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Index models to evaluate the potential metal pollution contribution from washoff of road-deposited sediment

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ARTICLE INFO

Article history:

Received 26 September 2013

Received in revised form

19 March 2014

Accepted 6 April 2014

Available online 18 April 2014

Keywords:

Road-deposited sediment

Index model

Pollution load

Pollution strength

Urban diffuse pollution

ABSTRACT

The proper evaluation of pollutant strength and loads associated with road-deposited sediment (RDS) is crucial for controlling diffuse pollution in urban areas. A new index model, which combines source and transport factors and is called the RDS index, was developed using RDS characteristics (e.g., the amount, grain size, mobility, and metal concentrations) and used in a case study in the Beijing region. The observed and weighted RDS characteristics along an urban–rural gradient, which included central urban (UCA), urban village (UVA), central suburban county (CSA), rural town (RTA), and rural village (RVA) areas, were used to calculate the RDS index for the pollutant load ($RDS_{index,load}$) and the pollutant strength ($RDS_{index,strength}$). Our results demonstrated that the $RDS_{index,load}$ and $RDS_{index,strength}$ values both changed significantly along the urban–rural gradient. $RDS_{index,strength}$ increased along the urban–rural gradient and the $RDS_{index,load}$ value along the main roads decreased in the order $RVA > UCA > CSA > RTA$. The method offers a new way of assessing metal pollution in RDS and provides an important scientific basis for controlling pollution caused by RDS washoff.

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1. Introduction

As point sources of pollution in China and many other countries decrease, urban runoff containing contaminated road-deposited sediment (RDS) is becoming an increasingly serious problem (Zhu et al., 2008; Zhao et al., 2010). RDS from impervious surfaces is an important carrier of contaminants,

often containing metals at elevated concentrations (Aryal et al., 2009; Xiang et al., 2010). Large amounts of pollutants, such as nutrients, metals, and hydrocarbons, are usually transported in RDS washoff (Sartor and Boyd, 1972; Huber and Dickinson, 1988; Al-Khashman, 2007). Quantifying the relationship between RDS and washoff particles in urban runoff could provide a new method for estimating the pollution load that a waterway receives (Herngren et al.,

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2005; Zhao et al., 2011). A range of diffuse pollution models for urban areas have been proposed, including STORM, HSPF, DR3M-QUAL, SWMM, and SLAMM, and they have been used to estimate pollutant loads in runoff (Egodawatta et al., 2007; Wang et al., 2011). Despite the fact that the models mentioned above are widely used to estimate diffuse pollution sources in urban areas, they generally require substantial amounts of parameterization and calibration data (Shaw et al., 2006). Consequently, there is a need for improved runoff pollution estimates.

Identifying and prioritizing critical source areas could greatly improve the efficiency of RDS washoff pollution controls. Index models have been widely used for identifying diffuse critical source areas of phosphorus (P) on farms, and are less likely than other models to be constrained by a lack of input data (Heathwaite et al., 2005; Sharpley et al., 2008). Index models allow critical source areas to be identified by quantifying the relative pollution risk (e.g., as a probability) as opposed to the actual pollutant loading, because it is difficult to quantify transport factors (Buczko and Kuchenbuch, 2007; Buchanan et al., 2013). The index models mentioned above were initially devised for diffuse pollution in agricultural areas, and no index model has yet been specifically designed for diffuse pollution in urban areas that can be used to identify critical source areas of urban runoff. Characterizing RDS (e.g., the amount, grain sizes, associated metals, and mobility) can make it possible to identify critical source areas of urban runoff. A risk index (called RI_{RDS}) that combines RDS characteristics with a potential ecological risk index (RI) was developed by Zhao and Li (2013). The RI_{RDS} can be used to evaluate the amount of pollution present per unit area, but not the total pollution load. There is a clear need for the development of new simple but effective index models that use RDS characteristics to quantify or semi-quantify urban pollutant loadings.

