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HEAVY METALS IN THE ENVIRONMENT

Heavy Metal Contamination and Assessment of Roadside and Foliar Dust along the Outer-Ring Highway of Shanghai, China

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Foliar and roadside dust samples were collected from five sites along the outer-ring highway of Shanghai, one of the biggest metropolitan areas of China, to assess heavy/toxic metal contamination. Concentrations of Zn, Cu, Ni, As, and Hg in foliar dust were higher than in roadside dust, whereas concentrations of Pb and Cd were higher in roadside dust. In the roadside dust, average concentrations of all metals except As in foliar and roadside dust samples were significantly above the background values of soil in Shanghai: the ratios between the average of samples and background values of Shanghai were in the order: Cd (25.1) > Zn (12.2) > Cu (6.16) > Pb (5.74) > Ni (5.50) > Hg (5.18) > As (1.05). By using the geo-accumulation index, the pollution grades of seven heavy metals at five sampling sites were calculated. Roadside dust was heavily to extremely contaminated with Cd; moderately to heavily contaminated with Zn; and moderately contaminated with Cu, Hg, Pb, and Ni. Foliar dust was heavily contaminated with Cd; moderately to heavily contaminated with Zn and Cu; and moderately contaminated with Hg, Pb, and Ni. The contamination level of heavy metals in the Puxi area was greater than that in the Pudong area, which might be related to the industrial distribution and land use. Combined with correlation analysis, hierarchical cluster analysis indicated that atmospheric deposition is the main source of Cd, Hg, As, and Pb in dust and that Cu and Zn in dust are mainly from heavy traffic on the highway. A portion of Ni in dust also comes from the parent soil.

Urban dust, the main contributor to urban pollution, is the carrier of different kinds of environmental pollutants (Inyang and Bae, 2006). Urban dust is a multicomponent aggregator that is composed of mineral constituents, natural biogenic materials, and anthropogenic inorganic/organic matter (Ferreira-Baptista and De Miguel, 2005; Shi et al., 2008a). Damage to the ecosystem caused by dust is hidden and long-term, whereas damage to the human body can be direct and immediate (Inyang and Bae, 2006). Due to the small size and strong transmissibility of dust examined in this study (ranging in size from 1 to 10,000 μm, with a large percentage having an aerodynamic diameter of 10 μm or less), it is hazardous to human health and to the environment (Inyang and Bae, 2006). It could easily and rapidly spread via wind or flushing of road runoff and then inhaled by humans during normal breathing (Shi et al., 2011). Dust particles could also be absorbed by the human body and accumulate to toxic levels.

Heavy metal pollution in roadside/street dust has caused concerns in the past years throughout the world (e.g., Banerjee, 2003; Ract et al., 2003; Ahmed and Ishiga, 2006; Kartal et al., 2006; Oliva and Espinosa, 2007; Salonen and Korkka-Niemi, 2007; Sindern et al., 2007; Srinivasa Gowd et al., 2010) and in China (Li et al., 2001; Li and Huang, 2007; Tanner et al., 2008; Shi et al., 2008a; Lu et al., 2009a; Lu et al., 2009b; Shi et al., 2010; Sun et al., 2010; Shi et al., 2011; Zhang et al., 2013). The main sources of the metals are traffic emissions, industrial exhaust, and atmospheric deposition. The forms of emission include point (e.g., industries and power plants), line (e.g., traffic), and surface (e.g., residential heating). Such pollution exists everywhere and poses potential threats to a person's daily life because heavy metals cannot be degraded or decomposed in the environment and can accumulate in the human body (Banerjee, 2003; Imperato et al., 2003).

To our knowledge, most previous research about heavy metal pollution in dust was performed near busy roads. These studies focused on the comparative analysis of the concentrations between roadside dust and soil dust rather than the dust on foliar surfaces or quantifying the heavy metal concentrations on

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Abbreviations: Igeo, geo-accumulation index; pcu, passenger car unit; TOC, total organic carbon.

roadside dust of urban areas compared with those of suburban areas (Shi et al., 2011; Zhang et al., 2013). Heavy metal content decreases with dust size because smaller dust particles have greater surface area and absorb more heavy metal elements. Foliar dust is of greater concern because the frequent hand-to-mouth activities of children increase their exposure (Shi et al., 2011), and the dust is deposited on roadside shrubs that are of the same height as some children (0.5–1.5 m), which could be an important contributor of dust intake by children through direct inhalation. In this study, we investigated heavy metal content in dust samples selected from roadside and foliage along the outer-ring highway of Shanghai (i) to quantify heavy metal accumulation in roadside and foliar dust and to assess heavy metal pollution; (ii) to compare the difference of heavy metal contents among urban roadside dust, suburban roadside dust, foliar dust, and soils in different cities; and (iii) to analyze the dust pollution spatial patterns in Shanghai and identify the main sources of metals in the dust.

