Heavy Metal Contents of Road-Deposited Sediment along the Urban–Rural Gradient around Beijing and its Potential Contribution to Runoff Pollution

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Supporting Information

ABSTRACT: Understanding the contribution of road-deposited sediment (RDS) and its washoff process is essential for controlling urban runoff pollution. Ninety-seven RDS samples were collected along the urban–suburban–rural gradient from areas of five administrative units in the Beijing metropolitan region, including central urban (UCA), urban village (UVA), central suburban county (CSA), rural town (RTA), and rural village (RVA) areas. RDS washoff was evaluated with different particle sizes using a rainfall simulator. Heavy metal elements (i.e., Cr, Cu, Ni, Pb, and Zn) were estimated in both RDS and runoff samples. The RDS mass per unit area increased in the order UCA ($21 \pm 24 \text{ g/m}^2$) $\approx$ CSA ($20 \pm 16 \text{ g/m}^2$) $<$ RTA ($59 \pm 63 \text{ g/m}^2$) $<$ RVA ($147 \pm 112 \text{ g/m}^2$) $\approx$ UVA ($147 \pm 198 \text{ g/m}^2$). Compared to RDS from the other administrative units, RDS from the UCA and CSA had higher metal concentrations and higher proportions of smaller particles, whereas that from the RVA and UVA had larger quantities of metals per unit area. UCA and CSA had lower potential runoff pollution contributions per unit area. Our findings imply that controlling the first flush in the UCA and CSA, and improving existing street cleaning methods and road surface conditions in the TRA, UVA, and RVA will be appropriate strategies for controlling runoff pollution from RDS.

INTRODUCTION

Road-deposited sediment (RDS) is an important carrier of heavy metal contaminants, and can contribute to urban runoff pollution.^{1,2} Rapid urban expansion has increasingly caused urban surface runoff pollution problems.³ Along with rapid urban expansion, a large proportion of urban land is being converted to roads and zoned for commercial, industrial, and residential use.⁴ Urbanization increases the population density and intensifies human activities such as traffic, industry, and energy consumption via coal and natural gas.⁵ These activities produce large quantities of RDS particles and contribute to delivery of heavy metal pollutants to urban water bodies.^{6,7} Vehicle emissions in particular are a major contributor to metals in RDS, including Pb, Cu, Cd, and Zn.⁸ Compared to nonindustrial land, industrial land commonly has higher concentrations of heavy metals in both RDS and soil.^{9,10} There are also a number of other sources of metals in RDS, including coal combustion, disintegration of vehicle brakes and tires, atmospheric deposition, road surface wear, municipal solid waste incineration, and residential heating.¹¹ These metals do not degrade, and they accumulate and contribute to air pollution and contamination of surface runoff.¹² At the same time as the pollution sources are increasing, the coverage of land with impervious materials such as asphalt, concrete, and stone is also rapidly increasing because of urbanization.⁴ Impervious surfaces typically cover 60–100% of a typical urban area.¹³ Because these surfaces are impenetrable, large quantities of the pollutants associated with RDS accumulate on them.¹⁴

Consequently, RDS polluted surface runoff from impervious surfaces contains a high pollutant load.¹⁵

The quantity, grain size, and particle mobility of RDS are critical to the assessment of the role of RDS in heavy metal pollution in urban runoff.¹⁶ Sartor and Boyd (1972) carried out one of the earliest studies on accumulation of RDS on impervious surfaces.¹⁷ Their study and subsequent studies have shown that the quantity of RDS is affected by many factors such as the land use zoning, street texture, and street cleaning method.^{18,19} Rough street surfaces can create a large permanent storage area for street dust.¹⁸ Regional industrial and other human activities also affect the quantity of RDS per unit area.¹⁶ The grain size has an important influence on the mobility of particles and their associated pollutant concentrations.²⁰ RDS less than 100 μm in diameter is the most important fraction in surface runoff.²¹ However, there is still a lack of understanding of how the urban–suburban–rural gradient affects the quantity, grain size, and particulate mobility of RDS.

Because of rapid urbanization in China, urban surface runoff containing contaminated RDS is becoming an increasingly serious environmental problem. The Beijing metropolitan region is a useful area for a representative study of the urban–suburban–rural

Received: January 28, 2011

Accepted: August 5, 2011

Revised: July 5, 2011

Published: August 05, 2011

gradient and rapid urbanization in China. Along the urban–suburban–rural gradient, there are obvious differences in land use zoning, population density, proportion of surfaces that are impervious, traffic density, energy consumption, street cleaning methods, and industry. In this study, the following aspects were addressed: How do the quantity and its grain size distribution of RDS vary along the urban–suburban–rural gradient in the Beijing metropolitan region? What are the influences of urban–suburban–rural gradient on the heavy metal distribution in RDS and potential pollution contribution in surface runoff?

