RESEARCH ARTICLE



Pollution and ecological risk assessment of nutrients associated with deposited sediments collected from roof and road surfaces

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Abstract

Surface-deposited sediment in urban areas is an essential environmental medium for assessing nutrient contamination. The total nitrogen (TN) and total phosphorus (TP) pollution associated with surface-deposited sediments can be transported into urban water bodies by runoff and can cause eutrophication of those water bodies. By analyzing the samples collected on roof surfaces and road surfaces, this study provides a comparison of the differences in TN and TP pollution loading in sediments on these two different impervious surface materials. Also, an assessment of the ecological risk of nutrients in surface-deposited sediments with respect to grain size fraction was performed. The results indicate that the TN and TP pollution loading in both road-deposited sediments and roof-deposited sediments indicated an asymmetric "W" trend along with grain size fraction, and both road-deposited sediments and roof-deposited sediments had the highest TN and TP pollution contribution when the particle size is between 250 and 500 µm. TN in roof-deposited sediments has high ecological risk when the particle size is less than 250 µm. These findings help to provide guidance for the management of surface-deposited sediment pollution.

Keywords Road and roof sediments \cdot TN \cdot TP \cdot Pollution loading \cdot Ecological risk \cdot Grain size faction

Introduction

As point source pollution has become effectively managed, non-point source pollution has become increasingly prominent and has thus been recognized as a major contributor to water pollution (U.S. Geological Survey 1999; Lai et al. 2011). Surface-deposited sediment is one of the most important sources of non-point source pollution, and it has received increased consideration in recent decades (Yu et al. 2015). Surface-deposited sediment is a potentially toxic medium, as it contains such pollutants as nutrients, metals, and hydrocarbons. These pollutants originate from a wide range of sources,

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² Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Jiangsu Joint Laboratory of Atmospheric Pollution Control, Nanjing 210044, China including wet and dry deposition, bodies of plants and animals, vehicle exhaust, vehicle and road wear, de-icing operations, abrasion of construction materials, and soil erosion (Rijkenberg and Depree 2010; Shi et al. 2011; Zhao et al. 2014; Bian et al. 2015; Charters et al. 2016). Therefore, serious aquatic pollution can result when sediments are transported into water bodies by stormwater runoff and are thus able to enter the food chain (Herngren et al. 2005; Kong et al. 2012; Ignacio et al. 2014). Surface-deposited sediment has been identified as an important source of urban nonpoint source pollution that can easily enter receiving waters of the USA (Sutherland et al. 1998). Thus, studies on surfacedeposited sediments typically aim to document the harmful effects of deposited sediments on receiving waters. One study investigating road runoff pollutant characteristics found that 60-80% of phosphorus and 50-60% of nitrogen in road runoff were associated with particulates (Hvitved-Jacobsen et al. 1994). If the nutrient load to surface water is too high, it can cause eutrophication of water bodies (Qin et al. 2013). Once a water body undergoes eutrophication, it can lose its primary functions and can subsequently impact the sustainable development of both economy and society (Yang et al. 2008). Therefore, assessments of nutrient ecological risk that can provide specific guidance for protecting aquatic ecosystems

are necessary. Assessments of nutrient ecological risk in surface-deposited sediment are limited.

Additionally, land use, surface type and condition, the proportion of surfaces that are impervious, local traffic volume and characteristics, surface cleaning methods, and cleaning frequency can strongly affect the composition of surface-deposited sediment and the pollutant types and concentrations (Charlesworth et al. 2003; Arnold and Gibbons 1996). Especially in urban areas, road surfaces and roof surfaces are two major impervious surfaces (Angino et al. 1972). Previous studies have indicated that the differences between road surfaces and roof surfaces have important effects on grain size distribution, pollution level, and mobility in rainfall runoff. Therefore, different contamination characteristics can be expected between road and roof sediments (Khanal et al. 2014; Yu et al. 2014).

In addition, many studies have found that finer sediments tend to have higher metal concentrations and more readily wash off into water bodies (Zhao et al. 2010; Zhao and Li 2013). It has been summarized that 55–90% of particles are smaller than 125 µm in urban road runoff, and the corresponding value was 55-85% for roof runoff (Charters et al. 2016). Therefore, pollutants associated with these fine particles can be carried into waterways and can cause a variety of environment problems (Bian et al. 2015). The prevailing belief is that controlling fine particles is the key to controlling sediment pollution. In terms of total amount contributed, understanding pollution loading is significant for assessing the environmental pressure on a region. Unfortunately, to date, analyses of pollution loading of total nitrogen (TN) and total phosphorus (TP) in surfacedeposited sediment have been limited.