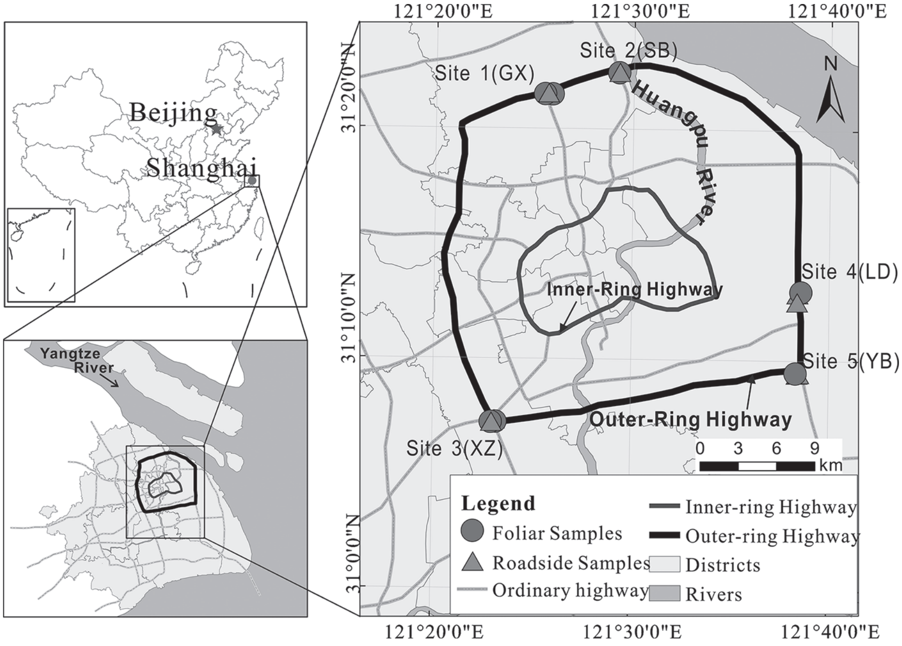

Fig. 1. Sampling sites along the outer-ring highway. GX, Gongfu Xincun; LD, Long Dong Avenue Interchange; SB, Songbin Road; XZ, Xinzhuang Interchange; YB, Ying Bin Avenue Interchange.

Materials and Methods

Location Description

Shanghai (30°40′–31°53′ N, 120°51′–122°12′ E) is one of the largest cities in China, with an area of 6340.5 km^2. Shanghai is located in the Yangtze River Delta of eastern China (Chan, 2007) and had a population of >23 million in 2010 (National Bureau of Statistics of China, 2010). The Huangpu River divides Shanghai into two parts: the Pudong area and the Puxi area. Adjacent to the Pacific Ocean, it belongs to the subtropical monsoon climate, with an annual rainfall of 1122 mm and an annual average temperature of 16.1°C (Shi et al., 2008a). The main framework of the urban green space system is developed along the main roads and river. The Shanghai outer-ring highway is a ring-shaped expressway that surrounds the inner districts of Shanghai, with a total length of 99 km. Its original designed capacity was 60,000 passenger car unit per day (pcu d^{-1}), but the current traffic flow has exceeded the design capacity. For example, in the Pudong New District, the average daily traffic concentration from January to June in 2007 was about 94,891 pcu d^{-1} on the Pudong section of the outer-ring highway, more than 1.5 times the highway's design capacity. Moreover, because of the main function of the outer-ring as the cargo route in and out of Shanghai, trucks account for more than 50% of all vehicles (Tao, 2011). These trucks, especially the heavy trucks, consume diesel instead of gasoline and therefore emit more toxic material when idling or running at low speeds (Riddle et al., 2007). There are also more pollutants emitted from truck tires and brakes (Adachi and Tainosho, 2004).

Five sampling sites were selected at the intersection of the local road and the outer-ring highway (Fig. 1). Site 1, Gongfu Xincun (GX) (31°21′30.54″ N, 121°25′53.55″ E), is located at the intersection of the North and South Highway and the outer-ring highway. Site 2, Song Bin Road (SB) (31°22′28.01″ N, 121°29′15.43″ E), is located at the intersection of the Song Bin road and the outer-ring highway. These intersections are the main connection between the city center and the Baoshan District, which is an important heavy industrial base. There is heavy traffic, especially truck traffic, at these two intersections. Site 3, the XinZhuang (XZ) Interchange (31°07′21.61″ N, 121°23′07.10″ E), is one of the biggest interchanges in Shanghai and is the connection between an important highway from Shanghai to Zhejiang Province and the outer-ring highway. Around Site 3, there are numerous overpasses with a large number of vehicles, which results in a heavy traffic jam every day. Site 4, the Long Dong (LD) Avenue Interchange (31°12′55.89″ N, 121°38′23.59″ E) and Site 5, the Ying Bin (YB) Avenue Interchange (31°09′26.56″ N, 121°38′24.45″ E) connect the outer-ring highway and the highway to the Pudong international airport. Site 1 (GX), Site 2 (SB), and Site 3 (XZ) are located in the Puxi area, which has been fully developed for more than 100 yr (Shi et al., 2008a). Sites GX and SB are near the heavy industrial base of Shanghai, and XZ is located in an area with industrial parks and factories. Site 4 (LD) and Site 5 (YB) are in the Pudong area, which developed slowly until 1990 and is mainly a suburban area, except for a financial center in its core area (Fig. 1).

