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Particle size distribution and characteristics of heavy metals in road-deposited sediments from Beijing Olympic Park

*Haiyan Li**, *Anbang Shi*, *Xiaoran Zhang*

Key Lab of Urban Storm-Water System and Water Environment, Beijing University of Civil Engineering and Architecture, Beijing 100044, China

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ABSTRACT

Due to rapid urbanization and industrialization, heavy metals in road-deposited sediments (RDSs) of parks are emitted into the terrestrial, atmospheric, and water environment, and have a severe impact on residents' and tourists' health. To identify the distribution and characteristic of heavy metals in RDS and to assess the road environmental quality in Chinese parks, samples were collected from Beijing Olympic Park in the present study. The results indicated that particles with small grain size (<150 μm) were the dominant fraction. The length of dry period was one of the main factors affecting the particle size distribution, as indicated by the variation of size fraction with the increase of dry days. The amount of heavy metal (*i.e.*, Cu, Zn, Pb and Cd) content was the largest in particles with small size (<150 μm) among all samples. Specifically, the percentage of Cu, Zn, Pb and Cd in these particles was 74.7%, 55.5%, 56.6% and 71.3%, respectively. Heavy metals adsorbed in sediments may mainly be contributed by road traffic emissions. The contamination levels of Pb and Cd were higher than Cu and Zn on the basis of the mean heavy metal contents. Specifically, the geoaccumulation index (I_{geo}) decreased in the order: Cd>Pb>Cu>Zn. This study analyzed the mobility of heavy metals in sediments using partial sequential extraction with the Tessier procedure. The results revealed that the apparent mobility and potential metal bioavailability of heavy metals in the sediments, based on the exchangeable and carbonate fractions, decreased in the order: Cd>Zn≈Pb>Cu.

Introduction

With the rapid growth of urbanization, the density of motor vehicles used to transport the increasing population and essential goods has increased dramatically. Aerosols and road-deposited sediments (RDS) are formed during the process, which has given rise to escalating levels of pollution along roadways in many parts of the world (Loganathan et al., 2013). RDS are the sinks and sources of inorganic and organic pollutants such as heavy metals, metalloids and polycyclic aromatic hydrocarbons (PAHs), which are derived from the emission of vehicles, vehicle tires, brakes and body frames, surfaces of asphalt roads, road railings/fences, deicing salt, paint markers, and pesticides and herbicides added to the pavement (Aryal et al., 2010; Murakami et al., 2008; Perry and Taylor, 2007).

Road-deposited sediments are widely recognized as a major non-point source of heavy metals that is difficult to categorize and manage, due to their dynamic characteristics. These emissions containing particulate matter are released to ambient

* *Corresponding author*. E-mail: lihaiyan@bucea.edu.cn (Haiyan Li).

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air, or deposited on the road surfaces in the form of street emissions (Hur et al., 2007). During rain/storm/street-washing events, the street dust particles laden with contaminants are washed off and finally end up in receiving water bodies (Murakami et al., 2008; Yu et al., 2001; Jain, 2004; Stead-Dexter and Ward, 2004). Furthermore, the mobilization of heavy metals into the atmosphere by industrial activities has become an important process in the geochemical cycling of these metals. This is acutely evident in urban areas, where large quantities of heavy metals from various stationary and mobile sources are released into the atmosphere, plants and soil, exceeding the natural emission levels (Shi et al., 2008; Fujiwara et al., 2011; Škrbić et al., 2012). In fact, heavy metals are considered to be toxic pollutants, because they can accumulate in the sediments due to atmospheric deposition by sedimentation interception and may affect human health if their concentration reaches a certain level (Ferreira-Baptista and De Miguel, 2005).

In recent decades, a number of studies have focused on the concentration, distribution and source identification of heavy metals in street side dusts (Manno et al., 2006; Andersson et al., 2010; Cao et al., 2011). Most of the studies have shown that the concentration of heavy metals (Cu, Fe, Cd, Mn, Ni, Pb, Zn) decreases with the increase of particle size, and the highest concentration was measured in the finest fraction of the particles with size < 63–75 μm (Zhao et al., 2010, 2011; Singh, 2011; Lee et al., 2013). Metals in the fine fraction are generally considered to arise from exhaust emissions, whereas metals in the coarse particles are considered to be derived from the components of wear and tear of vehicles (Duong and Lee, 2011; Lim et al., 2006). Besides, study focusing on the heavy metal chemical compositions is important to assess their mobility and hence bioavailability, using sequential extraction. Cd was identified to be the most bioavailable element among heavy metals, as it shows the greatest affinity to operationally defined exchangeable sites and carbonates in sequential extraction (Banerjee, 2003). Zn and Pb are mainly associated with carbonates > Fe/Mn oxides > the exchangeable fraction (Li et al., 2001). Moreover, the vast proportion of Cu was bound to organic matter and a small proportion was an exchangeable matter, therefore Cu is the least likely to release to the environment under natural conditions (Peng et al., 2009). In general, to the best of our knowledge, the index of bioavailability reported in the limited number of studies conducted shows the order of Cd > Zn = Pb > Cu (Charlesworth et al., 2003).

