Characterization of heavy metal desorption from road-deposited sediment under acid rain scenarios

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ABSTRACT

Road-deposited sediments (RDS) on urban impervious surfaces are important carriers of heavy metals. Dissolved heavy metals that come from RDS influenced by acid rain, are more harmful to urban receiving water than particulate parts. RDS and its associated heavy metals were investigated at typical functional areas, including industrial, commercial and residential sites, in Guangdong, Southern China, which was an acid rain sensitive area. Total and dissolved heavy metals in five particle size fractions were analyzed using a shaking method under acid rain scenarios. Investigated heavy metals showed no difference in the proportion of dissolved fraction in the solution under different acid rain pHs above 3.0, regardless of land use. Dissolved loading of heavy metals related to organic carbon content were different in runoff from main traffic roads of three land use types. Coarse particles (>150 μm) that could be efficiently removed by conventional street sweepers, accounted for 55.1%–47.1% of the total dissolved metal loading in runoff with pH 3.0–5.6. The obtained findings provided a significant scientific basis to understand heavy metal release and influence of RDS grain-size distribution and land use in dissolved heavy metal pollution affected by acid rain.

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Introduction

Road-deposited sediments (RDS) are carriers for potentially toxic pollutants such as heavy metals (Egodawatta et al., 2013). During rainfall, heavy metals absorbed on RDS will be washed into the urban receiving water bodies with runoff, where they may have a negative effect on water quality (Brown and Peake, 2006). During acid raining, desorption of heavy metal will increase in runoff (Sansalone and Ying, 2008). Therefore, a number of research studies have focused on RDS based heavy metals. Heavy metals exist in the form of the “particulate” (solid phase) and “dissolved” (aqueous phase) fractions in the runoff, with the aqueous phase fraction having a significant impact on waterway’s health (Dean et al., 2005). Many factors, such as particle size (Zhao et al., 2009), specific surface area (Liu et al., 2009; Vdović et al., 1991) and solid–liquid interface ion exchange (Gunawardana et al., 2012), facilitate heavy metals to exchange from the particle-adsorbed fraction to the dissolved fraction. Since most of the dissolution of heavy metals come from the exchangeable and carbonate bound, pH of acid rain may play a very prominent role in enhancing desorption (Fang and Qing, 2008; Jegadeesan et al., 2008; Kanti Sen and Khilar, 2006; Kraepiel

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et al., 1999; Qing et al., 1998; Van der Sloot, 2002). Consequently, RDS affected by acid rain may increase the solubility of toxic heavy metals, causing a serious issue for urban storm water quality (Morrison et al., 1988).

Previous studies primarily focused on the description and analysis of heavy metals behaviors from different carriers, including heavy metal adsorption and desorption, especially the latter (Dijkstra et al., 2004; Kaschl et al., 2002; Weng et al., 1994; Yüksel and Alpaslan, 2001). For the adsorption of heavy metals, the mostly focused factors include particle size, organic matter, Fe/Al/Mn oxides, minerals, heavy metal species, charge carrier surface properties, etc. (Gunawardana et al., 2014, 2015). Desorption of heavy metals mainly includes static and dynamic studies. For static studies, carriers are static during rainfall, while they are moving during raining for dynamic studies. Abollino et al. (2003) found that the low pH of the desorption solution led to an increase in the concentration of dissolved heavy metals, and pH had different influences on different metals through the static desorption of Na-montmorillonite. Similarly, Dijkstra et al. (2004) also proved that static desorption of heavy metals (Cd, Cu, Ni, Zn and Pb) from soil showed a V-shaped feature, indicating that acidic and alkaline conditions promoted desorption, while neutral condition did not conducive to heavy metal desorption. Through investigating static leaching of fly ash, Gitarì et al. (2009) found pH determined the charge properties of fly ash surface and the ionization degree/existing form of the elements. Therefore, less desorption of heavy metal occurred under high pH conditions, while a significant increase in the amount of desorption under low pH conditions, especially for natural heavy metals (such as Fe and Mn). Similarly, pH (from 3.50 to 9.60) did play a key role in solubility of heavy metal from construction/demolition waste, and for these materials, most metals existed a secondary dissolution at pH 5.3 (Galvín et al., 2012). Dynamic desorption of copper and zinc showed that the effect of acid rain erosion could be divided into two phases: initial flushing stage and balanced steady stage (He et al., 2001). Acidic rain pH played a catalytic role in desorption of heavy metals during these two phases. The above studies showed that increasing acidity promoted desorption of heavy metals, while few studies have focused on pH, especially under dynamic conditions of strong acid rain, effect on desorption of heavy metals from the RDS. Therefore, to make effective control strategies, the in depth analysis of the desorption process under natural conditions and the important factors affecting desorption of heavy metals from RDS are needed.