Sampling Methods

Samples were taken after several sunny days, so the dust could deposit and accumulate on the road and foliar surfaces. Roadside dust and foliar dust were sampled with print papers and small brushes. Roadside dust was collected from the road surface directly around the sampling site in a 5- to 20-m area, and foliar dust was collected from the leaf surface of the shrubs along the road (e.g., *Fatsia japonica* and *Aucuba japonica* 'Variegata'), with leaf height ranging from 0.5 to 1.5 m. The weight of the dust samples from roadside was about 20 to 100 g, and the weight of the dust samples from leaves was about 5 to 10 g. All the collected samples were stored in polyethylene bags at −20°C until being transported to the laboratory for analysis. A total of 10 foliar and 8 roadside dust samples were collected from the five sampling sites (Table 1).

Analysis

Samples were freeze-dried (ALPHA 1–4/LD, Martin Christ Inc.) and mixed thoroughly using mortars and pestles. A small portion of each sample (about 0.5 g) was passed through a 63-μm sieve, weighed, and placed in a polytetrafluoroethylene vessel. The samples were hot digested with concentrated $HClO_4$, HF, and HNO_3 (Deng et al., 2010). The digested solutions were each diluted to 50 mL with 1:1 (v/v) HNO_3 and stored in plastic bottles. The concentrations of Cd, Cu, Ni, Pb, and Zn were determined by a direct-reading inductively coupled plasma emission spectrometer analyzer (710-ES, Varian). Another 0.5-g sample was passed through a 63-μm sieve and digested for 2 h with aqua regia (HNO_3:HCl, 3:1) in a warm water bath (95°C). After cooling, part of the digestion solution was used for As concentration measurement directly, and another part was used for Hg measurement after being deoxidated by a reducing agent (thiocarbamide and ascorbic acid). Concentrations of Hg and As were determined by an atomic fluorescence spectrophotometer (AFS-9230, Jitian Inc.). Total organic carbon (TOC) was determined with potassium dichromate oxidation–outer heating (Shi et al., 2008b). Blanks and sediment standard reference material (GSD-9, National Research Center for Certified Reference Materials, Beijing, China) were performed synchronously in each analytical batch. The recoveries were between 86 and 109%.

Contamination Assessment Methodology

Pollution levels of heavy/toxic metals in dust samples can be characterized by the geo-accumulation index (Igeo) put forward by Müller (1969). This contamination assessment index has been used in many environmental studies (e.g., Lu et al., 2009a; Shi et al., 2010) and can be defined with the following equation:

$$\text{Igeo} = \log 2[Cn/(k \times Bn)] \quad [1]$$

where *C*n is the measured concentration of the metal n, and *B*n is the background value of the metal n. Because of the possible variations of the background data due to lithological variations, the factor 1.5 is used for compensating the background data (correction factor) due to lithogenic effects in Igeo calculating (Singh et al., 1997; Howari and Banat, 2001; Loska et al., 2004; Ghrefat et al., 2011). Müller (1981) proposed seven classes of the geo-accumulation index as follows: if Igeo ≤ 0, uncontaminated (class 0); if 0 < Igeo ≤ 1, uncontaminated to moderately contaminated (class 1); if 1 < Igeo ≤ 2, moderately contaminated (class 2); if 2 < Igeo ≤ 3, moderately to heavily contaminated (class 3); if 3 < Igeo ≤ 4, heavily contaminated (class 4); if 4 < Igeo ≤ 5, heavily contaminated to extremely contaminated (class 5); if Igeo > 5, extremely contaminated (class 6).

Table 1. Distribution and number of samples.

Sampling site	Number of samples	
	Foliar dust	Roadside dust
Gongfu Xincun (GX)	3	2
Songbin Road (SB)	3	1
Xinzhuang Interchange (XZ)	2	1
Long Dong Avenue Interchange (LD)	1	2
Ying Bin Avenue Interchange (YB)	1	2
Total	10	8

Results

Total Organic Carbon Content in Foliar and Roadside Dust

The average TOC content in foliar dust (6.86 mg g^{-1}) was 1.84 times higher than that in roadside dust (3.72 mg g^{-1}), and the largest value of TOC in foliar dust (11.3 mg g^{-1}) was almost two times higher than that in roadside dust (Table 2). Although we did not measure the particle size of the dust sample because of the sample shortage, TOC in foliar dust is significantly greater than that in roadside dust (independent sample *t* test; $p < 0.01$), implying that foliar dust is finer than roadside dust (Sternberg et al., 2010).