A great number of studies on heavy metals in RDS have focused on developed countries (Kumar et al., 2010, 2013a; Lee et al., 2013; Kurt-Karakus, 2012; Lau and Stenstrom, 2005), but little information on heavy metals in RDS is available for developing countries, including China (Zhao and Li, 2013; Zhao et al., 2011), especially in public parks. The heavy metal pollution is determined by calculating the value of the integrated pollution index (IPI), concentration factor (CF), element enrichment factor (EF) and geoaccumulation index (I_{geo}) (Chen et al., 2005; Gong et al., 2008) while the health risk assessment of the metal is determined by calculating the hazard quotient (HQ) and health index (HI) in surface soils of urban parks in Beijing (Luo et al., 2012). The concentrations of Cu, Zn, Cd and Pb were much higher than their background values in Chinese soil and the health risk assessment of heavy metals in road dusts in Beijing urban parks indicated that ingestion, dermal contact and inhalation were the three main exposure pathways for people (Du et al., 2013). In addition, the results on the distribution of heavy metals in sediment from a public park lake suggested that potentially large contributions from point sources were related to human activities in highly urbanized regions (Yang et al., 2014). Although these studies of heavy metals in urban park soil and road sediment have been a central issue in China, there is little detailed data on the origin, distribution and concentration of heavy metals in the RDS in the Beijing parks. Among the different species of contaminants, heavy metals such as Cu, Cd, Pb and Zn are of particular concern due to their prevalence and persistence in the environment (Stead-Dexter and Ward, 2004). Therefore, the main objectives of this study are: (1) to determine the relationship between the particle size characteristics and the chemical compositions of several heavy metals (*i.e.*, Cu, Zn, Pb and Cd), using partial sequential extraction procedures, in sediment samples collected from Beijing Olympic Park (a famous tourist attraction in China), (2) to investigate the heavy metal contamination assessment in order to evaluate the road environmental quality of the dust in the urban parks and the potential risks to residents and tourists based on the geoaccumulation index (I_{geo}), and (3) to identify the potential heavy metal pollution contribution to the receiving park water bodies based on their availability due to the apparent mobility and potential metal bioavailability.

1. Materials and methods

1.1. Study area

Beijing, the capital of China, (39°54′N, 116°24′E) is located at the northern tip of the roughly triangular North China Plain, and spans 16,800 km^2. The Olympic Park is located at the northern end of the central axis of Beijing and is bounded by the Qing River and the North 5th Ring Road. The traffic load of the North 5th Ring Road is very heavy (300,000 vehicles per day). Furthermore, the park is bounded by the North 4th Ring Road (comparably busy) to the south, the Anli Road to the east, and the Lincui Road to the west. The national stadium, swimming center, and gymnasium are all sited near the Olympic Park (Qiao et al., 2011).

Particles were collected from eight different sampling sites (Table 1 and Fig. 1) within the Beijing Olympic Park, including industrial areas, areas with heavy and low traffic density, commercial areas and residential districts. The roads where the sampling was performed were major intersections. The samples were collected in the afternoon (15:00–17:00) of 14th, 28th Oct. and 12th, 25th, Nov. 2013 when the traffic loads were low. Before the samples of the last three times were collected, the length of the dry period was 7, 2, and 15 days, respectively.

1.2. Sample collection

The dust samples were collected from both sides of the road at the intersection on a dry day using a plastic dustpan and brush. An area generally ranging within 0.5 m of the curb of the road (Sartor and Boyd, 1972) and 30–50 m in length was swept in order to obtain a sufficient amount of sample for analysis and to

Table 1 – Sampling sites.

Sample location	Street side	AADT	Area sampled (m^2)	Sampling time
B&D	The intersection of East Beichen Road and North Datun Road	2500	20	15:00–15:30
NC	The bird's nest	NA	33	15:00–15:30
T&H	The intersection of West Tianchen Road and Hui Zhonglu Road	NA	28	15:30–16:00
B&R	The intersection of West Beichen Road and Ruyi Bridge	2100	30	16:00–16:30
GWH	The front of the Park Management Committee	NA	30	15:00–15:30
D&T	The intersection of Datun Road and East Tianchen Road	600	25	15:00–15:30
SLF	The Water Cube	NA	35	16:30–17:00
K&T	The intersection of South Kehui Road East Tianchen Road	NA	22	15:00–15:30

ADDT: annual average daily traffic (in thousands).
NA: not available.

overcome the problem of localized spatial variation. The sample of road dust in each location was 100–500 g in mass. All samples were kept in the sealed polyethylene bags after removal of large debris on-site, and then transported to the laboratory and stored at 4°C. The sampling procedure is similar to that reported previously (Lau and Stenstrom, 2005; Duong and Lee, 2009).

1.3. *Particle sieving*

Sieving was conducted in the laboratory to fractionate the samples without contaminating them. Stainless steel screens were purchased in six size scales with openings from 38.5 to 2000 μm. All samples were air-dried in the laboratory for 3 days before sieving. Road dusts were sieved through a stainless steel sieve with a mesh diameter of 2000 μm to remove refuse and small stones. Samples were sorted into six grain size fractions, with six different size distribution ranges: 830–2000, 300–830, 150–300, 76–150, 38.5–76 and <38.5 μm. Then the samples were weighed and heavy metals were analyzed for each size fraction.