MATERIALS AND METHODS

Study Area. The Beijing metropolitan region is located between latitudes 39°26′ N and 41°03′ N and longitudes 115°25′ E and 117°30′ E, and has an area of 16 808 km². The average annual temperature is 12.7 °C, and the average precipitation is 539.5 mm/yr, as recorded from 1978 to 2008. The population is 17 million and the population density is 1,033 people per km². The population density along the urban-to-suburban gradient varies from 7629 people per km² in the urban area to 433 people per km² in the suburban area.²²

The Beijing metropolitan region includes 16 administrative subdivisions, which are county-level units governed directly by the municipality. There are six districts in the urban area, and eight districts and two counties in the suburban area. The whole region can be divided into urban, suburban, and rural areas. In suburban and rural areas, each county consists of a group of towns, and each town consists of a group of villages. We established five sampling areas along the urban–suburban–rural gradient, including in the central urban (UCA), urban village (UVA), central suburban county (CSA), rural town (RTA), and rural village areas (RVA).

RDS Sample Collection and Grain Size Fractionation. RDS samples were collected using a domestic vacuum cleaner (Philips FC8264) during September 2–10, 2009 following a dry weather period of about two weeks.

The RDS samples were collected from 97 RDS sampling sites in the urban, suburban, and rural areas of Beijing metropolitan region (Figure 1). Our sampling sites covered UCA, UVA, CSA, RTA, and RVA areas throughout the whole Beijing area. A smaller number of sampling sites for UVA (6 sites) were selected because all urban villages were distributed within the central urban area rather than the entire Beijing area. A larger number of CSA sampling sites (40 sites) were selected because of the possible high variation of RDS among widely spatial distributed central suburban county areas throughout Beijing. The characteristics of sampling sites are described in Table S1 in Supporting Information (SI). For each RDS sample collection, a variable area was vacuumed from the central road marking to the curb. The sampling area size was measured with a ruler. The RDS sample at each sampling site was measured with an electronic weighing scale. Sample mass collected was between 0.8 and 1.5 kg. Samples were sorted into grain size fractions of <44, 44–62, 62–105, 105–149, 149–250, 250–450, 450–1000, and >1000 μm using polyester sieves. More details about the RDS sample collection and grain size fractionation described in SI.

Estimation of Heavy Metal Loads in RDS. To determine the contribution of particles with different grain sizes to the overall contamination of the RDS, we calculated the pollutant load percentage for each individual RDS sample. The grain size

fraction load (GSF_{Load}) was calculated using the following eq 1:

$$\text{GSF}_{\text{Load}} = \frac{C_i \times \text{GS}_i}{\sum_{i=1}^m C_i \times \text{GS}_i} \quad (1)$$

where C_i is the heavy metal concentration associated with a RDS sample for a given grain size; GS_i is the percentage by mass of the size fraction i in the total sample; and m is the number of grain size fractions.

Experimental Design of RDS Runoff by Rainfall Simulation. There are many important factors that impact the potential contribution of RDS to surface runoff and runoff pollution. In the RDS runoff experiment, we focused on the RDS particle size, particle size composition, and rainfall intensity. The RDS percentage contribution for different particle sizes could be calculated by the combination of each particle size percentage. A specially designed rainfall simulation and small impervious surface plots were applied to estimate the removal rate of RDS in surface runoff. The experimental design was based on Herngren et al.²³ and Egodawatta et al.⁷ An area of 1.5 m × 2.0 m for the RDS washed off the surface was chosen so that optimum rainfall uniformity was achieved. The selected road for RDS washoff experimental plot was paved with asphalt two years ago, and its roughness and slope were 0.625 mm and 2.36°, respectively. Seven size fractions (fraction >1000 μm was discarded) at three levels of RDS mass per unit area were washed off with a rainfall intensity of 46.7 mm/h. Five other rainfall intensities were also simulated for the middle level of RDS mass per unit area. The rainfall duration was one hour, and surface runoff samples were collected manually at 2-min intervals during the first 10 min, at 5-min intervals during the second and third 10 min, and at 10-min intervals thereafter until there was no more surface runoff. In total, 56 different rainfall events were carried out with the five rainfall intensities (range 12.87–101.37 mm/h) and three levels of RDS mass per unit area (see more details about the experimental design of RDS surface runoff by rainfall simulation in SI).