Heavy Metal Contents in Foliar Dust and Roadside Dust

Total contents of the seven observed metals are given in Table 2, and soil background values and environmental quality standard for soils were used as reference values. The average contents of Cu, Zn, Hg, and As in foliar dust were higher than those in roadside dust, and the average contents of Ni, Pb, and Cd were higher in roadside dust. The larger standard deviations of Zn, Pb, and Ni imply the great disturbance and noticeable inputs from human activities (Dantu, 2009). The ratios between foliar and roadside dust values decreased in the order: Hg (1.35) > Cu (1.20) > As (1.15) > Zn (1.10) > Cd (0.96) > Ni (0.94) > Pb (0.88). All of the mean values in the foliar and roadside dust samples, except As in roadside dust, were significantly higher than the background values of Shanghai soil, especially Cd (25.05 times) and Zn (12.22 times). The percentages of samples with Cd, Cu, Ni, Pb, Zn, Hg, and As concentrations exceeding the corresponding values in class 2 were 94.4, 94.4, 77.8, 5.56, 94.4, 5.56, and 0% of the total samples, respectively. These elements may pose unfavorable effects to agricultural production and human health. Furthermore, metal contents in measured urban dusts were above the numbers in class 3, which clearly showed that almost all values of Cd (94.4%) and Zn (94.4%) and nearly half of Ni values (44.44%) surpassed the corresponding limits for the normal growth of plants (i.e., class 3).

Spatial Distribution of Heavy Metal Concentrations

The spatial distribution of metal levels in foliar and roadside dust along the outer-ring highway is shown in Fig. 2. Each element shows different distribution patterns on foliar and roadside dust. On the whole, the contents of heavy metals in foliar dust were higher than those in roadside dust, especially for samples observed in the Puxi area (GX, SB, and XZ), whereas the level of Pb in roadside dust in XZ was over three times higher than its level in foliar dust. In cities, Pb and Zn could indicate the level of traffic pollution for their specific source (Li et al., 2001); the XZ area has a complex highway interchange with large amounts of traffic every day. Therefore, there was a high Pb value in the roadside dust from XZ. A similar condition was found with the Cd level at the SB site, which was largely associated with tire tread and lubricating oil (Ellis and Revitt, 1982); tire wear and oil spills directly contributed to high Cd accumulation in dust. Moreover, the dust with larger particle size discharged from vehicle exhaust and tire wear tends to deposit and accumulate more along the roadside rather than on foliage, which explains why almost all sampling stations had higher Cd concentrations in roadside dust than in foliar dust. However, Cu, Zn, As, and Hg content in foliar dust higher was higher than in roadside dust at more than half of the sampling sites. In general, the highest metal concentrations in foliar or roadside dust was on the west side of the Huangpu River (i.e., the Puxi area [sites GX, SB, and XZ]) and lowest on the east side (i.e., the Pudong area [sites LD and YB]). The higher Hg concentration in the Puxi area (GX, SB, and XZ) might be related to emissions (e.g., from coal combustion) from heavy industrial factories in the GX and SB areas and factories in the XZ area.

Along the outer-ring highway of Shanghai, not only does foliar dust have a higher level of metal contamination, but roadside dust also has a higher level of heavy metal accumulation (except for Pb) than in other studies (Fig. 3). In these results, metal levels in urban road dust were much higher than those in urban soil (Akhter and Madany, 1993; De Miguel et al., 1997; De Miguel et al., 1998; Li et al., 2001; Imperato et al., 2003; Biasioli et al., 2006; Shi et al., 2011; Zhang et al., 2013). The levels of Pb were comparatively low, especially compared with samples from such old or large industrial cities as New York, London, Madrid, Seoul, and Hamilton (Fergusson and Ryan, 1984; Harrison et al., 1981; Chon et al., 1995; De Miguel et al., 1997; Droppo et al., 1998). Copper content in this study was comparable to the data in urban dust of other cities but higher than its content in urban soil. Compared with metal content in dust from other cities (e.g., New York [Fergusson and Ryan, 1984], Seoul [Chon et al., 1995], Madrid and Oslo [De Miguel et al., 1997], Hamilton [Droppo et al., 1998], Hong Kong [Li et al., 2001], London [Harrison et al., 1981], and Torino [Biasioli et al., 2006]), the mean values of Cd, Ni, and Zn in this research were much higher than in urban road and soil.

Discussion

Metal Pollution Assessment Based on Igeo Contamination Degree

The average Igeo value of all seven heavy metals in foliar dust at five sites is 1.93 and belongs to the moderately contaminated level, which means that the average concentration of heavy metals in foliar dust is about six times the background concentration. The average Igeo values of each heavy metal in foliar dust were in the order: Cd (3.84, class 4) > Zn (2.85, class 3) > Cu (2.07, class 3) > Hg (1.81, class 2) > Pb (1.71, class 2) > Ni (1.59, class 2) >> As (−0.38, class 0). Therefore, Cd in foliar dust is in the heavily contaminated level; Zn and Cu are in the moderately to heavily contaminated level; Hg, Pb, and Ni are in the moderately contaminated level; and As is in the uncontaminated level. For the each site, the Igeo values of heavy metals in foliar dust were in the order GX > SB > XZ > YB > LD (Table 3). Cadmium in foliar dust at GX and SB is from class 5 areas; Ca at XZ, YB, and LD is from class 4. At GX and SB, Zn in foliar dust is class 4 and belongs to class 3 at XZ, YB, and LD. For Cu, dust was class 3 at

Table 2. Statistic values of total organic carbon and heavy metal concentrations on foliar and roadside dust.