The aim of the current study was to evaluate desorption of widely concerned heavy metals in polluted RDS at different functional areas under simulated acid rain with a range of pH values. Given the toxicity to the environment, Cr, Mn, Cu, Ni, Zn and Pb were selected, while Fe and Al were excluded as they are common metal elements in soils and present as metal oxides showing low toxicity. Understanding desorption and migration characteristics of the selected metals could help on undertaking effective management to improve urban water quality (Galvín et al., 2012). Mn representing the natural source, would mostly originate from soil sources (Gunawardana et al., 2012), while other five metals originate primarily from human activities as the anthropogenic source (Herngren et al., 2006; Sartor et al., 1974; Sutherland, 2003). As any dissolved heavy metals from RDS will flow via runoff into the river, quantifying total dissolved heavy metals into the river provides a scientific basis to control and design reasonable measures to deal with the heavy metal pollution. The study provides some fundamental knowledge in relation to metals desorption from RDS, guidance to control pollution of dissolved metals.

1. Materials and methods

1.1. Study sites

Study sites (Fig. 1) are located at Shenzhen, Guangdong Province, China, which was seriously subjected to acid rain due to the wide use of sulfur-generated fuel such as coal (Dianwu et al., 1988). Shenzhen is a typical megacity in South China (the area of about 2000 km² and a population of over 11 million) and is a subtropical zone with abundant annual rainfall (ranging from 1400 to 2000 mm). An urban catchment named Dakan (with a coverage of 1.3 km² and a population of 35,000), located at the northern part of Shenzhen, was selected as the study site since it encompassed typical urban land uses, namely industrial, commercial and residential areas (the area of road were 51,200 m², 12,100 m² and 31,800 m², respectively). In the study area, roads are primarily paved with asphalt and runoff water finally entered the Dakan River through the catchment rainfall drainage pipe system.

1.2. Build-up sample collection and laboratory analysis

Collection of RDS samples on the main road surfaces of the three functional areas, was carried out using a domestic vacuum cleaner by the dry method. Each sampling area from three to six plots with each plot having an area of 1.5 m², was carefully vacuumed three times with the aid of a fine brush attached to the vacuum nozzle to ensure that as much solids and particle size distribution were tested. Samples were air-dried for 5 days, followed by a dry sieving and separation. The sample was divided into five groups with size ranges of <75, 75–150, 150–300 and >300 μm (Liu et al., 2015), respectively, and then stored in the closed polyethylene bags. It is noteworthy that some small particles were likely to attach to large particles since a large particle as a whole and referred their combination as the “large particle”. For each build-up sample, organic carbon content (TOC), total RDS, RDS load on the five particle sizes and particle size distribution were tested. Samples were sprinkled onto plates, coated by Au and characterized by Scanning Electron Microscope (SEM) (S4800) to observe erosion affected by acid rain on the surface of particles.
1.3. Simulated rain and batch desorption experiments

At the sampling sites, $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ in the acid rain were 50–26 and 38–39 $\mu$mol/L from 2006 to 2012, respectively, while the pH of rainfall varied from 3.18 to 8.73 (Chen, 2014). Simulated acid rain was designed according to main ion composition and pH of the local rain water. Synthetic acid rain with pH of 3.0, 3.5, 4.0, 4.5, and 5.6 was prepared from a stock $\text{H}_2\text{SO}_4$–HNO$_3$ solution mixture (1:1.3, V/V), and pH 7.0 and 8.5 were selected to compare with the acid rain. The concentrations of $\text{K}^+$, $\text{Na}^+$, $\text{Ca}^{2+}$, $\text{Mg}^{2+}$, $\text{NH}_4^+$, $\text{Cl}^-$, $\text{SO}_4^{2-}$, $\text{NO}_3^-$, and $\text{F}^-$ were 6, 30, 29, 5, 41, 30, 38, 38 and 3 $\mu$mol/L, respectively.