The urban, suburban, and rural areas in and near large cities in China are generally divided into areas classed as urban districts, suburban county areas, rural townships, rural villages, and urban villages (Zhao et al., 2011). Previous studies have shown that RDS characteristics (including the amount of RDS present, its grain size distribution, metals associated with it, and its mobility) vary significantly along urban–rural gradients (Zhao et al., 2011). It is important to be able to evaluate the metal load associated with RDS correctly along urban–rural gradients, and developing such an ability will improve our understanding of the risks posed by heavy metal pollution in RDS being transported into urban water. The RDS characteristics mentioned above are critical to the assessment and evaluation of the role of RDS in metal pollution in urban runoff (Zhao and Li, 2013). However, there is still no index model available for identifying critical source areas of pollutants associated with RDS. Quantifying pollutant loadings will assist managers to focus remediation actions on decreasing RDS washoff into water bodies.

In this study, we aimed to 1) determine the source and transport factors for heavy metals in RDS using observed and weighted RDS characteristics in a multiplicative index, and 2) combine a number of functions for evaluating the pollution load and strength into one RDS index.

2. Materials and methods

2.1. Study area and RDS sampling

In general, big cities in China are divided by the government into urban, suburban, and rural areas, and the classifications used are urban district, suburban county, rural township, rural village, and urban village. We collected RDS samples from areas that were typical of each of these administrative divisions within the Beijing metropolitan region. The Beijing metropolitan region includes 16 administrative sub-divisions, which are county-level units that are governed directly by the municipality. There are six such districts in the urban area, and ten (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 chose five sampling areas along an urban–suburban–rural gradient, including a central urban area (UCA), an urban village area (UVA), a central suburban county area (CSA), a rural town area (RTA), and a rural village area (RVA). In general, the population density, traffic density, energy consumption, and frequency of road sweeping all decrease moving along the urban–rural gradient. Roads in the UCA and CSA generally have relatively smooth and undamaged impervious surfaces, and are regularly (daily) mechanically swept, whereas roads in the RTA, RVA, and UVA generally have very damaged surfaces, and are rarely swept.

RDS samples were collected using a domestic vacuum cleaner (Philips FC8264; Philips, Amsterdam, Netherlands) between 2 and 10 September 2009, following a period of about two weeks of dry weather. Three sampling sites were selected in each of the study areas. An unspecified area at each site was vacuumed from the central road marking to the curb until a reasonable amount of RDS was collected, then the size of the area sampled was measured with a ruler. Each RDS sample was weighed with an electronic balance, and sample masses of 0.8–1.5 kg were found to have been collected at each site. The mass of RDS per unit area was calculated by dividing the RDS mass collected by the size of the sampling area, and it ranged from 2 to 570 g/m² for all of the samples that were collected. The amount of RDS collected at a site was generally larger in the RTA, RVA, and UVA than in the UCA and CSA. The samples were separated into the grain size fractions <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

Metals were measured after the RDS samples had been digested with a mixture of HF and HClO₄ on a hotplate (Tessier et al., 1979). All solutions were stored at 4 °C until they were analyzed. The Cr, Cu, Ni, Pb, and Zn concentrations were determined using an Elan 6000 inductively coupled plasma-optical emission spectroscopy instrument (Perkin–Elmer, Waltham, MA, USA). Certified geochemical soil reference materials (CRMs) GSS-1 and GSS-2 were also analyzed to provide quality assurance and quality control (QA/QC) information. No RDS CRMs are available, but using soil CRMs has

been found to be an acceptable way of determining the quality of RDS analyses in previous studies (Sutherland, 2003; Kartal et al., 2006; Zhao et al., 2010). The recoveries of the five metals that were analyzed were 75–110% (75–95% for Cr, 90–108% for Cu, 90–104% for Ni, 90–110% for Pb, and 98–107% for Zn). The detection limits for Cr, Cu, Ni, and Zn ranged from 0.1 to 1 µg/L, and the detection limit for Pb ranged from 1 to 10 µg/L. Duplicate aliquots of 2% of the RDS samples were analyzed, and the metal concentrations found in the duplicates were always within ±10% of the mean concentration. Reagent blanks were included in each batch of samples that was analyzed.

2.3. Source and transport factors in the RDS indices

2.3.1. Source factors in the RDS indices

The amount of pollutants that is present in RDS is important because it determines the amount of pollutants in surface runoff. The source factors used in the RDS index models included the amount of RDS, the grain sizes, and the amounts and types of pollutants associated with the RDS. To obtain the best possible RDS parameter estimates, we studied a large number of sites (167 in total, 97 of which were in areas with busy main roads and 70 of which were in residential areas) along an urban–rural gradient. The mass of RDS per unit area varied widely, with a range of 2–570 g/m² and a mean of 70 g/m². The metal concentrations were measured in each RDS grain size fraction.