1.4. *Analytical methods*

The sequential extraction methods were adapted from that reported by Tessier et al. (1979). Extraction helps to quantify five element fractions with different solubilities under different environmental conditions in the road dusts, namely exchangeable, carbonate bound, iron and manganese oxide bound, organic matter bound and residual (Peng et al., 2009). To minimize the source of error, blanks and duplicate samples were conducted in the analytical procedures to assess the precision and bias. All the extractions and analyses were conducted in triplicate (n = 3) for quality control and the average values were reported. Heavy metals in the road dust were extracted by the HF-$HClO_4$ digestion method on a hotplate. The concentrations of Zn and Cu were determined by flame atomic absorption spectrometry while the concentrations of Pb and Cd were measured by graphite furnace atomic absorption spectrometry (Hitachi Z-2010, Tokyo, Japan). If the spectrometer gave a reading that exceeded the calibrated range of a heavy metal, the sample was diluted. All

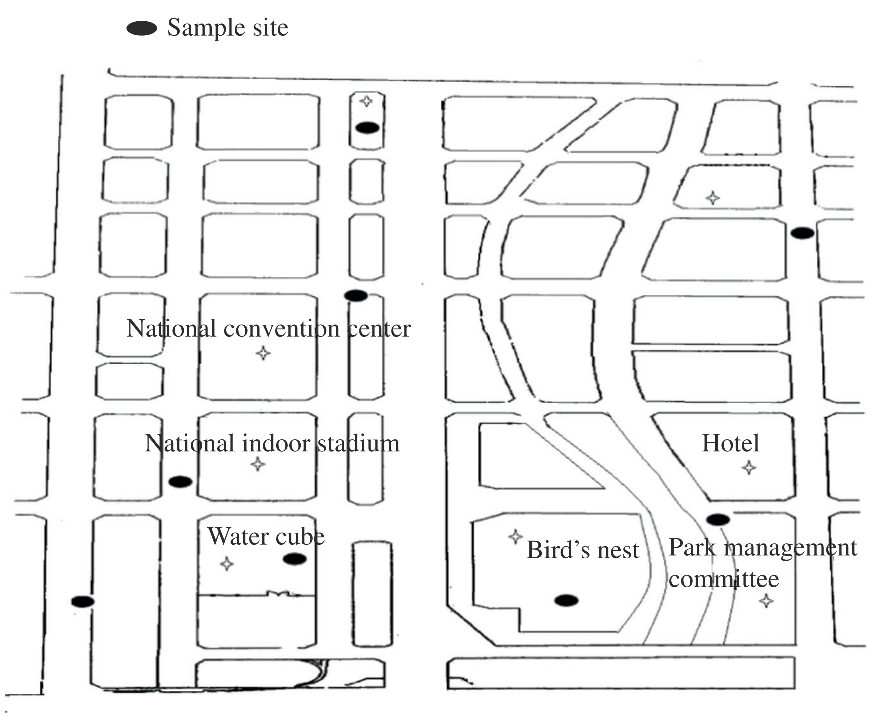

Fig. 1 – Sampling sites of RDS in the Beijing Olympic Park area, China.

the plastic vessels and glassware were treated by dilute (1:1) nitric acid for 24 hr, then rinsed with distilled water before use. The analytical precision, measured as relative standard deviation, was routinely between 3% and 5%. Accuracy of analyses was checked by standards and duplicate samples. The recovery metal concentrations from the reference materials were as follows: 102% (Zn), 98% (Cu), 96% (Pb), 97% (Cd) and 93% (Cr). Precision can be considered satisfactory for environmental analysis, as it was within 5% of the relative percentage difference.

1.5. *Methods of contamination assessment*

A number of calculation methods have been put forward to quantify the degree of metal enrichment or pollution in sediments and dusts (Odewande and Abimbola, 2008; Lu et al., 2009). In the present study, the geoaccumulation index (I_{geo}) was calculated to assess the level of heavy metal contamination in the RDS. I_{geo} is calculated by Eq. (1):

$$I_{geo} = \log_2(C_n/1.5B_n) \qquad (1)$$

where, C_n represents the measured concentration of the element and B_n is the geochemical background value of the element in fossil argillaceous sediment (average shale). In the present study, B_n is the background content of the element in Chinese soil. The constant 1.5 is introduced to minimize the effect of possible variations in background values that may be attributed to lithological variations in the sediments.

2. Results and discussion

2.1. *Particle size distribution*

The grain size distribution of RDS is a particularly important factor because it determines the mobility of particles and the concentrations of the associated pollutants (Zhao et al., 2010). The concentration and total amount of pollutants in different particle size fractions of street dusts are important parameters to assess the transport of sediment-bound pollutants and pollution control by various remedial methods. Different studies based on road sediments have shown that the concentration of heavy metals (*i.e.*, Cu, Fe, Cd, Mn, Ni, Pb and Zn) decreases with the increase of particle size, and the highest concentrations were measured in the finest fraction, <63–75 μm (Ewen et al., 2009; Singh, 2011).

