



Risk assessment of metals in road-deposited sediment along an urban–rural gradient

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ARTICLE INFO

Article history:

Received 1 November 2012

Received in revised form

6 December 2012

Accepted 13 December 2012

Keywords:

Road-deposited sediment

Metal pollution

Risk assessment methods

Urban–rural gradient

ABSTRACT

We applied the traditional risk assessment methods originally designed for soils and river sediments to evaluation of risk associated with metals in road-deposited sediment (RDS) along an urban–rural gradient that included central urban (UCA), urban village (UVA), central suburban county (CSA), rural town (RTA), and rural village (RVA) areas in the Beijing metropolitan region. A new indicator RI_{RDS} was developed which integrated the RDS characteristics of mobility, grain size and amount with the potential ecological risk index. The risk associated with metals in RDS in urban areas was generally higher than that in rural areas based on the assessment using traditional methods, but the risk was higher in urban and rural village areas than the areas with higher administration units based on the indicator RI_{RDS} . These findings implied that RDS characteristics variation with the urban–rural gradient must be considered in metal risk assessment and RDS washoff pollution control.

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1. Introduction

Road surfaces serve as sinks and sources of metals and other contaminants in urban environments (Deletic and Orr, 2009; Yuen et al., 2012). Elevated concentrations of metals are ubiquitous in road-deposited sediment (RDS) owing to a wide range of human activities including vehicle emissions, coal combustion, disintegration of vehicle brakes and tires, atmospheric deposition, road surface wear, municipal solid waste incineration, and residential heating (Andrews and Sutherland, 2004; Duzgoren-Aydin, 2007; Eriksson et al., 2007; Duong and Lee, 2009). RDS acts as a significant carrier of potentially toxic elements such as metals including chromium (Cr), copper (Cu), nickel (Ni), lead (Pb) and zinc (Zn) (Al-Khashman, 2004; Tian et al., 2009; Sutherland et al., 2012). RDS contaminated by metals is becoming an important threat because of the possible transmission of their pollutants to aquatic systems by urban runoff (Herngren et al., 2005; Kong et al., 2012). Excessive accumulation of metals in RDS also results in increased human exposure to metals due to their close proximity to anthropogenic activities (Christoforidis and Stamatis, 2009; Charlesworth et al., 2011). As point source pollution is reduced in China and many other countries, urban runoff with contaminated RDS has become an increasingly serious problem (Zhu et al., 2008; Zhao et al., 2010). Thus, a proper understanding of RDS contamination is crucial in urban ecosystem.

Good pollution assessment methods are powerful tools for processing, analyzing and conveying raw environmental information to decision makers, city planners, engineers and public health managers (Caeiro et al., 2005; Gong et al., 2008). In general, assessment of metal contamination of RDS is still in the initial stages. The earliest studies conducted to investigate metal contamination associated with RDS focused on comparison among land uses in one city or among different cities (Lau and Stenstrom, 2005; Lu et al., 2009). In recent years, a variety of methods have been introduced to investigate the metal risk associated with RDS posed to water, soil or river sediments, with traditional methods including the (1) geoaccumulation index (I_{geo}), (2) enrichment factor (EF), (3) Nemerow synthetic pollution index (PI_N) and (4) potential ecological risk index (RI) (Buccolieri et al., 2006; Cheng et al., 2007; Liu et al., 2008; Shi et al., 2010; Wei and Yang, 2010). The applicability of these traditional assessment methods to the risks associated with metals in RDS is still unclear. These methods were originally designed for the water, soil and river sediment, and only with consideration of metal concentrations or toxicology risks, without coupling the pollutant amounts and mobility to the grain size of the RDS. Clearly there is a need to develop a new method for assessment of metal risk associated with RDS characteristics.

The urban–suburban–rural gradient pattern is common in the context of rapid urbanization in China, and strongly affects land use zoning, population density, the proportion of surfaces that are impervious, traffic density, energy consumption, street cleaning methods, and spatial distribution of manufacturers (Callender and Rice, 2000; Zhao et al., 2011). Previous studies indicated that the

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urban–rural gradient had an important effect on RDS amounts, grain size composition, metal types and concentrations associated with it (Rogge et al., 1993; Kupiainen et al., 2005; Kim and Sansalone, 2008; Thorpe and Harrison, 2008). Zhao et al. (2011) reported that metal concentrations decreased and quantities of metals per unit area increased from urban to rural areas along the urban–rural gradient in Beijing. Therefore, the urban–rural gradient could strongly influence the spatial distribution of the potential risk of metals associated with RDS.