2.3.2. Transport factors in RDS indices

The particle mobility for each RDS grain size fraction was measured using simulated rainfall. A specially designed rainfall simulator and small impervious surface plots were used to estimate the RDS removal rates in surface runoff. The amount of each RDS size fraction in the runoff from the impervious surface was expressed as a percentage of the total mass of RDS in the runoff with respect to the initial total mass of RDS on the surface, calculated using Equation (1) (Zhao et al., 2011).

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

where F_w is the percentage of an 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 the size fraction on the surface (mg), $C(t)$ is the concentration of the size fraction in the surface runoff water (mg/L) at a particular sampling time t , and $Q(t)$ is the surface runoff flow rate at the sampling time (m³/min).

To obtain F_w , 35 different simulated washoff scenarios using seven RDS size fractions (<44, 44–62, 62–105, 105–149, 149–250, 250–450, and 450–1000 µm) and five simulated rainfall intensities (12.87, 32.70, 46.7, 82.67, and 101.37 mm/h) were each conducted for 1 h. F_w was used to calculate the amounts of RDS washoff for different particle sizes and to calculate the amounts of the heavy metals that were associated with the washed off particles. The F_w for each grain size particle was independent of the amount and size of the particles, but the F_w for the bulk RDS particles was dependent on

the grain size distribution (Shaw et al., 2006; Zhao et al., 2011). We used the washoff percentage for each RDS grain size to calculate the bulk RDS washoff percentage and amount for the corresponding sampling area, to properly represent the RDS washoff scenarios in our calculations. We used the F_{wi} values for each RDS size fraction to calculate the amount of RDS washoff that occurred for the RDS samples taken from each of the urban–rural gradient area types, i.e., UCA, UVA, CSA, RTA, and RVA. Further details of the experimental design are available elsewhere (Zhao et al., 2011).

2.3.3. Ranking the source and transport factor ratings

No standard methods are available from the literature for assessing RDS source and transport factors and to allow rankings to be allocated. We assigned weightings to the source and transport factors according to our in situ investigations into the RDS characteristics. The RDS mass per unit area was ranked into six levels (M_{weighted}), in which 0–30 g/m² was given a value of 1.00, 31–60 g/m² a value of 1.75, 61–90 g/m² a value of 2.50, 91–140 g/m² a value of 3.00, 141–190 g/m² a value of 3.50, and >190 g/m² a value of 3.75 (Zhao and Li, 2013). The RDS mass per unit area ranking levels were primarily based on the results of our in situ investigation into the RDS mass per unit area (using a total of 167 sites, 97 of which were from areas on main roads and 70 of which were in residential areas), because no standard values are available for dividing RDS masses per unit area into levels. We found that places that had higher RDS masses per unit area also had higher proportions of relatively coarse particles. We assigned comparatively low values to the higher RDS masses per unit area because coarser particles give lower levels of risk associated with their metal contents and the mobility of those metals (Zhao and Li, 2013). Metals associated with RDS were ranked by comparing the measured concentrations of the metal of interest with its background concentrations. The background heavy metal concentrations that were used were determined in 120 soil samples that did not suffer from heavy metal pollution, and these had been selected from a total of 803 soil samples (Chen et al., 2004).

Transport factors can be ranked according to the RDS washoff percentages for each grain size fraction (F_{wi}), as described by Zhao et al. (2011). The transport factors were incorporated into the RDS index by giving the lowest F_w value for a certain grain size fraction a value of 1 (see Table 1). Other F_w values for other grain size fractions were assigned by dividing the F_w value by the lowest F_w value (Zhao and Li, 2013). The transport factor ratings for each grain size fraction are listed in Table 1.

2.4. Calculating the RDS index

In this section, we will describe the general principles involved in calculating the RDS index. The RDS index was based on the phosphorus index that is used for modeling diffuse pollution in agricultural areas (Sharpley et al., 2008; Buchanan et al., 2013), but the RDS index was designed specifically to deal with the RDS characteristics found in diffuse pollution in urban areas.