Fig. 2 shows the results of particle size distributions of street dusts from the eight sampling sites in Beijing Olympic Park. Consistent with the trend reported in the literature (Herngren et al., 2006), particles with the size of 300–830, 150–300, and 76–150 μm were the most abundant, which indicated that the particles collected were most numerous in the middle size ranges. Particles with the size of 830–2000 μm showed the greatest abundance in NC, GWH and SLF areas with very low vehicle traffic. On the contrary, the percentages by weight of these particles were lower than 5% in B&D and B&R areas, where the traffic density was >2000 vehicles/hr. The reasons may be attributed to the different traffic volumes at the various street junctions, with the brake and acceleration behavior enhancing road abrasion and causing this diversity (Zereini et al., 2007). The results revealed that the size distributions of street dusts were affected by the traffic activity very obviously.

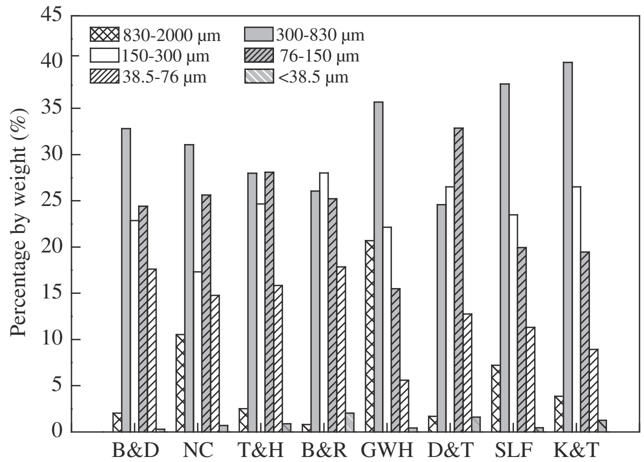

Fig. 2 – Size fractionation as a function of particle size from different sampling sites. Locations are referred to Table 1.

As shown in Fig. 3, the size fraction of the dusts collected from the B&R site indicated that the effect of the length of dry period on the particle size distribution was significant (Egodawatta et al., 2007; Egodawatta and Goonetilleke, 2008), which was a typical and representative experimental result among all sites. Overall, a higher percentage by weight of fine particles (<150 μm) was observed with increasing numbers of dry days. With dry days changing from 2 to 15 days, the percentage by weight of particles <38.5, 38.5–76, and 76–150 μm increased 62.70%, 99.89%, and 29.58%, respectively. Furthermore, previous researchers (Zhao et al., 2010, 2011; Singh, 2011; Lee et al., 2013) suggested that the highest concentration of heavy metals was measured in the finest fraction, <63–75 μm, therefore the sweeping frequency needs to be strengthened for dust removal (<150 μm). However, the accumulation of particles with grain sizes of 150–300 μm remained stable (26.52%–28.02%), while the decrease of the contents of particles with grain sizes of 300–830 and 830–2000 μm was clearly demonstrated. Specifically, the decrease rate of particles with grain sizes of 300–830 and 830–2000 μm reached 34.85% and 79.02% respectively. This could be due to the influence of braking, acceleration and compaction by rolling behavior over extended dry periods, enhancing road abrasion, on the particle-size distribution of material that accumulates at the edge of the road. Therefore, it could be postulated that the length of dry period was one of the main factors that affected the distribution of road dusts, and the effect on particles with different grain sizes was distinctly different. The size fraction of the dusts collected from other sites was not shown in the figure as the trend was similar.

2.2. *Heavy metal concentrations of RDS*

The RDS grain size distribution has a significant effect on mediating transport and chemical interactions (Duong and Lee, 2011; Herngren et al., 2006). An important index of contamination for RDS is the mass load of a heavy metal in a given grain size fraction (Zhu et al., 2008). The statistical results of

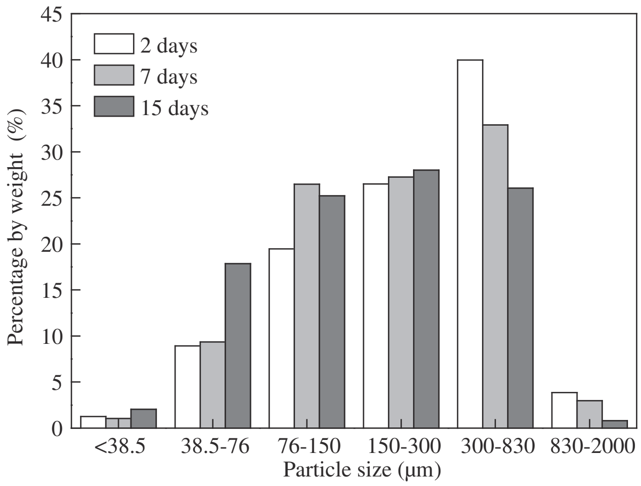

Fig. 3 – Particle size distribution of street dusts loading over dry period.

heavy metal (*i.e.*, Cu, Zn, Pb, and Cd) concentrations investigated in the studied samples in each grain size fraction are presented in Fig. 4. The results indicated that the concentrations of heavy metals (Pb and Zn) decrease with the increase in particle size as a whole. The exception was the observation of fluctuations in the concentrations of Cu and Cd. The highest metal concentrations were measured in the <38.5 μm fraction, with the exception of Cu and Zn. The highest concentrations of these two metals were observed in the 38.5–76 and 76–150 μm fractions, respectively.