In order to evaluate the effect of acid rain and RDS particle size on heavy metal desorption of dynamic desorption experiments of RDS, five types of particle sizes were tested in water with 7 pH values. For desorption of heavy metals from RDS, samples were placed in 100 mL Erlenmeyer flask (1 g solid particles with 50 mL of solution) and shaken at 240 r/min at 25°C to simulate rainfall runoff (Pitcher et al., 2004). According to local drainage system, the time that runoff went into the surrounding river through pipe system was generally less than 2 hr. So, the experiments were applied for 2 hr to simulate the period of runoff. The pH of solution was measured every 5 min.

Due to random sampling or incomplete digestion, the mean value was used to analyze the percentage of dissolution. Each solution in flask should ultimately analyze the dissolved metal concentrations (named $C_{Di}$) after filtration through 0.45 μm nitrocellulose filter (Waterman). Total metal concentrations (named $C_{Ti}$) after the samples had been digested using the nitric acid digestion procedure according to Standard Methods for the examination of Water and Wastewater (APHA, 2005). The concentrations of Cr, Mn, Cu, Ni, Zn, and Pb were determined using an Elan 6000 inductively coupled plasma-optical emission spectroscopy (PerkinElmer, Waltham, MA, USA). As a part of quality control, certified geochemical soil reference materials (GBW07430-GSS16) were used in this research for QA/QC purposes. For the analysis of heavy metals, recoveries varied but all fell within the range of 75.6%–105.8%, which is within the acceptable recovery range of 75%–120% (Herngren et al., 2006). The precision was nearly 6% with the confidence level of 95%.

Each sample was tested for $C_D$ and $C_T$ of heavy metals. Desorption of heavy metal from each size fraction under different pH values was expressed as the percentage to the total heavy metal, which was calculated by the following equation (Zhao and Li, 2013):

$$DR_i(\%) = \frac{CD_i}{CT_i} \times 100\%$$

where $DR_i$ (Dissolved Ratio) is the percentage of dissolved heavy metal in the RDS particle with a grain-size fraction of i (%); $CD_i$ (mg/L) is the dissolved metal concentration of the size fraction i in runoff (sample filtered through 0.45 μm nitrocellulose filter); $C_{Ti}$ (mg/L) is the total (particulate and dissolved) metal concentration of the size fraction i in runoff.

1.4. Dissolved heavy metal contribution to river

In order to determine the contamination degree of dissolved metals at different rain pH values, it was necessary to obtain loading of dissolved metals that could go into river in the whole study area. This was calculated by the following equations:

$$D_{Li} = \text{RDS}_{Li} \times \frac{C_{Di}}{SS}$$

$$TD_L = D_L(\text{Pb}) + D_L(\text{Cr}) + D_L(\text{Mn}) + D_L(\text{Cu}) + D_L(\text{Zn}) + D_L(\text{Ni})$$
Metal concentrations were in the 75–150 μm fraction, was apparent at three sites, with the exception of smaller grain-size fractions had higher heavy metal concentrations, and would affect the quality of receiving water, especially during acid rain seasons. The importance of fine particles in the adsorption of metals, and genic heavy metals from the runoff deposits was observed for all metals combined (named TDL); TDL values for all metals combined with land uses (named UTDL).

Table 1 - Road-deposited sediments mass per unit area (loading) of three land uses in each size fraction.