The RDS index is used in this study not only to identify critical source areas by quantifying relative pollution risks, but

Table 1 – Transport factor ratings in RDS index for pollutant strength.

Transport factor	Grain size fraction of RDS (μm)						
	<44	44–62	62–105	105–149	149–250	250–450	450–1000
F _{wi,weighted}	17.0	10.0	4.5	4.3	2.9	1.5	1.0

also to quantify actual pollutant loadings. Both semi-quantitative and quantitative applications use the RDS characteristics and take source and transport factors into account. Semi-quantitative and quantitative pollutant loadings were calculated using the weighted RDS characteristics and the measured RDS characteristics, respectively. The RDS index was calculated using Equation (2).

$$RDS_{index} = F_{source} \times F_{transport} \quad (2)$$

where F_{source} and $F_{transport}$ are the RDS source and transport factors, respectively.

2.4.1. RDS index model for the load

The RDS index model for the load was calculated using the observed source and transport factor values, using Equation (5). The source and transport factors that were included for the load estimations ($F_{source,load}$; $F_{transport,load}$) were calculated separately using Equations (3) and (4).

$$F_{source,load} = \sum_j^m \sum_i^n (M_i \times C_{ij} \times A) \quad (3)$$

$$F_{transport,load} = F_{w,observed} \quad (4)$$

$$RDS_{index,load} = F_{source,load} \times F_{transport,load} \\ = \sum_j^m \sum_i^n (M_i \times C_{ij} \times A \times F_{wi,observed}), \quad (5)$$

where $RDS_{index,load}$ is the potential amount of pollution in the runoff (kg), M_i is the mass of a particular RDS size fraction per unit area (mg/m²), C_{ij} is the measured concentration of the metal of interest j in the RDS with a grain size i (mg/kg), i and j are the numbers of grain size fractions and metal species in the study, respectively, and A is the street surface area in the administrative division unit along the urban–rural gradient (in units of 10⁴ m²).

2.4.2. RDS index model for pollution strength

The RDS index for pollution strength was calculated using the weighted values for the source and transport factors, using Equation (8). The source and transport factors that were included for the strength estimations ($F_{source,strength}$, $F_{transport,strength}$) were calculated separately using Equations (6) and (7).

$$F_{source,strength} = \sum_j^m \sum_i^n \left(T_r^j \times \frac{C_{ij}}{C_{rj}} \times P_i \times M_{weighted} \right) \quad (6)$$

$$F_{transport,strength} = F_{w,weighted} \quad (7)$$

$$RDS_{index,strength} = F_{source,strength} \times F_{transport,strength} \\ = \sum_j^m \sum_i^n \left(T_r^j \times \frac{C_{ij}}{C_{rj}} \times P_i \times M_{weighted} \times F_{wi,weighted} \right), \quad (8)$$

where $RDS_{index,strength}$ is the potential pollution strength in the runoff, T_r^j is the toxic response factor for the metal of interest according to Hakanson (1980) (the values for each element of interest were, in order, Zn = 1, Cr = 2, Ni = 3, and Cu = Pb = 5), C_{rj} is the background concentration of the metal in soil (31.1, 19.7, 27.9, 25.1, and 59.6 mg/kg for Cr, Cu, Ni, Pb, and Zn, respectively (Chen et al., 2004)), P_i is the amount of RDS with grain size i expressed as a percentage of the total RDS mass, and $M_{weighted}$ is the level of the bulk RDS mass per unit area for each sampling site. C_{ij} , i , and j have the same meanings as described above for Equation (5).

Values and categories were established for the $RDS_{index,strength}$ method according to the products of the source and transport factors. $RDS_{index,strength}$ was classified as indicating low levels of risk when $RDS_{index,strength} \leq 150$, moderate levels of risk when $150 < RDS_{index,strength} \leq 300$, considerable levels of risk when $300 < RDS_{index,strength} \leq 600$, and high levels of risk when $RDS_{index,strength} > 600$ (Zhao and Li, 2013).

3. Results

In this section, we will present the results of using the RDS index in a case study in the Beijing metropolitan region. Developing the RDS index for use at the scale of an urban region could allow it to be used to reflect the spatial distributions of the potential environmental risks posed by metals in RDS along the urban–rural gradient. We will first describe the RDS characteristics (the amounts present, grain sizes, associated metals, and its mobility) along the urban–rural gradient.