The results indicated that Pb was the most abundant element in particles for all of the size fractions. Although unleaded gasoline has been widely used in China, possible explanation for this observation was that the sources of Pb were coal combustion, construction materials, paint, brake linings, proximity to the oldest urbanized sectors (downtown and surrounding old neighborhoods) and lead-based weights added to vehicles for tire balance (Thorpe and Harrison, 2008; Del Rio-Salas et al, 2012; Root, 2000). Moreover, the lowest concentration of Pb, in the 300–830 μm fraction, was six times higher than the highest concentration of Zn in the 76–150 μm fraction, at 10.16 mg/kg. The distributions of these two metals may be due to the fact that Zn derived from brake pads was difficult to remove and transfer into the sediments, unlike Pb (Thorpe and Harrison, 2008). The concentration of Cu in the <38.5 μm size fraction was only 9.93%. The result was quite different from other metals, as the level suggested that the smallest concentration of Cu was measured in the finest size. In contrast, the highest concentration (1.11 mg/kg) for Cd was observed in the finest size fraction (<38.5 μm). In general, the results indicated that the heavy metal concentration was the lowest in the >300 μm fraction of all the samples. Therefore, the fine particles (<300 μm) from RDS during street sweeping is important to remove.

In addition, the mean total concentration of Pb was generally similar to those observed in earlier studies (Kumar et al., 2013b; Herngren et al., 2006). The concentration of Cd in each grain size fraction in the present study was similar to the values reported in other studies (Zhao et al., 2010; Li et al., 2001), but was significantly higher than the background values in the soil of

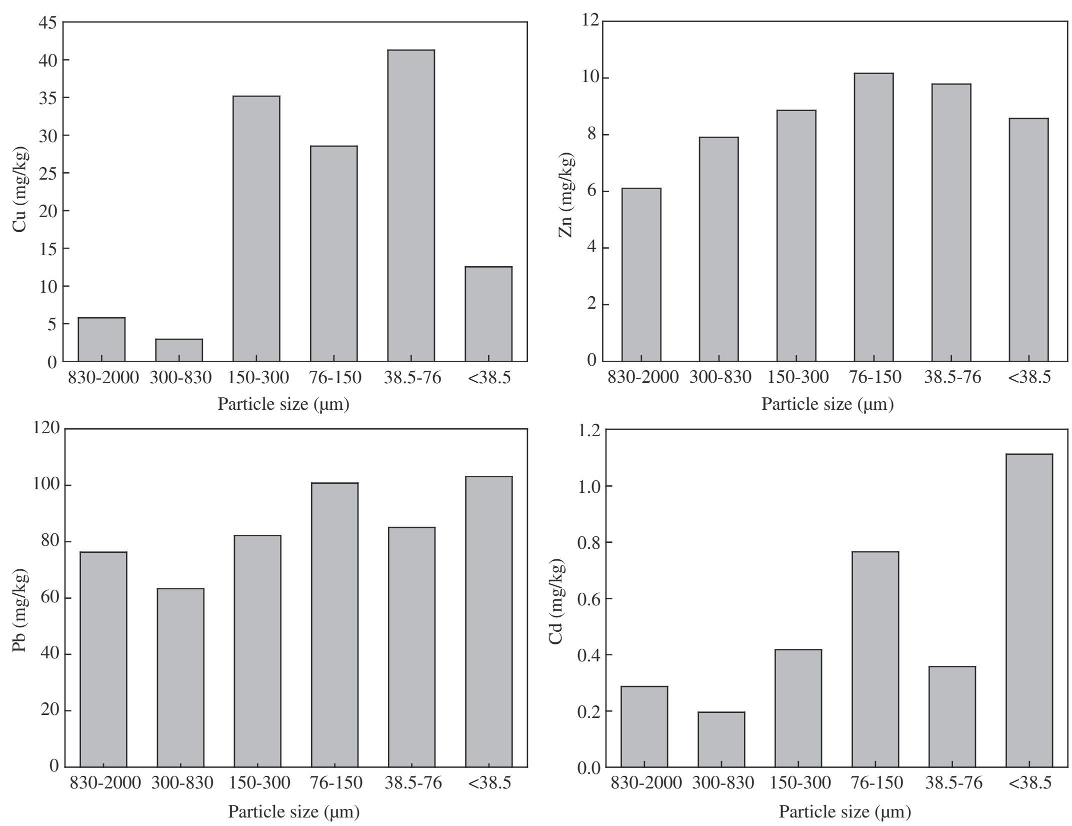

Fig. 4 – Average heavy metal concentrations in street dusts from Beijing Olympic Park.