Parameters	Total organic C	Cd	Cu	Ni	Pb	Zn	Hg	As
	mg g^{-1}	mg kg^{-1}						
Foliar dust								
Average	6.86	3.20	191.8	170.6	136.6	1070	0.59	10.2
Minimum	1.66	2.02	145.6	41.0	91.1	509.4	0.23	3.08
Maximum	11.3	5.49	235.8	537.0	191.0	1930	1.22	13.4
SD	3.05	1.23	32.1	149.9	31.7	518.6	0.31	2.88
Roadside dust								
Average	3.72	3.32	160.2	180.6	155.6	976.1	0.44	8.87
Minimum	1.53	0.45	42.6	50.4	30.9	162.4	0.04	3.06
Maximum	6.55	5.61	217.9	388.6	384.9	2152	0.82	12.4
SD	1.74	1.68	53.8	107.6	103.7	598.8	0.25	2.66
Ratio 1†	1.84	0.96	1.20	0.94	0.88	1.10	1.35	1.15
Average of both samples		3.26	176.0	175.9	146.1	1022.84	0.52	9.54
Ratio 2‡		25.1	6.16	5.50	5.74	12.2	5.18	1.05
Background values of Shanghai soil§		0.13	28.6	31.9	25.5	83.7	0.10	9.10
National Standard–Class I¶		0.2	35	40	35	100	0.15	15
National Standard–Class II#		0.6	100	60	350	300	1.0	25
National Standard–Class III††		1.0	400	200	500	500	1.5	40

† Ratios between foliar and roadside dust average values.

‡ Ratios between average of foliar and roadside dust and soil background values of Shanghai.

§ Data from Wang (1992).

¶ Environmental quality standard for soils in China (National Environmental Protection Agency of China, 1995) and numbers in class I are threshold levels of the nationwide natural background.

Metal levels in class II are threshold values established to protect agricultural production and maintain human health.

†† Values in class III are established to maintain normal growth of plants, particularly trees.