3.1. Source factors in the RDS index

The source factors in the RDS index in this study included the amount of RDS present, the proportions of RDS particles of each grain size, and the metals associated with the RDS. The source factors were investigated along an urban–rural gradient in the Beijing metropolitan region. The mass of RDS per unit area clearly varied along the urban–rural gradient, with low masses per unit area being found in the UCA (23.9 ± 18.8 g/m²) and CSA (24.2 ± 17.9 g/m²), medium masses per unit area being found in the RTA (54.1 ± 56.4 g/m²), and high masses per unit area being found in the UVA (134.5 ± 153.8 g/m²) and RVA (188.2 ± 202.6 g/m²) (Fig. 1). About 70% of the $M_{weighted}$ values for the UCA and CSA sampling sites fell into level 1, and about 35–40% of the $M_{weighted}$ values for the RVA and UVA sampling sites fell into levels 5 and 6. The RDS particle size distribution also varied along the urban–suburban–rural gradient (Fig. 2). The median RDS diameter, d_{50} , was less than 149 μm in the UCA and CSA, but more than 149 μm in the RTA, RVA, and UVA. In general, the rural areas had higher proportions of coarser RDS particles and larger amounts of RDS. Our site investigations indicated that the

Fig. 1 – Box plot of the amounts of road-deposited sediment (RDS) found along the urban–rural gradient in Beijing. Values are given as the mean $\pm$ the standard deviation. Dashed lines in the figure refer to the values for the M_{weighted} levels. UCA = central urban area, UVA = urban village area, CSA = central suburban county area, RTA = rural town area, and RVA = rural village area.

RVA and UVA were seldom swept (0.2–0.3 times/d) and had high proportions of bare soil or broken and rough road surfaces. The streets in the RVA and UVA are often swept with brooms, whereas mechanical sweepers, which give higher RDS removal efficiencies, are used in urban areas.

The metal concentrations in the RDS along the urban–rural gradient are shown in Fig. 3. The mean Cr, Cu, Pb, and Zn (i.e., excluding Ni) concentrations in the RDS samples from across Beijing were approximately two to four times higher than the background concentrations in soils from Beijing (Chen et al., 2004). The mean total concentrations of the metals analyzed decreased along the urban–rural gradient in the order UCA > CSA > UVA > RTA > RVA. Zn was the most abundant element in the RDS from all of the areas.

3.2. Transport factors in the RDS index

The transport factors that were assessed in this study were mainly chosen for their potential to affect surface runoff that would affect the RDS index. The percentage of each RDS size fraction washed off the surface (F_{wi}) was used to represent the transport factors in the RDS index. The $F_{wi, \text{observed}}$ values ranged from 1% to 66%, and are shown in Table S6 in a publication by Zhao et al. (2011). The washoff amount was calculated for each of the RDS size fractions from the $F_{wi, \text{observed}}$ values by adjusting the amounts and particle size compositions in the RDS to match those in the sampling areas (Shaw et al., 2006; Zhao et al., 2011). The RDS washoff amounts along the urban–rural gradient are shown in Fig. 4. The amount of particles washed off per unit area increased as the rainfall intensity increased, but the washoff as a percentage of the RDS decreased along the urban–rural gradient.

Fig. 2 – Box plot of the mass percentages of road-deposited sediment (RDS) particles in each grain size fraction along the urban–rural gradient in Beijing. UCA = central urban area, UVA = urban village area, CSA = central suburban county area, RTA = rural town area, and RVA = rural village area.

3.3. RDS index for the pollutant load

The RDS index for the pollutant load provides a new way of estimating pollutant loads in large-scale urban runoff. The RDS mobility, amount, and associated metals, and the area of each administrative division along the urban–rural gradient were integrated into the RDS index for the pollutant load,

Fig. 3 – Box plot of the metal concentrations in the road-deposited sediment (RDS) collected along the urban-rural gradient in Beijing. Dashed lines in each subfigure refer to the background concentrations in soils in the Beijing area (SBC, C_{ri}) (Chen et al., 2004). UCA = central urban area, UVA = urban village area, CSA = central suburban county area, RTA = rural town area, and RVA = rural village area.