Beijing (China National Environmental Monitoring Center, 1990).

2.3. Results of heavy metal contamination assessment in RDS

The total concentrations of heavy metals in street dusts of Beijing Olympic Park, derived by summing the sub-concentration of each heavy metal and accounting for the corresponding mass fraction of the classified particle size group, were compared with the data reported among different particle grain size ranges and different cities worldwide (Table 2). It was evident that Cu, Pb and Cd pollution existed in the park dust samples, with the values of these metals higher than the background concentrations in the soil of Beijing, and in China. In addition, the mean concentration of Zn in this study was lower than those sampled in 13 different urban parks of Beijing, while the mean concentration of other metals was higher. The results indicated that the contamination by these metals in Olympic Park was exacerbated, to a certain extent. Furthermore, the mean concentration of Cu in the studied samples was similar to those sampled in Baoji, Kavala, but lower than Barcelona, Guangzhou, Birmingham, Istanbul, Huludao, Shanghai and Buenos Aires. Moreover, the mean concentration of Zn was lower than for any city shown in the table. However, the concentration of Pb was similar to that in dust sampled in Huludao, Baoji and higher than other cities, except for Banja Luka and Newcastle upon Tyne. On the other hand, the mean concentration of Cd was only lower than the cities of Huludao and Istanbul. In general, each city has its own characteristic combinations of elemental compositions, and the observed similarities as well as variations may not reflect actual natural and anthropogenic diversities among the different urban settings. To represent the level of pollution, the geoaccumulation index is widely used, which has been established as a standard procedure to represent and analyze urban sediments.

The calculated results of I_{geo} of heavy metals in Olympic Park street dusts were computed from the mean total Cu, Zn, Pb, and Cd concentrations (Table 3). The I_{geo} for Cu, Zn, Pb and Cd were 1.87, −1.51, 3.79 and 5.03, respectively, which indicated that the values of I_{geo} decreased in the order of Cd > Pb > Cu > Zn. The I_{geo} of Cu, which falls into class 2, indicates that the dust samples were moderately polluted. Moreover, the I_{geo} obtained for Zn indicated little or no pollution, as the value fell into class 1. Furthermore, the fact that I_{geo} of Pb fell into class 5 showed that there was significant Pb pollution in street dust. Finally, the I_{geo} of Cd in class > 5 revealed that the park samples were extremely polluted.

2.4. Metal speciation in RDS

In sediments, metals could be bound to various compartments in different ways: occluded in amorphous materials; adsorbed by clay surfaces or iron/manganese oxyhydroxides; present in the lattice of secondary minerals such as carbonates, sulfates or oxides; complexed with organic matter or in the lattice of primary minerals such as silicates (Yu et al., 2001; Banerjee, 2003). Since each form shows different remobilization potentials, affecting its respective bioavailability and toxicity, the measurement of the total amount of metal may not be able to provide essential information on the characteristics of pollution. To clearly evaluate the toxicity of heavy metals to aquatic biota, in the past decades, different sequential extraction procedures for partitioning the metals bound to various mineral components were developed (Jain, 2004). Among all these sequential extraction methods, the five-step method mainly established by Tessier et al. (1979) has been most commonly used. Normally, the sum of the mobile and exchangeable fractions can be used to assess the environmentally available components. The fractions bound to Mn oxides and organic materials are assumed to represent

Table 2 – Comparison of the mean heavy metal contents in road dust among Beijing and other cities worldwide.

City	Cu (mg/kg)	Zn (mg/kg)	Pb (mg/kg)	Cd (mg/kg)	Particle size	Reference
Newcastle upon Tyne, UK	132	421	992	1.0	<250 μm	Okorie et al., 2012
Birmingham, UK	466.9	534.0	48.0	1.62	<63 μm	Charlesworth et al., 2003
Barcelona, Spain	1332	1572	248	3	<10 μm	Amato et al., 2011
Buenos Aires, Argentina	190	751	208	NA [a]	<50 μm	Fujiwara et al., 2011
Banja Luka, Bosnia and Herzegovina	77.7	272	608	1.39	<2000 μm	Škrbić et al., 2012
Istanbul, Turkey	1039	227	222	3.9	<500 μm	Sezgin et al., 2004
Kavala, Greece	124	272	301	0.2	<63 μm	Christoforidis and Stamatis, 2009
Baoji, China	123.2	715.3	433.2	NA [a]	<75 μm	Lu et al., 2009
Huludao, China	264.4	5271	533.2	72.84	<1000 μm	Zheng et al., 2010
Guangzhou, China	176	586	240	NA [a]	<2000 μm	Duzgoren-Aydin et al., 2006
Shanghai, China	196.8	734	295	1.2	<125 μm	Shi et al., 2008
Urban parks of Beijing	72.13	219.20	201.82	0.64		Du et al., 2013
Beijing Olympic Park, China	126.3	51.38	510.7	3.14	<2000 μm	This study
Background in soil of China	23.1	97.2	24.7	0.053		China National Environmental Monitoring Center
Background in soil, Beijing	18.70	57.50	24.60	0.12		China National Environmental Monitoring Center

[a] Not available.

potentially mobile components under changing conditions, and are viewed as the most important components in sediments for metal binding.

