

Acosta, J.A., Faz, A., Kalbitz, K., Jansen, B., Martínez-Martínez, S., 2014. Partitioning of heavy metals over different chemical fraction in road dust of Murcia (Spain) as a basis for risk assessment. J. Geochem. Explor. 144, 298–305.
Banerjee, A.D.K., 2003. Heavy metal levels and solid phase speciation in road dusts of Delhi, India. Environ. Pollut. 123, 95–105.
Charlesworth, S., De Miguel, E., Ordóñez, A., 2011. A review of the distribution of particulate trace elements in urban terrestrial environments and its application to considerations of risk. Environ. Geochem. Health 33, 103–123.
Chen, M., Li, X.M., Yang, Q., Zeng, G.M., Zhang, Y., Liao, D.X., et al., 2008. Total concentrations and speciation of heavy metals in municipal sludge from Changsha, Zhuzhou and Xiangtan in middle-south region of China. J. Hazard. Mater. 160, 324–329.
Chen, J.Q., Wang, Z.X., Wu, X., Zhu, J.J., Zhou, W.B., 2011. Source and hazard identification of heavy metals in soils of Changsha based on TIN model and direct exposure method. Trans. Nonferrous Metals Soc. China 21, 642–651.
Chen, H., Lu, X.W., Li, L.Y., 2014. Spatial distribution and risk assessment of metals in dust based on samples from nursery and primary schools of Xi'an, China. Atmos. Environ. 88, 172–182.
Cook, A.G., Weinstein, P., Centeno, J.A., 2005. Health effects of natural dust: role of trace elements and compounds. Biol. Trace Elem. Res. 103, 1–15.
De Miguel, E., Llamas, J.F., Chacón, E., Mazadiego, L.F., 1999. Sources and pathways of trace elements in urban environments: a multi-elemental qualitative approach. Sci. Total Environ. 235, 355–357.
Gao, P., Liu, S., Ye, W.Y., Lin, N., Meng, P., Feng, Y.J., et al., 2015. Assessment on the occupational exposure of urban public bus drivers to bioaccessible trace metals through resuspended fraction of settled bus dust. Sci. Total Environ. 508, 37–45.
Gay, J.R., Korre, A., 2006. A spatially-evaluate methodolody for assessing risk to a population from contaminated land. Environ. Pollut. 142, 227–234.
Guo, W.H., Liu, X.B., Liu, Z.G., Li, G.F., 2010. Pollution and potential ecological risk evaluation of heavy metals in the sediments around Dongjiang harbor, Tianjing. Procedia Environ. Sci. 2, 729–736.
Hakanson, L., 1980. An ecological risk index for aquatic pollution control. A sedimentological approach. Water Res. 14, 975–1001.
He, Z.L., Yang, X.E., Stoffella, P.J., 2005. Trace elements in agroecosystems and impacts on the environment. J. Trace Elem. Med. Biol. 19, 125–140.
Jiang, M., Zeng, G.M., Zhang, C., Ma, X.Y., Chen, M., Zhang, J.C., et al., 2013. Assessment of heavy metal contamination in the surrounding soils and surface sediments in Xiawangang river, Qingshuitang District. PLoS One 8, e71176.
Jiang, Y.F., Hu, X.F., Yves, U.J., Zhan, H.Y., Wu, Y.Q., 2014. Status, source and health risk assessment of polycyclic aromatic hydrocarbons in road dust of an industrial city, NW China. Ecotoxicol. Environ. Saf. 106, 11–18.
Leeuwen, C.J.V., Vermeire, T.G., 2007. Risk assessment of chemicals: an introduction. 2nd ed. Springer Press, Netherlands.
Li, Z.W., Zeng, G.M., Zhang, H., Yang, B., Jiao, S., 2007. The integrated eco-environment assessment of the red soil hilly region based on GIS—a case study in Changsha city, China. Ecol. Model. 202, 540–546.
Li, F., Huang, J.H., Zeng, G.M., Yuan, X.Z., Liang, J., Wang, X.Y., 2012a. Multimedia health risk assessment: a case study of scenario-uncertainty. J. Cent. South Univ. 19, 2901–2909.
Li, L., Xu, Z.R., Zhang, C.L., Bao, J.P., Dai, X.X., 2012b. Quantitative evaluation of heavy metals in solid residues from sub- and super-critical water gasification of sewage sludge. Bioresour. Technol. 121, 169–175.
Li, F., Huang, J.H., Zeng, G.M., Yuan, X.Z., Li, X.D., Liang, J., et al., 2013a. Spatial risk assessment and sources identification of heavy metals in surface sediments from the Dongting lake, middle China. J. Geochem. Explor. 132, 75–83.
Li, H.M., Qian, X., Hu, W., Wang, Y.L., Gao, H.L., 2013b. Chemical speciation and human health risk of trace metals in urban road dusts from a metropolitan city, Nanjing, SE China. Sci. Total Environ. 456–457, 212–221.
Li, Z.G., Feng, X.B., Li, G.H., Bi, X.Y., Zhu, J.M., Qin, H.B., et al., 2013c. Distributions, sources and pollution status of 17 trace metal/metalloids in the road dust of a heavily industrialized city of central China. Environ. Pollut. 182, 408–416.