<table>
<thead>
<tr>
<th>Particle size (μm)</th>
<th>Industrial (g/m²)</th>
<th>Commercial (g/m²)</th>
<th>Residential (g/m²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt;300</td>
<td>10.17</td>
<td>11.21</td>
<td>7.54</td>
</tr>
<tr>
<td>150–300</td>
<td>13.10</td>
<td>10.84</td>
<td>5.93</td>
</tr>
<tr>
<td>100–150</td>
<td>7.96</td>
<td>5.60</td>
<td>4.81</td>
</tr>
<tr>
<td>75–100</td>
<td>5.72</td>
<td>3.49</td>
<td>2.78</td>
</tr>
<tr>
<td>&lt;75</td>
<td>27.23</td>
<td>12.43</td>
<td>12.95</td>
</tr>
<tr>
<td>Total</td>
<td>64.19</td>
<td>43.37</td>
<td>34.02</td>
</tr>
</tbody>
</table>

2.2. Effluent final pH values after heavy metal desorption process

H⁺ ion consumption was an important factor affecting the solution pH (Wang et al., 2009). From acid rain pH change, solution pH reached steady state after 2 hr. In order to identify the effects of H⁺ ion consumption, the final pH values as a function of acid rain were investigated in Fig. 4. Land use, particle size and initial pH were important factors affecting the final pH in runoff.

Land use played an important role in the final runoff pH. The order of areas for final pH for the same particle size was Industrial > Residential > Commercial. The difference in the final pH among three functional areas showed that industrial RDS were easily eroded by acids. The particle size distribution also contributed to the final pH. The final pH increased with the decreasing particle size, which could be attributed to the higher alkaline materials of smaller particles compared to the larger particles. As to the initial pH, the solution of initial pH 3.0 (red rectangle data) remained the lowest final pH, while other initial pH events got the similar final pH between each other. It implied that there was no significant difference in pollutants desorption from RDS, when the initial pH was higher than 4.0.

2.3. Dissolved Ratio (DR) of heavy metals from RDS

Fig. 5 shows DR of heavy metals based on five particle size fraction from three functional areas. Overall, the industrial RDS had the highest DR to most anthropogenic heavy metals (Pb, Cr, Ni and Zn), and the commercial RDS had the lowest, but the difference was not obvious. The TOC content of the particles was in the following sequence: commercial area > residential area > industrial area in Fig. 6, corresponding to order of heavy metal DR in three functional areas. This showed that desorption of heavy metals might be correlated with TOC. DR would reflect the contribution of dissolved heavy metals pollution. This was a serious concern for industrial RDS, which had higher risks to the environment.

No great difference in the desorption of most anthropogenic heavy metals from the runoff deposits was observed for a certain particle size under different initial pHs. Zhang and Wang (2009) investigated the distributions of heavy metals in the four defined solid fractions, including acid-extractable, reducible, oxidizable and residual fractions, and found that Mn and Zn had the highest percentage of acid-extractable fraction, while Cu, Pb, Ni, and Cr had relatively low proportion. So, Mn and Zn were more sensitive to acidity. The DR of Mn and Zn was highest at the initial pH 3.0 (red arrows in Fig. 5), but there were no much difference in DR when initial pH > 3.0, indicating that although acid-extractable fraction had the high distribution in Mn and Zn, only the difference was made by the extreme acidity (pH 3.0). It was also proved by the final pH which was the same values from the initial pH > 3.0.

At the same initial pH for different particle sizes, the DR was generally higher for large particle size fraction than that for small particle size fraction. It was also noteworthy that the DR was more variable with pH values for large particles while less for small particles (such as <75 μm). These phenomena could be attributed to the particle surface characteristics.
Compared to large particles, small particles hold heavy metals tightly due to the relatively uneven and rough surfaces. This led to the difficulty of heavy metals to be desorbed from small particles. This was also supported by SEM images shown in Fig. 3a1 and a2, where small particle surfaces were rougher with a number of grooves than large particles, irrespective of pH conditions. Gunawardana et al. (2015) also found that heavy metals created more stable bonds with fine solids than those adsorbed to the coarse solids particles, resulting in that desorption from fine particles was relatively low compared to the coarse particles. Organic matters could enhance adsorption by providing binding sites for cation adsorption (Filep et al., 1999). Furthermore, organic matter content in solids provided substantial binding sites for surface reactive pollutants, including heavy metals. The small size range particles showed the relatively higher TOC irrespective of location or land use (Fig. 6). Although high TOC content was found in the large particles at three sites, the TOC content seemed to have a less effect on the heavy metal adsorption in large particle fractions (Herngren et al., 2006). So, coarse solids had the higher desorption ratio of heavy metals.