In this study, we aimed to 1) compare and analyze the applicability of traditional assessment methods to metal risk associated with RDS; 2) develop a new metal risk assessment method with consideration of RDS characteristics; and 3) investigate the spatial pattern of metal risk associated with RDS along an urban–rural gradient.

2. Materials and methods

2.1. Study area and RDS sampling

The administrative division of China has historically consisted of several levels. There are currently five practical levels of local government: the province, prefecture, county, township, and village. The administrative division in China is divided by political boundaries and not defined by quantified criteria, which results in several categories according to the characteristic of urban–rural function. The patterns of division differ only slightly and are followed by most large cities across the country. The administrative division of the urban–rural gradient in China is as follows: central urban (UCA), central suburban county (CSA), rural town (RTA), and rural village (RVA) areas. In addition, urban village areas (UVA) consist of villages within large cities that still look like rural villages and lack urban infrastructure and services, but are primarily occupied by residents that work in cities and have an urban lifestyle (Deng and Huang, 2004). There were obvious differences in land use zoning, population density, proportions of impervious surfaces, traffic density, energy consumption, street cleaning methods, and industry along the urban–rural gradient. Further details regarding the characteristics of sampling sites along the urban–rural gradient are available elsewhere (Supporting Information; Zhao et al., 2011). To account for variation in RDS characteristics on roads of areas, the number of sampling sites varied depending on our field investigation. For example, we selected four main traffic roads as our sampling sites of the CSA for each of the ten suburban counties (Fig. 1). We collected the RDS from all the ten suburban counties so that we could assure our sampling sites cover all suburban counties. The total sampling site number at the CSA was 10 CSA areas \times 4 main traffic roads = 40 sampling sites. Based on our on-site investigation, we found that the characteristics of the UCA RDS had relatively lower variation. Eleven sampling sites were selected to represent the major roads of the UCA. Because of the smaller area of RTA and lower RDS variation of the RTA, RVA and UVA, we selected 20 (10 RTA \times 2 main traffic roads), 20 (10 RVA \times 2 main traffic roads) and 6 (3 UVA \times 2 main traffic roads) sampling sites, for RTA, RVA, and UVA, respectively.

We collected RDS samples (1) during a dry weather period of about 2 weeks; (2) at a time that was half-way between the last sweeping time and the next; (3) so that the collection of all RDS samples in the study were completed in as short a time period as possible (eight days in this study). RDS samples were collected using a domestic vacuum cleaner (Philips FC8264) from September 2–10, 2009. For each RDS sample collection, a variable area was vacuumed from the central road marking to the curb, after which the sampling area length and width was measured with a ruler. All RDS samples were air-dried for 7 days and then weighed using an electronic scale. The sample mass ranged from 0.8 to 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.

2.2. Analytical methods and quality control

Total metals (Cr, Cu, Ni, Pb and Zn) were measured after HF–HClO₄ digestion on a hotplate (Tessier et al., 1979). A quality control programme, including reagent blanks, replicate samples and standard reference material, was used to assess data precision and accuracy (Sutherland and Tolosa, 2000; Al-Khashman, 2004). All chemicals used for metal measurements were guaranteed reagent. All glassware, polyethylene labware and Teflon tubes used in the analyses of metals were washed with detergent, acid-soaked and then rinsed thoroughly with deionized water. GBW07401 (GSS-1) and GBW07402 (GSS-2), which are reference materials for soil certified by the General Administration of Quality Supervision, Inspection and Quarantine of the People's Republic of China (CRMs), were used for quality control during the digestion of RDS. The digests were stored at 4 °C prior to analysis. The concentrations of Cr, Cu, Ni, Pb and Zn were determined by inductively coupled plasma-optical emission spectrometry (ICP-OES) (Perkin–Elmer, USA). The

recoveries varied among metals, but all fell within the range of 75–95%. The precision was nearly 90% with a confidence level of 95%.

2.3. Traditional risk assessment methods originated from soils and sediments

Direct comparison, geoaccumulation index (I_{geo}), Nemerow synthetic pollution index (P_{IN}), and potential ecological risk index (RI) were used to assess the metals contamination of RDS with different grain sizes along the urban–rural gradient in Beijing (Table 1).