Fig. 4 – Box plot of the amount of road-deposited sediment (RDS) washoff that will occur along the urban-rural gradient in Beijing. UCA = central urban area, UVA = urban village area, CSA = central suburban county area, RTA = rural town area, and RVA = rural village area. The simulated rainfall in the tests lasted 1 h.

Table 2 – Load of metals combined along the urban–rural gradient/RDS_{index,load} unit: kg.

Sampling site ^a	Rainfall intensity (mm/h) ^c				
	12.87 ^b	32.7	46.7	82.67	101.37
UCA	47.9	197.3	192.6	373.0	444.0
CSA	39.8	136.9	141.9	258.1	318.0
RTA	33.7	111.1	116.3	210.0	258.1
RVA	104.8	344.7	363.3	661.9	805.3

^a Sampling site area: 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.

^b The area of main traffic road for each administrative division units along the urban–rural gradient ($10^4 \times \text{m}^2$): UCA: 9869, CSA: 12071, RTA: 5603, RVA: 11591. The areas above were calculated according to that the traffic road area accounting for 30% of the total traffic areas in Beijing city. The data of urban village area was not available.

^c The simulated 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.

which was calculated using Equation (5). Street area data for the residential areas were not available, so we only calculated pollutant loads for the main roads. The RDS_{index,load} results are presented in Table 2. The RDS_{index,load} values ranged from 33.7 to 805.3 kg at the five rainfall intensities tested, and they decreased in the order RVA > UCA > CSA > RTA. The results indicated that the RDS characteristics (the amount present, the grain size, its mobility, and the metal concentrations in it) varied significantly along the urban–rural gradient, and that they could be combined to determine the potential for pollution to be caused by surface runoff in each administrative division. The urban runoff pollutant load increased as the rainfall intensity increased, indicating that diffuse pollution caused by RDS washoff in heavier rainfall events should be paid special attention.

3.4. RDS index for the pollutant strength

In a similar way to the RDS index for the pollutant load, the RDS index for the pollutant strength provided a new way of identifying risks posed by pollutants in large-scale urban runoff. The RDS index for pollutant strength integrated the RDS mobility, amount, grain size distribution, associated metal concentration, and the toxicities of the metals, and it was calculated using Equation (8). The RDS_{index,strength} results are shown in Fig. 5. The RDS_{index,strength} values for the runoff in the areas that were studied decreased in the order UVA > RVA > RTA > CSA > UCA, which is slightly different to the order for the $F_{\text{source,strength}}$ values, indicating that $F_{\text{source,strength}}$ was more important than $F_{\text{transport,strength}}$ in determining the order for the RDS_{index,strength} values along the urban–rural gradient. The median RDS_{index,strength} for the metals in the RDS collected from the UVA fell into the considerable risk category, and the values in the other areas all fell into the moderate risk category. Our results help to improve our understanding of the risks associated with metal pollutants associated with RDS in urban water.

Fig. 5 – Box plot of the source factors and the road-deposited sediment (RDS) index for pollution strength along the urban–rural gradient in Beijing. UCA = central urban area, UVA = urban village area, CSA = central suburban county area, RTA = rural town area, and RVA = rural village area.

4. Discussion

4.1. Further considerations for the RDS index

There are no index models specifically designed for quantifying and identifying the potential for diffuse pollution caused by surface runoff in urban areas. In this study, we have summarized our research into the relationship between RDS and pollution in urban surface runoff, including in terms of risk assessment (Zhao and Li, 2013) and estimating pollution loads (Zhao et al., 2011), and we developed an RDS index model based on the phosphorus index that is used to rank diffuse pollution risks in agricultural areas. Although the RDS index model could be used for two purposes, quantifying

Table 3 – The washoff load and strength of RDS per unit area along the urban–rural gradient.

Sampling site ^a	Washoff load		Washoff strength	
	RDS washoff load (g/m ²) ^b	Normalization ^d	RDS washoff strength ^c	Normalization
UCA	2.95	0.01	34.60	0.00
CAS	2.73	0.00	37.20	0.04
RTA	7.22	0.30	63.40	0.42
RVA	17.70	1.00	103.90	1.00
UVA	15.22	0.83	102.00	0.97

^a Sampling site area: 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.

^b RDS washoff load was the product of $F_{wi,observed}$ and M_i at rainfall intensity = 46.7 mm/h.