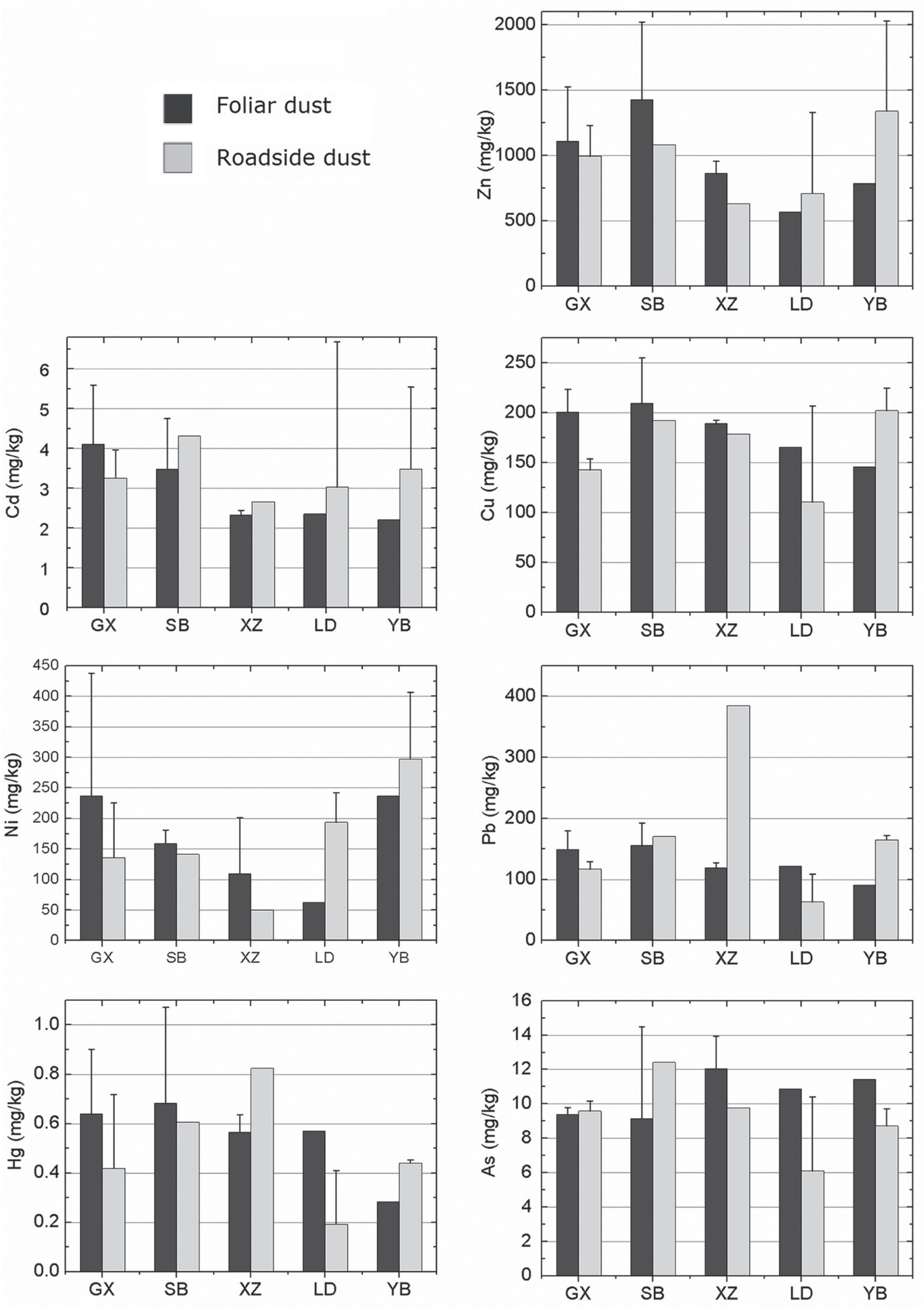

Fig. 2. Spatial distribution of metal levels in foliar and roadside dust along the outer-ring highway. Error bars represent SD of the heavy metal concentrations. GX, Gongfu Xincun; LD, Long Dong Avenue Interchange; SB, Songbin Road; XZ, Xinzhuang Interchange; YB, Ying Bin Avenue Interchange.

GX, SB, and XZ and class 2 at YB and LD. Mercury in GX and SB was also higher than at other three sites, belonging to class 3, whereas it was class 2 at XZ and LD and class 1 at YB. Only at SB was Hg in class 3; the Igeo of Pb was class 2 at the other four sites. Nickel was class 3 at GX and YB and class 2 at SB and XZ but was class 1 at LD. Arsenic was class 0 at all the sites. The spatial distribution patterns of heavy metal pollution in foliar dust is clear: the contamination level decreases from the north to the southeast of Shanghai. The moderately to heavily contaminated areas are located in the northern part of Shanghai (GX and SB), which is the heavy industrial base of Shanghai, with heavy truck traffic and high particle deposition from local factories.

Compared with foliar dust, the average Igeo value of all seven heavy metals in roadside dust at the five sites is 1.91, (i.e., moderately contaminated). The average Igeo values of each heavy metal in roadside dust were in the order: Cd (4.08, class 5) > Zn (2.87, class 3) > Pb (1.99, class 2) > Cu (1.91, class 2) > Hg (1.57, class 2) > Ni (1.56, class 2) > As (−0.59, class 0). Cadmium in roadside dust is class 5, which is higher than that in foliar dust. In roadside dust, only Zn is class 3, and Cu, Hg, Pb, and Ni are class 2. Arsenic belongs to class 0. The spatial distribution of Igeo