Estimation of the Percentage of RDS in Surface Runoff. The surface runoff samples were collected and their volumes were measured at set intervals for each simulated rainfall event. Each size fraction of RDS in the runoff from the impervious surface was expressed as percentage of the total RDS mass in the runoff with respect to the initial total mass of RDS on the surface. This was calculated by the following equation:

$$\text{Fw} (\%) = \frac{M_{\text{Fw}}}{M_{\text{initial}}} \times 100\% = \frac{\int_0^1 C(t) \times Q(t) dt}{M_{\text{initial}}} \times 100\% \quad (2)$$

where Fw is the percentage of each RDS size fraction washed off the surface (%); M_{Fw} is the mass of the size fraction washed off the surface over the entire rain event (mg); M_{initial} is the initial mass of RDS with a corresponding grain size on the surface (mg); $C(t)$ is the mass of RDS with a corresponding grain size in the surface runoff water (mg/L) at each sampling time; and $Q(t)$ is surface runoff flow rate at each sampling time (m³/min).

Potential Heavy Metal Contribution in Surface Runoff. The potential contribution of pollution washed off the surface to contamination of the runoff was determined by the

Figure 1. Study area and sampling sites of road-deposited sediments in Beijing metropolitan region.

following equation:

$$Pw = \sum_{i=1}^7 M_i \times C_i \times Fw_i \quad (3)$$

where, Pw is the potential pollution contribution to runoff per unit area ($\mu\text{g}/\text{m}^2$); M_i is the mass of RDS of one size fraction per unit area (mg/m^2); C_i is the mass of heavy metal in each size fraction of RDS (mg/kg); and Fw_i is the percentage of each RDS size fraction washed off the surface (%).

Analytical Methods. Analyses for five heavy metal elements (Cr, Cu, Ni, Pb, and Zn) were run for all RDS and surface runoff samples using standard methods.²⁴ RDS samples were digested with $\text{HF}-\text{HClO}_4$. (See details about the analytical methods in SI)

RESULTS

RDS Grain Size Distribution. The cumulative particle volume distribution curves of the RDS grain size from the main traffic

Figure 2. (a) Grain size distribution of road-deposited sediment, (b) mean mass percentage of road-deposited sediment versus grain size fraction, and (c) road-deposited sediment mass per unit area of different land uses at different administrative division levels versus grain size fraction. Values expressed as mean $\pm$ standard deviation. UCA = the central urban area; UVA = the urban village area; CSA = the central suburban county area; RTA = the rural town area; RVA = the rural village area.

road surface showed a similar pattern at each of the sites along the urban environmental gradient (Figure 2). The median diameter of RDS, d_{50} , from UCA and CSA ranged from 100 to 150 μm , while d_{50} from RTA and RVA ranged from 150 to 200 μm . The RDS particles from all impervious road surfaces were predominantly smaller particles ($<250 \mu\text{m}$, 65–78%). RDS particles with grain sizes of 62–105 μm were the most abundant among the eight size fractions; notably this size fraction accounted for $38 \pm 11\%$ and $32 \pm 7\%$ of the total RDS in UCA and CSA, respectively (Figure 2b). The three dominant mass fractions in the RDS particles were as follows: 62–105 μm , $30 \pm 10\%$; 149–250 μm , $21 \pm 5\%$; 250–450 μm , $14 \pm 4\%$. There was an obvious gradient in the RDS mass per unit area for all grain size fractions along the urban–suburban–rural gradient as follows: low (UCA, $21 \pm 24 \text{ g/m}^2$; CSA, $20 \pm 16 \text{ g/m}^2$), medium (RTA, $59 \pm 63 \text{ g/m}^2$), and high (UVA, $147 \pm 198 \text{ g/m}^2$; RVA, $147 \pm 112 \text{ g/m}^2$) mass per unit area (Figure 2c). UVA and RVA had the highest values of RDS mass per unit area among all the sites. Our on-site investigation indicated that these areas were seldom swept (0.2–0.3 times/d), had high proportion of bare soil, and had broken and rough road surfaces.