2.3.1. Geoaccumulation index (I_{geo})

Pollution levels of metal in RDS could be characterized by the I_{geo} value put forward by Müller (1969), which can be calculated by Eq. (1):

$$I_{\text{geo}} = \log_2 \left[\frac{C_{ij}}{1.5C_{ri}} \right] \quad (1)$$

where: C_{ij} is the measured concentration of the examined metal i in RDS with grain size j , and C_{ri} is the background concentration of metal i . The C_{ri} for Cr, Cu, Ni, Pb and Zn was set as 31.1, 19.7, 27.9, 25.1 and 59.6 mg/kg, respectively (Chen et al., 2004). Factor 1.5 was selected because of possible variations in background values for a given metal in the environment as well as very small anthropogenic influences. Müller (1969) proposed seven classes of the I_{geo} , which are given in Table 1.

2.3.2. Nemerow synthetic pollution index (P_{IN})

P_{IN} was applied to assess soil environmental quality in previous studies (Cheng et al., 2007). In the present study, this method was utilized to measure the degree of RDS environmental pollution as follows:

$$P_{\text{IN}} = \sqrt{\frac{\text{Max}P_{ij}^2 + \bar{P}_{ij}^2}{2}} = \sqrt{\frac{\left[\frac{C_{ij}}{C_{ri}} \right]_{\text{Max}}^2 + \left[\frac{1}{m} \sum_{i=1}^m \frac{C_{ij}}{C_{ri}} \right]^2}{2}} \quad (2)$$

where: $\text{Max}P_{ij}$ and \bar{P}_{ij} are the maximum and average value of the pollution indices of all metals, respectively, m is the number of metal species and C_{ij} and C_{ri} have the same meaning as in Eq. (1). Cheng et al. (2007) proposed five classes of P_{IN} , which are shown in Table 1.

2.3.3. Potential ecological risk index (RI)

RI, which represents the sensitivity of the biological community to toxic substance and illustrates the potential ecological risk caused by the overall contamination, was introduced to assess the contamination degree of RDS in previous studies. The equation used to calculate RI was as follows (Hakanson, 1980):

$$RI = \sum_{i=1}^m T_r^i \times \frac{C_{ij}}{C_{ri}} \quad (3)$$

where: RI is the sum of all five risk factors for metals in RDS, T_r^i is the metal toxic response factor according to Hakanson (1980), and the values for each element are in the order of Zn = 1 < Cr = 2 < Cu = Ni = Pb = 5. C_{ij} and C_{ri} have the same meaning as in Eq. (1). Hakanson (1980) proposed four classes of RI, which are shown in Table 1.

2.4. A new index for assessment of metal risk associated with RDS (RI_{RDS})

The new method for assessment of metal risk associated with RDS, which coupled concentration, toxic response factor, and the pollutant amounts and mobility, was primarily based on RI. The equations comprising the potential ecological risk index of RDS (RI_{RDS}) are as follows:

$$\begin{aligned} RI_{\text{RDS}} &= \sum_i^n \left(\sum_j^m RI_j \right) \times F_{\text{amount}} \times F_{\text{mobility}} \\ &= \sum_i^n \left(\sum_j^m RI_j \right) \times \left(\sum_j^m A \times P_i \times M_j \right) \\ &= \sum_j^m \sum_i^n \left(T_r^i \times \frac{C_{ij}}{C_{ri}} \times A \times P_j \times M_j \right) \end{aligned} \quad (4)$$

where: T_r^i has the same meaning as in Eq. (3) and C_{ij} and C_{ri} have the same meaning as in Eq. (1). A was the amount of RDS per unit area. The amounts of RDS per unit area varied with the site averaging from 26 to 220 g/m^2 (Sartor and Boyd, 1972) and were divided into six levels: 0–30 $\text{g}/\text{m}^2 = 1.00$; 31–60 $\text{g}/\text{m}^2 = 1.75$; 61–90 $\text{g}/\text{m}^2 = 2.50$; 91–140 $\text{g}/\text{m}^2 = 3.00$; 141–190 $\text{g}/\text{m}^2 = 3.50$; >190 $\text{g}/\text{m}^2 = 3.75$. P_j was the percentage of RDS with grain size j accounting for the total RDS mass and M_j is the grain size response factors for potential mobility in runoff (Zhao et al., 2011) (Table 2).

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