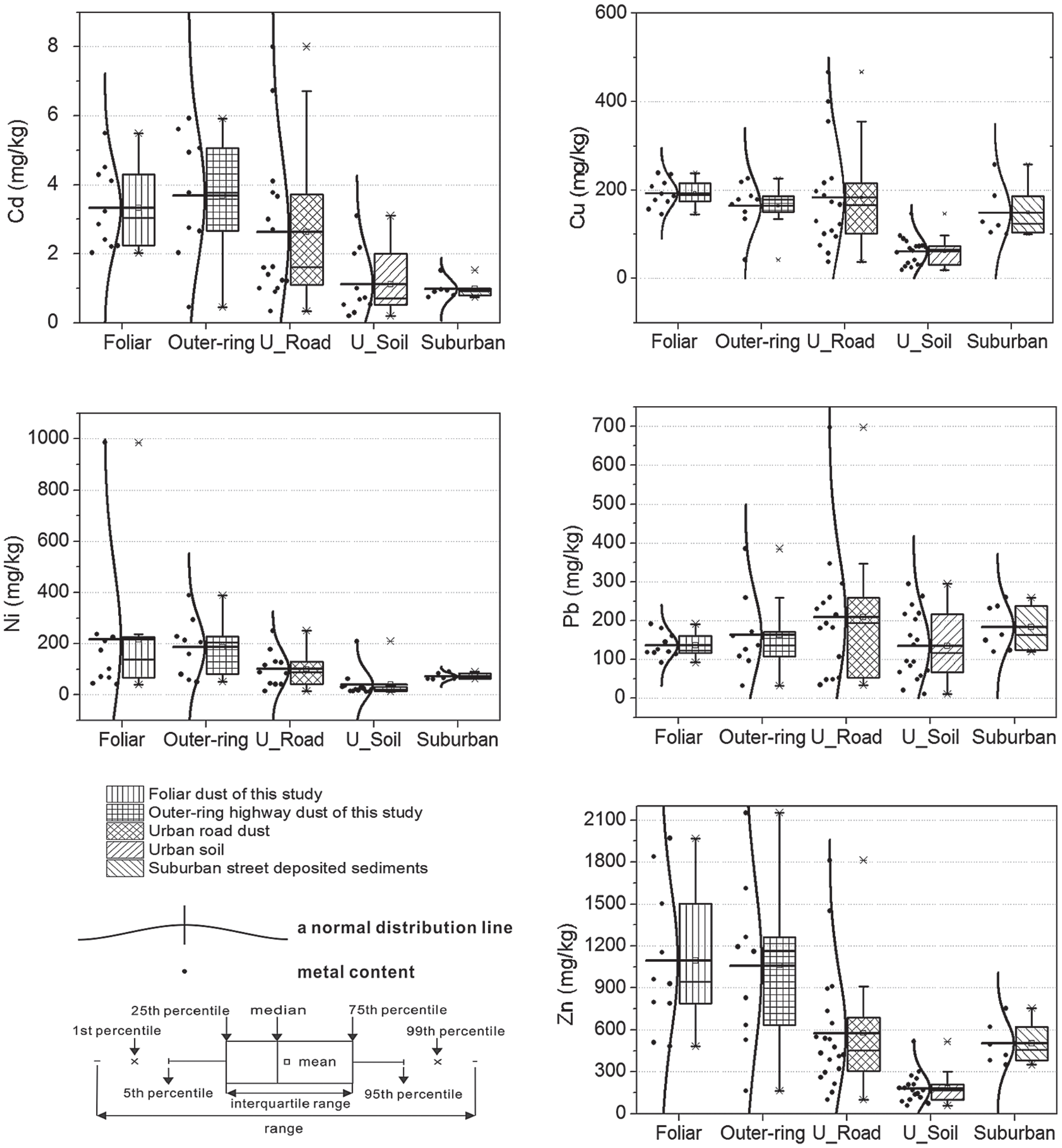

Fig. 3. Comparison of total metal contents between in foliar dust and outer-ring roadside dust in this study and in urban roadside dust, urban soil, and suburban roadside dust in other studies. U_Road, U_Soil, and Suburban data from Akhter and Madany (1993), De Miguel et al. (1997, 1998), Li et al. (2001), Imperato et al. (2003), Biasioli et al. (2006), Shi et al. (2011), and Zhang et al. (2013).

Table 3. Geo-accumulation index values of seven heavy metals in roadside dust and foliar dust.

Site†	Cd	Zn	Cu	Hg	Pb	Ni	As
Roadside dust							
GX	4.06	2.99	1.73	1.48	1.51	1.61	−0.51
SB	4.47	3.11	2.16	2.02	1.56	2.16	−0.14
XZ	3.77	2.34	2.06	2.46	0.08	3.33	−0.48
YB	4.16	3.42	2.24	1.55	2.63	2.11	−0.65
LD	3.96	2.50	1.37	0.36	2.02	0.73	−1.16
Foliar dust							
GX	4.40	3.15	2.22	2.09	2.31	1.97	−0.54
SB	4.16	3.51	2.29	2.19	1.73	2.03	−0.58
XZ	3.57	2.78	2.14	1.91	1.19	1.64	−0.18
YB	3.50	2.65	1.76	0.92	2.31	1.25	−0.26
LD	3.59	2.18	1.95	1.93	0.39	1.67	−0.33

† GX, Gongfu Xincun (Site 1); LD, Long Dong Avenue Interchange (Site 4); SB, Song Bin Road (Site 2); XZ, XinZhuang Interchange (Site 3); YB, Ying Bin Avenue Interchange (Site 5).

Table 4. Pearson correlation coefficients of metal contents in foliar and roadside dust.

Elements	Cu	Ni	Pb	Zn	Hg	As
Cd	0.638**	−0.128	0.225	0.828**	0.494*	0.363
Cu		0.154	0.486*	0.705**	0.582*	0.598**
Ni			−0.107	−0.010	−0.258	0.034
Pb				0.194	0.482*	0.264
Zn					0.515*	0.408
Hg					1.00	0.600**

* Significant at the 0.05 probability level.

** Significant at the 0.01 probability level.

values differ between foliar dust and roadside dust. The average Igeo values of heavy metal in roadside dust at each site were in the order: YB > SB > XZ > GX > LD (Table 3). Cadmium and Zn are the two most important pollutants at the five sampling sites. At XZ, Pb in roadside dust is class 4, and Hg is class 3, which is different at the other sites (Table 3). Except for As, which belongs to class 0 at all sites (the same as in foliar dust), contamination levels for all metals were different at each site. The spatial diversity of heavy metal pollution in roadside dust is not as clear and great as it of foliar dust.