Heavy Metal Concentrations of RDS. Five metals (Cr, Cu, Ni, Pb, and Zn) were analyzed in each grain size fraction of the RDS (Figure 3 and Table S4). The smallest RDS particles ($<44 \mu\text{m}$) had the highest metal concentrations (mg/kg) as follows: Cr 78.4 ± 25.1 , Cu 62.5 ± 43.8 , Ni 28.4 ± 9.6 , Pb 54.4 ± 19.2 , and Zn 340.0 ± 146.6 . The lowest heavy metal concentrations occurred in these fractions with grain size $>250 \mu\text{m}$ in most areas. Generally, the smaller grain size fractions had higher heavy metal concentrations. The metal concentrations for each grain size fraction decreased along the urban gradient in the order UCA $>$ CSA $>$ RTA $>$ UVA $>$ RVA. Zn was the most abundant element in all size fractions of the RDS in all areas (Figure 3).

Heavy Metal Loads of RDS along the Urban–Rural Gradient. The metal mass per unit area showed interesting patterns along the urban–suburban–rural gradient in Beijing. The metal mass per unit area for each RDS fraction in the UVA was the largest of all the areas at approximately 2 times that in the RVA, approximately 3–4 times that in the RTA, and approximately 6–8 times that in the UCA and CSA (Figure 4). The percentage contribution of each RDS size fraction to the heavy metal load was estimated using GSF_{Load} . The percentage contributions of the RDS size fractions to the mean GSF_{Load} of all the metals was in the following order (Table S5 in SI): 62–105 μm ($28.3 \pm 8.3\%$) $>$ 149–250 μm ($22.3 \pm 3.9\%$) $>$ 105–149 μm ($14.5 \pm 2.5\%$) $>$ 250–450 μm ($11.5 \pm 3.0\%$) $>$ 44–62 μm ($9.0 \pm 2.9\%$) $>$ 450–1000 μm ($7.3 \pm 3.3\%$) $>$ 0–44 μm ($5.3 \pm 1.7\%$) $>$ 1000–2000 μm ($1.8 \pm 1.2\%$). Generally, heavy metals in the RDS were mainly in the particles $<250 \mu\text{m}$ in diameter (80%). RDS with a grain size of $<100 \mu\text{m}$ is commonly referred to as the grain size of suspended solids in runoff.²⁵ Consequently, the contribution of particles $<105 \mu\text{m}$ in diameter to the overall metal contamination of the RDS affects the potential emission to runoff pollution. The percentage contributions of the $<105 \mu\text{m}$ particles in the different areas to the mean GSF_{Load} for all metals was in the following order (area, percentage contribution): UCA, $49.9 \pm 4.1\%$ $>$ CSA, $44.8 \pm 2.2\%$ $>$ RVA, $41.7 \pm 4.0\%$ $>$ RTA, $39.1 \pm 3.4\%$ $>$ UVA, $37.4 \pm 3.5\%$.

Estimation of RDS and Heavy Metal Loads in Surface Runoff from Simulated Rainfall. An understanding of how the different RDS size fractions are washed off impervious surfaces is essential to estimate the contribution of RDS to urban runoff. The percentages of RDS (Fw) washed off the impervious surface for each grain size fraction were calculated using a rainfall simulation at three levels of RDS mass per unit area (Table S6 in SI). Fw was proportional to the rainfall intensity at the same spatial density of particles on the surface, and was independent of spatial density.²⁶ The smaller size fraction of RDS was rapidly washed from the pavement. The first flush phenomena occurred, and was more pronounced with the smaller particles than the larger particles.

The potential contribution of RDS washed off per unit area of impervious surface to surface runoff pollution is shown in Table S7 in SI. The order of the areas for potential pollution contribution per unit area of impervious surface for Cr and Ni (rainfall intensity 12.87–101.37 mm/h) and Pb and Zn (rainfall intensity 12.87–46.7 mm/h) was UVA $>$ RVA $>$ RTA $>$ UCA $>$ CSA. For Cu (rainfall intensity 12.87–101.37 mm/h) and Pb and Zn (rainfall intensity 46.7–101.37 mm/h) the order was UVA $>$ RVA $>$ UCA $>$ RTA $>$ CSA. The difference in the potential pollution contribution per unit area between RDS and the metals associated with it becomes larger with increasing rainfall intensity.

Figure 3. Heavy metal concentrations in RDS. Values expressed as mean $\pm$ standard deviation.

This could be attributed to the higher mobility and higher GSF_{Load} of the smaller particles compared to the larger particle. The potential pollution contribution per unit area varied along the urban–rural environment gradient and with rainfall intensity. The contribution increased with the increasing rainfall intensity. Overall, the proportion of potential contribution from RDS to runoff pollution of heavy metals was higher in UCA and CSA than in RTA, RVA, and UVA. The higher potential pollution contribution of heavy metals was attributed to higher proportion of smaller particles in RDS in UCA and CSA, which had higher metal concentration and was more easily washed off in runoff.