Fig. 2 - Heavy metal concentration in each size fraction.

<table>
<thead>
<tr>
<th>Grain size fraction (μm)</th>
<th>Pb (mg/kg)</th>
<th>Cu (mg/kg)</th>
<th>Mn (mg/kg)</th>
<th>Ni (mg/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>&gt;300</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>150-300</td>
<td></td>
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<tr>
<td>100-150</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>75-100</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>≤75</td>
<td></td>
<td></td>
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</tbody>
</table>
2.4. Heavy metal dissolved loading from RDS in runoff

Calculation of dissolved heavy metal loadings of RDS affected by acid rain dissolved heavy metal loadings from RDS affected by acid rain, could provide necessary parameters for the design of storm water treatment facilities. Dissolved elements which were detached from the carrier, could be readily moved with runoff. As shown in Fig. 7, TDL was calculated in each particle size range and showed different performances in different land uses. The desorption of heavy metals in most size ranges was highest at commercial area, which was due to its heaviest accumulated metal elements in dry period. The highest TDL was found in the <75 μm size range in industrial and residential area, while the highest TDL was found in the 150–300 μm in commercial area. The difference in metal pollutant loading between coarse particles and fine particles was caused by DR and mass distribution of RDS particles in each grain size.

However, the influence of pH to TDL of metals would be limited in runoff. The TTDL values decreased from 0.376 to 0.341 mg/m² in the industrial area, from 0.721 to 0.267 mg/m² in the commercial area and from 0.149 to 0.104 mg/m² in the residential area as the initial rain pH from 3.0 to 4.0, respectively. But there was no significant influence of pH on total desorption of metals with the initial pH from 4.0 to 8.5. Although commercial area had higher TDL at pH 3.0 (extreme condition), industrial area has higher TDL at pH from 4.0 to 8.5 (normal condition). Therefore, considering the normal condition, appropriately controlling acid rain could relatively reduce the ecological risk posed by dissolved heavy metals, especially in the case of industrial areas.

2.5. Impacts of particle sizes on dissolved pollution contribution to river

Heavy metals accumulated on impervious surfaces from dry deposition could be washed off during a rainfall-runoff event, transferring a pollutant load eventually into river (Sansalone et al., 1998). According to previous studies of RDS transport in runoff, RDS on road were coarser than the RDS in wash-off, and 80%–100% of downstream RDS was finer than 100 μm (suspended) (Drapper et al., 2000; Ying and Sansalone, 2010). The suspended solids in runoff water accounted for less than 17% of the RDS, while the particulate heavy metal loads in the...
urban runoff water accounted for less than 7% (Zhao et al., 2010). Many runoff events removed only a small proportion of surface pollutant loads and particle matter to receiving water (Vaze and Chiew, 2003a, 2003b), because most particles could not travel the long distance between the RDS source and the target water bodies. All those studies demonstrated that dissolved pollution was more important than particulate pollution in most runoff events. Fig. 8 shows the order of UTDL of heavy metals in each grain size compared in the cases of initial pH = 3.0, 4.0 and 5.6. Specifically, (>300) μm (0.383 mg/m²) > (<75) μm (0.327 mg/m²) > (100–300) μm (0.180 mg/m²) > (>300) μm (0.156 mg/m²) > (100–150) μm (0.096 mg/m²) > (75–100) μm (0.054 mg/m²) at initial pH 4.0 and (<75) μm (0.226 mg/m²) > (150–300) μm (0.175 mg/m²) > (>300) μm (0.151 mg/m²) > (100–150) μm (0.090 mg/m²) > (75–100) μm (0.051 mg/m²) at initial pH 5.6.