On the whole, heavy metal contamination levels in foliar and roadside dust are higher in the western part of Shanghai (i.e., the Puxi area, on the west side of Huangpu River) than in the eastern part (i.e., the Pudong area) due to the different levels of development and land use. Especially at the GX and SB sites, atmospheric particle deposition from the factories is a main contributor of foliar dust, which is also a main source of heavy metals.

Source Identification Based on Multivariate Statistics Analysis

To identify the source of measured metals along the outer-ring highway, correlation analysis was conducted (Table 4), and connections for interdependence among the different metals in the researched dusts were established by means of cluster analysis, which is efficient for metal source identification (Davis et al., 2009). The results of hierarchical cluster analysis are presented in Fig. 4.

There are four obviously distinguished clusters of seven heavy metals in dusts (Fig. 4): Cd-Hg-As, Cu-Pb, Ni, and Zn. Arsenic concentration in the analyzed roadside and foliar dust was lower than its background value in Shanghai, which was calculated from the local parent material. Therefore, the dust on the road and foliar surface is not mainly from the local soil; the particles originate from outer Shanghai or are produced by other sources, such factories and vehicles. Researches have demonstrated that Zn and Cd derive mainly from aging and wearing of automobile tires (e.g., De Miguel et al., 1997), and gasoline spills and vehicle body aging contribute to the increasing accumulation of Cd and Zn in dust (Li et al., 2001). Although there is significant correlation between Cd and Zn concentration in dust (Table 4), hierarchical cluster analysis implies that in this study Cd not only comes from traffic-related materials (e.g., tire tread, lubricating oil, and brake dust) with Zn but also from the deposition with Hg and As, which are specific markers of coal combustion (Scudlark and Church, 1988; Couture et al., 2008). Atmospheric deposition is the main source of Cd (Gunawardena et al., 2013; Connan et al., 2013) in addition to the traffic sources.

Lead and Cu are in one cluster, which means they derive from the same source or from sources that are close to each other. Lead also has a significant positive correlation with Cu (Table 4), implying the same main source. The sources of Pb in the ambient air are mainly from coal-fired fly ash, industrial activities, and the exhaust of leaded gasoline (Puxbaum et al., 2004). Copper is used in car lubricants (Al-Khashman, 2007) and can come from engine wear (Jaradat and Momani, 1999). Therefore, Pb and Cu in dust mainly come from vehicle engines. Compared with other metals, except As, the ratio of Ni between dust and background value is lowest. Nickel is commonly associated within mafic and ultramafic rock, suggesting that the parent materials of soil controlled Ni concentration (Shi et al., 2010). Moreover, Ni was poorly correlated with the other metals, coupled with long distance to other metals in the dendrogram, suggesting that part of Ni in dust comes from the parent materials of soil.

Conclusions

The mean concentrations of heavy metals (except As) in foliar and roadside dust were significantly higher than the soil background values of Shanghai, especially for Zn and Cd, reaching up to 12.2 and 25.1 times higher than the background values, respectively. The ratios of heavy metal concentrations in foliar dust and roadside dust was in the order Hg (1.35) > Cu (1.2) > As (1.15) > Zn (1.1) > Cd (0.96) > Ni (0.94) > Pb (0.88). Contamination assessment based on the geo-accumulation index revealed that, except for As, the metals investigated in this study were accumulated significantly in the dust along the Shanghai out-ring highway. Cadmium in

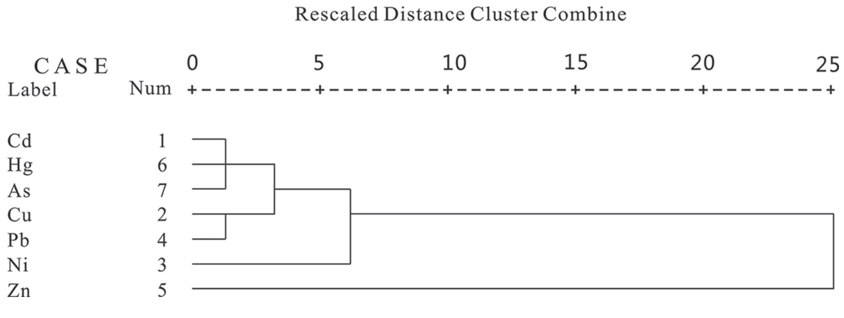

Fig. 4. Dendrogram of the hierarchical cluster analysis of metal contents in foliar and roadside dust. The data were normalized first using within-group linkage as cluster method.

dust samples appeared to be heavily contaminated in both foliar and roadside dust, and Zn was in the moderately to heavily contaminated class. The higher Cd, Zn, and Pb levels were found on road junctions (XZ) and/or in regions near industrial zones (GX and SB). This implies the influence of traffic and industry on the accumulation process of metals in the urban environment. Cadmium, Hg, and As mainly come from atmospheric deposition, and Pb, Cu, and Zn are mainly from heavy traffic on the highway, with Pb and Cu mainly from vehicle engines and Zn mainly from exhaust. Nickel in dust likely comes from parent soils.

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