DISCUSSION

The urban–rural environment gradient has an important influence on the quantity and grain size distribution of RDS. Anthropogenic sources such as industrial processes, vehicle emissions, and tire and road surface wear have been identified as dominant sources of RDS and the heavy metals associated with them.⁴ The urban–rural gradient affects the spatial distribution and quantity of RDS from anthropogenic sources.^{5,6} Different types of pollution sources produce RDS particles with a range of diameters.⁷ Kobringer and Geinopolos (1984) reported that tire wear produces particles with an average diameter of 20 μm .²⁷

Figure 4. Mass of heavy metal per unit area on different impervious surfaces. Values expressed as mean $\pm$ standard deviation.

Habibi (1973) found that 57% of the Pb emitted in vehicle exhaust was associated with particles $>9 \mu\text{m}$ in diameter.²⁸ It is well-known that the grain size distribution of RDS is an important factor in determining the mobility of particles and their associated pollutant concentrations.^{29,30} In Beijing, the most important source of Cu, Pb, Ni, and Zn in RDS is a combination of traffic, gas consumption for domestic cooking and heating, and some small factories; and the Cr source in RDS is construction materials such as concrete and mortar.³¹ In addition, the road texture and street cleaning method and sweeping frequency along the urban–rural gradient also affected the RDS load and grain size of RDS.³² In our study, the urban–rural gradient affected the proportion of RDS present as smaller size particles ($<105 \mu\text{m}$) and the corresponding heavy metal concentration in this size fraction. Along the urban–rural gradient, UCS and CSA had low mass of RDS ($21 \pm 24 \text{ g/m}^2$ and $20 \pm 16 \text{ g/m}^2$, respectively) but higher proportion of smaller size particles of RDS because of a combination effect of higher population density, higher traffic density, higher gas consumption for cooking and heating, but smooth road surface, more efficient mechanical sweeping method, and higher sweeping

frequency (2–3 times/d) (Table S1). In RTA, RVA, and UVA, higher proportion of bare soil, hand-sweeping method, lower sweeping frequency (0.2–2 times/d), and relatively rough road surface (Table S1) were important factors causing more RDS accumulation (59 ± 63 , 147 ± 112 , and $147 \pm 198 \text{ g/m}^2$ RDS for RTA, RVA, and UVA, respectively) and higher proportion of coarse particles of RDS. Therefore it is helpful to mitigate RDS pollution through improvement of existing cleaning methods (including cleaning frequency) and road surface condition.

Rainfall simulations for different size RDS particles in surface runoff can be used to provide insight into how the urban gradient affects the potential washoff load and ratio of heavy metals associated with RDS. In the present study, the amount and proportion of RDS particles with each grain size varied along the urban–rural gradient. It is well-known that storm runoff selectively transports RDS of particular grain sizes, and the percentage of RDS of each size fraction in the surface runoff is different.³³ To assess and compare the effect of the urban gradient on pollutant concentrations in surface runoff, the mobility of each size fraction needs to be known. The percentage of each size fraction of RDS washed off the surface was determined using a rainfall simulation.

This was then used to estimate the potential pollution contribution of surface runoff per unit area along the urban–rural gradient. In our previous field research, the total suspended solids (TSS) in surface runoff accounted for about 20% of the RDS based on two rainfall events,¹⁶ and the potential pollution of surface runoff calculated in the present study was similar. The potential pollution of surface runoff by each RSD size fraction per unit area of impervious surface provides a new method for estimating large-scale urban runoff pollutant loads. However, there are still many deficiencies in our rainfall simulation calculations because they are made without regard to the distances the particles travel, and with only one slope and roughness for the road surface.

■ ASSOCIATED CONTENT

S Supporting Information. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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■ ACKNOWLEDGMENT

We thank Xiaoyan He, Xiaoxue Wang, Huiliang Wang, Yang Ouyang, Xinzhong Du, Wenzan Li, Shuxia He, Yuguo Han, Fei Mo, Hongtao Guo, Di Tian, and Lina Wang for their help in collecting road-deposited sediment and in the rainfall simulation experiments. Funding was supported by the One Hundred Talents Program of The Chinese Academy of Sciences, the CAS/SAFEA International Partnership Program for Creative Research Teams of “Ecosystem Processes and Services” (KZCX2-YW-T13), and the State Key Laboratory of Urban and Regional Ecology (Grant SKLURE2008-1-05).

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