Liu, E.F., Yan, T., Birch, G., Zhu, Y.X., 2014. Pollution and health risk of potentially toxic metals in urban road dust in Nanjing, a mega-city of China. Sci. Total Environ. 476–477, 522–531.
Lu, X.W., Wang, L.J., Li, L.Y., Lei, K., Huang, L., Kang, D., 2010. Multivariate statistical analysis of heavy metals in road dust of Baoji, NW China. J. Hazard. Mater. 173, 744–749.
Lu, X.W., Wu, X., Wang, Y.W., Chen, H., Gao, P.P., Fu, Y., 2014. Risk assessment of toxic metals in road dust from a medium-sized industrial city of China. Ecotoxicol. Environ. Saf. 106, 154–163.
Luo, X.S., Ding, J., Xu, B., Wang, Y.J., Li, H.B., Yu, S., 2012. Incorporating bioaccessibility into human health risk assessments of heavy metals in urban park soils. Sci. Total Environ. 424, 88–96.
Ma, X.L., Zuo, H., Tian, M.J., Zhang, L.Y., Meng, J., Zhou, X.N., et al., 2016. Assessment of heavy metals contamination in sediments from three adjacent regions of the Yellow river using metal chemical fractions and multivariate analysis techniques. Chemosphere 144, 264–272.
Muller, G., 1969. Index of geoaccumulation in sediments of the Rhine river. GeoJournal 2, 108–118.
National Environmental Protection Agency of China, 1995. Environmental Quality Standard for Soils (GB 15618–1995) (in Chinese).
Pan, Y.M., Yang, G.Z., 1988. Hunan soil background values and research methods. Chinese Environmental Science Press, Beijing, pp. 275–285 (in Chinese).
Peijnenburg, W.J., Zablotskaja, M., Vijver, M.G., 2007. Monitoring metals in terrestrial environments within a bioavailability framework and a focus on soil extraction. Ecotoxicol. Environ. Saf. 67, 163–179.
Saeedi, M., Li, L.Y., Salmanzadeh, M., 2012. Heavy metals and polycyclic aromatic hydrocarbons: pollution and ecological risk assessment in road dust of Tehran. J. Hazard. Mater. 227–228, 9–17.
Saeedi, M., Salmanzadeh, M., Li, L.Y., 2014. Response to the comments of Zhang et al. (2014) on "heavy metals and polycyclic aromatic hydrocarbons: pollution and ecological risk assessment in street dust of Tehran". J. Hazard. Mater. 279, 389–391.
Saleem, M., Iqbal, J., Shah, M.H., 2014. Non-carcinogenic and carcinogenic health risk assessment of selected metals in soil around a natural water reservoir, Pakistan. Ecotoxicol. Environ. Saf. 108, 42–51.
Shao, M.F., Zhang, T., Fang, H.H.P., 2009. Autotrophic denitrification and its effect on metal speciation during marine sediment remediation. Water Res. 43, 2961–2968.
Soltani, N., Keshavarzi, B., Moore, F., Tavakol, T., Lahijanzadeh, A.R., Jaafarzadeh, N., et al., 2015. Ecological and human health hazards of heavy metals and polycyclic aromatic hydrocarbons (PAHs) in road dust of Isfahan metropolis, Iran. Sci. Total Environ. 505, 712–723.
Tang, R.L., Ma, K.M., Zhang, Y.X., Mao, Q.Z., 2013. The spatial characteristics and pollution levels of metals in urban road dust of Beijing, China. Appl. Geochem. 35, 88–98.
Tessier, A., Campbell, P.G.C., Bisson, M., 1979. Sequential extraction procedure for the speciation of particulate trace metals. Anal. Chem. 51, 844–851.
Wang, X.S., Qin, Y., Chen, Y.K., 2006. Heavy meals in urban roadside soils, part 1: effect of particle size fractions on heavy metals partitioning. Environ. Geol. 50, 1061–1066.
Yang, B., Chen, Z.L., Zhang, C.S., Dong, J.H., Peng, X.C., 2012. Distribution patterns and major sources of dioxins in soils of the Changsha-Zhuzhou-Xiangtan urban agglomeration, China. Ecotoxicol. Environ. Saf. 84, 63–69.
Zhang, Y., Hu, X.N., Yu, T., 2012. Distribution and risk assessment of metals in sediments from Taihu lake, China using multivariate statistics and multiple tools. Bull. Environ. Contam. Toxicol. 89, 1009–1015.
Zhao, X.M., Dong, D.M., Hua, X.Y., Dong, S.F., 2009. Investigation of the transport and fate of Pb, Cd, Cr(VI) and As(V) in soil zones derived from moderately contaminated farmland in northeast, China. J. Hazard. Mater. 170, 570–577.
Zhao, H.T., Li, X.Y., 2013. Understanding the relationship between heavy metals in road-deposited sediments and washoff particles in urban stormwater using simulated rainfall. J. Hazard. Mater. 246–247, 267–276.
Zhou, T., Xi, C.Z., Dai, T.G., Huang, D.Y., 2008. Comprehensive assessment of urban geological environment in Changsha city. Guangdong Trace Elem. Sci. 15, 32–38 (in Chinese).
Zhu, Z.M., Sun, G.Y., Bi, X.Y., Li, Z.G., Yu, G.H., 2013. Identification of trace metal pollution in urban dust from kindergartens using magnetic, geochemical and lead isotopic analyses. Atmos. Environ. 77, 9–15.