The results of the chemical fractionation patterns of Cu, Zn, Pb and Cd in the street dusts according to the sequential extraction method adapted from that of Tessier et al. (1979) are shown in Fig. 5. The fractionation pattern of metals showed a certain similarity between all the selected samples, irrespective of their wide range of metal levels. The results are graphed as leaching percentages, reflecting individual fraction removal against the sum of all fractions.

Copper partitioning was dominated by the organic (F4) fraction (>70% of total Cu except for the largest particles) in all the dusts with different sizes, indicating that the fraction was of major importance as Cu carrier in the roadside sediments, and the exchangeable fraction accounted for a very low percentage of Cu, varying from 0.11% to 1.38% with an average of 0.57%. The present results are in agreement with previous studies on road sediments (Charlesworth et al., 2003; Robertson et al., 2003). The high percentage of Cu measured in the organic fraction may be due to its strong tendency to form complexes with organic matter. Although the proportions of Cu in the different size particles present in the Fe–Mn oxide fraction varied considerably, the presence of Cu in the Fe–Mn oxide fraction (0.02%–46.51%) suggested that it was the second most important non-residual fraction, which was in broad agreement with a previous study on urban park soils and dusts in Hong Kong (Li et al, 2001). The role of the exchangeable (F1), carbonate (F2) and residual (F5) fractions was not significant, accounting on average for < 3.5% of total Cu (Fig. 5). Exchangeable Cu was quite low, which was probably due to strong specific (covalent) interactions of Cu with organic matter and other surfaces (Peng et al., 2009).

Zn partitioning was dominated by the carbonate (F2) fraction (average 32.11% of total Zn) and the reducible (F3) fraction (average 33.17% of total Zn), indicating that majority of Zn was associated with the carbonate and Fe–Mn oxide fractions. Our results showed a similar trend to that reported in earlier studies on soils, sediments and street dusts (Banerjee, 2003; Lee et al., 2005). These results could be explained by the following reasons: the stability constants of Zn oxides are high, and $CaCO_3$ may act as a strong adsorbent for heavy metals and could complex as double salts such as $(Ca, Zn)CO_3$. Furthermore, the percentage of the residual fraction of the dusts tended to decrease while the distribution pattern in the carbonate fraction increased with decreasing particle size. The results suggested that the enrichment capability of the residual and carbonate fractions of Zn might be related to particle size. Moreover, the percentages of total Zn in the exchangeable fraction observed in the present study were very low, and little enrichment of Zn in the residual fraction was noted.

Pb in the street dust (38.5–2000 μm) in the same size range showed the order of association: Fe–Mn oxide bound > residual > organic. The percentage of the total concentration of Pb in these fractions remained stable with changes in sediment size. Moreover, the exchangeable fraction accounted for a low percentage (0.77%–4.81%) in the road dusts. Except for the dusts < 38.5 μm, with very high concentration of Pb in the carbonate fraction, all of the other samples showed a similar pattern for Pb, with a low percentage. In addition, the percentage of Pb in the residual fraction appeared to decrease with decreasing particle size. These results indicated that the reducible, oxidizable, and carbonate fractions contained most of the Pb (total sum of 83.8%) in road dusts in Olympic Park.

Different from Pb, the exchangeable fraction proportions of Cd of the six size fractions were 9.58%, 22.57%, 9.19%, 20.79%, 23.98% and 4.64%, respectively, which is consistent with a previous observation (Duong and Lee, 2009). The present results show that Cd is the most bioavailable element, and highlighted the high potential mobility of Cd. The exchangeable fraction allowed Cd to get into the water systems of biological organisms most easily. On average, 21.19% of Cd was present in the carbonate fraction, with the substitution of Cd^{2+} for Ca^{2+} in calcite and precipitation of $CdCO_3$ at higher pH expected (Peng et al., 2009). The concentrations in the five fractions were nearly independent of the size of the road dusts, as each fraction contained a certain amount in each size. The smallest percentage of Cd (average 11.67% of total Cd) was observed in the organic fraction, which might mainly due to the fact that Cd–organic complexes, if present, are only loosely bound and are easily removed.

The percentages of total Zn and Pb in the non-residual fraction both rose as the size of the road sediments decreased. In fact, the environmental impact of sequential speciation categories depends on the ease of remobilization, and the metals in non-residual fractions could release their metal loads with changes in the environment more easily than metals in the residual fraction. Generally, the first two fractions could release their metal loads on lowering of the pH and are more mobile than the other fractions. Therefore, based on the first two fraction values, the results indicated that the apparent mobility and potential metal bioavailability for these park sediments was: Cd > Zn ≈ Pb > Cu. To reduce the bioavailability and mobility of the pollutants, suitable remediation measures should be adapted.

Table 3 – Relationship of geo-accumulation index (I_{geo}) and the pollution level and I_{geo} of heavy metals in road dusts.