^c RDS washoff strength was the product of $F_{w,weighted}$ and $M_{weighted}$.

^d The method of normalization is $(X-min)/(max-min)$.

pollution loadings and identifying risks from pollution, there are still many deficiencies in the source and transport factors that are used in the index.

In fact, the index cannot be directly verified using measured values because the index cannot be measured. The RDS washoff loads and strengths in the five areas that were studied were compared using the paired t-test after the values had been normalized (Table 3), and this showed that the values that were calculated using different methods correlated significantly ($P < 0.001$) but were not significantly different ($P = 0.132$). The comparison above indirectly analyzed the effectiveness of this index.

A key point is that the transport factor used in the RDS index was mainly derived from RDS washoff experiments using a rainfall simulator, and the impacts of differences between simulated rainfall and natural rainfall values on the accuracy of the transport factors are still unclear (Herngren et al., 2005; Zhao et al., 2010; Brodie and Egodawatta, 2011). The use of simulated rainfall could provide greater flexibility and control of the fundamental rainfall parameters such as intensity and duration and thereby helped to eliminate some of the variables which inherently increase the complexity of stormwater quality research. It can also overcome the constraints of variability and random nature associated with natural rainfall events. But further study was needed to reveal the differences of RDS washoff between simulated rainfall and actual rainfall. The transport factor that we used was focused only on the percentage of the RDS that was washed off, and did not take in account the distances that the particles could travel, being calculated for only one slope and one road surface roughness. The distance between the source and the target water body should be added to the model, so that an understanding of the hydraulic connectivity involved is included, to improve the ability of the RDS index to estimate large-scale urban runoff pollutant loads.

Using geo-statistical techniques may also allow pollution sources and the spatial distributions of released pollutants to be identified (Wang et al., 2012), which would be helpful in evaluating the release of pollutants from their sources, in targeting water bodies, and in defining hydraulic connectivity.

4.2. Important role of particle grain size

The particle grain size is a very important indicator because it links many aspects of the RDS, e.g., the mobilities and

amounts of the particles present, the sources and sinks of the RDS, and the pollutant concentrations and species that are present, including the types of particles to which the pollutants are attached. It has been shown in many previous studies that the RDS particle grain size distribution has an important relationship with the RDS characteristics because it affects the amount of RDS present, the concentrations and speciation of the associated metals, and the mobility of the RDS (Sutherland, 2003; Deletic and Orr, 2005; Lau and Stenstrom, 2005; Zhao et al., 2010). The particle grain size distribution affects the source and transport factors in the RDS index model. We even think that a new RDS index, called the grain size threshold for RDS pollution (for $PM_{2.5}$, a particulate matter diameter $\leq 2.5 \mu m$, for example) could be used in future research, and that such as index would provide a simple and effective way of assessing heavy metal pollution in RDS.

5. Conclusions

We improved an RDS index model and used it to assess pollution loads using data from different types of areas in Beijing (UCA, UVA, CSA, RTA, and RVA). The RDS indices for the pollutant load ($RDS_{index,load}$) and the pollutant strength ($RDS_{index,strength}$) varied strongly along the urban–rural gradient, and the $RDS_{index,strength}$ values increased along the urban–rural gradient. The $RDS_{index,load}$ for the main roads decreased in the order $RVA > UCA > CSA > RTA$. The RDS index integrates the RDS characteristics, such as the amount of RDS, the grain sizes present, the mobility of the RDS, and the associated metals, so the $RDS_{index,load}$ and $RDS_{index,strength}$ results did not simply match the variations in the amounts of RDS found or in the metal concentrations in the RDS along the urban–rural gradient. The RDS index is a new tool for assessing pollution caused by RDS washoff, to help in the management and control of this source of pollution.

Acknowledgments

This study was financially supported by the National Natural Science Foundation of China (41171395), the Major Program of

the Chinese Academy of Sciences (KZZD-EW-10-02), and a Zhenjiang City scientific project (SH2012006). The authors would like to thank Xiaoyan He, Xiaoxue Wang, Xiaomei Wang, Huiliang Wang, Yang Ouyang, Xinzhong Du, Wenzan Li, and Shuxia He for their help collecting road-deposited sediment and with the rainfall simulation experiments.

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