In study area, the general rain pH values were 4.0–5.6 and contribution of each size fraction was similar. Decrease of dry solid particles was an effective mitigation measure to remove dissolved elements. Previous studies

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Fig. 5 – Impacts of pH and grain size on D_R of heavy metals from industrial area, commercial area and residential area.
reported that there was an interrelationship between sweeper efficiency and particle size. So the street sweeper efficiency decreased with decreasing grain sizes (Zhao et al., 2010). It was difficult to remove 80% of metal load by conventional street sweepers because it was stored in the <250 μm fraction (Pitt, 1979; Sartor et al., 1974; Zhao et al., 2010). However, our results showed that a particle size >150 μm contributed 55.1%, 47.1% and 47.1% (calculated from Fig. 8) of the total dissolved heavy metals in RDS at initial pH 3.0, 4.0 and 5.6, respectively, which indicated that particles >150 μm comprised the critical size fraction that contributed to heavy metal dissolved pollution caused by acid rain. Combined with the previous research (Sartor et al., 1974), Fig. 8 also shows the potential removal UTDL in each grain-size fraction and the sweeper efficiency of conventional street sweepers for UTDL was about 43.0%, 40.3% and 40.2% (proportion of black bar in Fig. 8), in which the removal of coarser fraction accounted for 26.3%, 31.9% and 32.5% at initial pH 3.0, 4.0 and 5.6, respectively. This suggested that conventional street cleaning efforts could potentially remove a relatively large percentage of dissolved heavy metals in acid rain sensitive areas of China. But we also need more effective methods to remove RDS, especially for fine particles (<75 μm) that was the most polluted at each site in the antecedent dry period and had high dissolved pollution.

Fig. 6 – Concentration of TOC versus grain-size fraction for industrial, residential and commercial land uses. TOC: organic carbon content.

Fig. 7 – TD₅ₐ of heavy metals in runoff of main road in three typical land uses. TD₅ₐ: D₅ₐ for all metals combined.

Fig. 8 – UTDL and removal efficiency of conventional street sweepers of heavy metals under acid rain of pH 3.0 (worst) and 4.0–5.6 (general) in study area. UTDL: TD₅ₐ values for all metals combined with land uses.
in runoff, from road surfaces in urban areas through improvement of road surface condition and existing cleaning methods (Herngren et al., 2006) (such as cleaning frequency and cleaning tools) in order to safeguard water quality.

The above results suggested that the distribution of RDS particle size was the key factor determining desorption of heavy metals. Coarse particle size of RDS with higher dissolved loadings was more hazardous to water bodies. In addition, land use types exerted an important influence on the pollutant desorption of RDS. Commercial RDS had higher DR, while industrial RDS had the higher DR. So they had higher CDR. But higher DR was directly related to desorption, which dissolved pollution. In a word, we need to strengthen the separated from RDS with low TOC content, resulting in erosion of acidic rain, and these elements were easily RDS from these metallic materials were affected by the extreme acidity (pH 3.0).

Therefore, it implied that many heavy metals in industrial RDS from these metallic materials were affected by the erosion of acidic rain, and these elements were easily separated from RDS with low TOC content, resulting in dissolved pollution. In a word, we need to strengthen the control of the industrial area to reduce desorption of heavy metals from RDS.

3. Conclusions

Based on the data analyses, the following conclusions were obtained.

(1) In urban areas, RDS contained high concentrations of heavy metals. Urban land use played an important role in accumulating the RDS and metal contents in RDS. The order of heavy metal loading of main road was commercial > industrial > residential.

(2) Most anthropogenic source heavy metals (Pb, Cr, Cu, Ni) showed no difference in the D90 under different pHs, regardless of land use. The D90 of Mn and Zn was highest at initial pH 3.0, but there were no much difference in D90 of them when initial pH > 3.0, only the difference was made by the extreme acidity (pH 3.0).

(3) Industrial RDS had the highest D90 in most particle size fractions, and industrial area had higher risks posed to the environment than other typical areas.

(4) Particle sizes played an important role in desorption of heavy metals. The highest TD90 was found in the <75 μm size range in industrial and residential area, while the highest TD90 was found in the 150–300 μm in commercial area.

(5) Contribution of each size fraction was similar in three typical land uses affected by general acid rain (pH 4.0–5.6). Our results showed that a particle size > 150 μm contributed more than 50% of the total dissolved heavy metals in RDS at pH from 4.0 to 5.6, which indicated that particles > 150 μm comprised the critical size fraction that contributed to heavy metal dissolved pollution caused by acid rain.

(6) The removal dissolved loading in each grain-size fraction and the sweeper efficiency of conventional street sweepers for dissolved pollution was high at rain pH from 3.0 to 5.6.

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