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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.

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; Yukselen 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 conduce to heavy metal desorption. Through investigating static leaching of fly ash, Gitari 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 km^2 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 km^2 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 m^2, 12,100 m^2 and 31,800 m^2, 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 m^2, was carefully vacuumed three times with the aid of a fine brush attached to the vacuum nozzle to ensure that as much solids as possible were collected, including industrial (3 × 1.5 m^2), commercial (3 × 1.5 m^2) and residential (6 × 1.5 m^2) in Fig. 1. The antecedent dry at sample collection time was 10 days, which ensured that heavy metal accumulation tends to reach at equilibrium. As noted by Wicke et al. (2012), the heavy metal build-up amounts leveled off after 6 antecedent dry days, while Chow et al. (2015) also noted that the build-up generally reached a maximum after 5 dry days. Adequate samples (200–300 g) were collected from each road surface.

The collected 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, 75–100, 100–150, 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 "dry-sieving" approach was used. In this case, it was considered the large particle as well as those small particles attached to the 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.

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