I_{geo}	Pollution level	Heavy metals	I_{geo}
<0	Practically unpolluted	Cu	1.87
0–1	Unpolluted to moderately polluted		
1–2	Moderately polluted	Zn	−1.51
2–3	Moderately to strongly polluted		
3–4	Strongly polluted	Pb	3.79
4–5	Strongly to extremely polluted		
>5	Extremely polluted	Cd	5.03

3. Conclusions

The results indicated that the road dust grain size distribution was the key factor involved in determining the particle mobility and its associated heavy metal load. The cumulative percentage of sediments with a smaller grain size (<150 μm) accounted for 43.57% (on average), as the percentage of metal content in these particles accounted for 74.7% of Cu, 55.5% of Zn, 56.6% of Pb and 71.3% of Cd, respectively. High concentrations of the metals were observed, especially for Pb and Cd. In addition, the data on heavy metal concentrations of street dusts showed that roadside

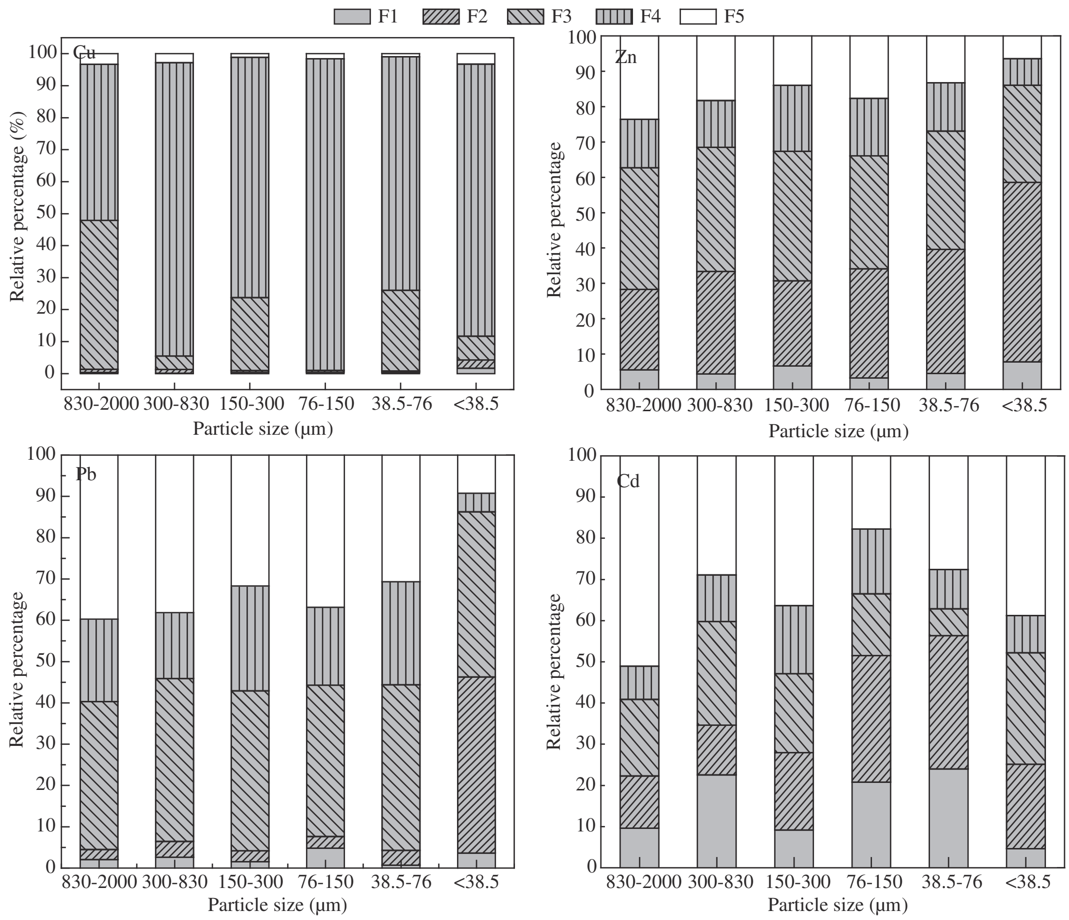

Fig. 5 – Operationally defined solid phase fractionation for Cu, Zn, Pb and Cd for street dust samples from Beijing Olympic Park. F1: exchangeable; F2: carbonate bound; F3: Fe–Mn oxide bound; F4: Organic bound; F5: residual.

sediments received considerable inputs of anthropogenic metals, primarily from automobiles.

Speciation data indicated that Cu was mainly found in the organic fraction, while Zn and Pb were preferentially bound to Fe–Mn oxides. The exchangeable and residual fraction of Cd was the highest among all these four metals and low amounts of metals were found in the residual form. The calculated results of I_{geo} decreased in the order of Cd > Pb > Cu > Zn. The high I_{geo} for Cd and Pb in street dusts indicated that there was a considerable Pb and Cd pollution, which mainly originated from traffic activities. Beijing Olympic Park is a famous tourist attraction in China, therefore, it would be advisable to limit or reduce moving vehicles in the park and use a combination of mechanical and vacuum-assisted sweepers to achieve the removal of particles of small size, as well as the great volume of road dusts and pollutants.

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