Nowadays, research on surface-deposited sediment mainly focuses on research methods, content, distribution characteristics, and source analyses of heavy metals associated with surface-deposited sediment (Sutherland et al. 2004; Ellis and Revitt 1982; Charlesworth and Lees 1999). Also, this research is mostly focused on roaddeposited sediments, so there is a lack of in-depth research on roof-deposited sediments, especially comparisons between road-deposited sediments and roofdeposited sediments. The aim of this paper is to assess contamination and ecological risk of nutrients in surfacedeposited sediments in the northern suburb of Nanjing, China. We divided the sampling area into road and roof areas according to the underlying materials. The particle size distribution of surface-deposited sediments, the concentration of TN and TP, and the pollution loading of TN and TP in surface-deposited sediments were analyzed. This paper seeks to solve two questions: (1) the pollution characteristics of nutrients in surface-deposited sediments of different particle sizes on different underlying surfaces and (2) the ecological risk of nutrients in surfacedeposited sediments with respect to grain size fraction.

The results of this research will provide a scientific basis for the source control and hierarchical management of pollution from surface-deposited sediments.

Materials and methods

Site description

The Nanjing University of Information Science & Technology is located in the northern suburb of Nanjing. Samples were collected in the campus area, near the national Ningliu Road. There are different land uses adjacent to our sampling points, including residential, agricultural, and industrial areas. Nanjing has a humid subtropical climate influenced by the East Asian monsoon, with damp conditions throughout the year (the annual average relative humidity is around 76%). It is very hot in summer (June–August), with an average temperature of 27.7 °C. The annual average rainfall depth is around 1062 mm, and half of the total rainfall occurs in summer.

Sample collection and pre-treatment

A total of ten sampling trips were conducted from September 2014 to February 2016. The sample information has already been published in a previous study (Yu et al. 2017). The sampling road is constructed of asphalt with nearly zero slope, and the surrounding area is a campus greenbelt. The aging and abrasion of the road surface is apparent from more than 10 years of use, even though traffic intensity is light (fewer than 100 vehicles per day). The sampling roof is on the top of a 25-m-high building and is mainly concrete, with some asphalt slim belt surrounding and some rusted cast iron pipes and other steel appurtenances installed approximately 25 cm above the roof top. The roof slope is flat. The major experimental procedures are as follows.

First, a fiber brush was used to dislodge most of the fine particles attached to the surface of the sampling areas. Second, a 250-W vacuum cleaner (Philips FC8264) was used to collect the sediment samples. Particulates smaller than around 1500 μ m can be collected efficiently. This process was repeated three times to ensure that all the possible particles were collected. Finally, all the sediment collected was mixed and transferred to a polyethylene container for transport back to the laboratory for analysis.

The collected surface samples were dried in natural conditions for 2 weeks and then dried to constant weight in an oven at 105 °C for 24 h. The prepared samples were size-fractionated by separating them using stainless steel sieves into seven particle size ranges: < 63, 63–90, 90–125, 125–250, 250–500, 500–850, and > 850 μ m. TN and TP were analyzed according to the Standard Method for the Examination of Water and Wastewater (APHA et al. 1995).

Calculation methods

Pollution loading

In this study, in order to analyze the pollution characteristics of TN and TP in surface-deposited sediments, a pollution loading term was applied. Pollution loading means the proportion of all the pollutants contained in a sample of a particle size range to the total amount of pollutants in all samples. Pollution loading is determined using the following formula:

$$P = \frac{C_i \times M_i}{\sum_{i=1}^7 C_i \times M_i} \times 100\%,$$

where C_i is the TN and TP concentration of sample i (mg/g) and M_i is the mass of sample i (g).

Ecological risk assessment

Currently, there is no systematic standard or index for assessing the ecological risk from nutrients in surface-deposited sediments in China. Surface-deposited sediments can be easily washed into receiving waters during runoff and can settle to the bottom of water bodies, eventually becoming a part of the water sediments. Thus, studies on surface-deposited sediments aim to document the harmful effects of deposited sediments to receiving waters.

Therefore, we try to assess ecological risk based on the nutrient concentration of sediments in aquatic ecosystems by applying the sediment quality guidelines for the protection of aquatic life in Ontario, Canada (CCME 2002) (Table 1). In this standard, LEL represents low effect level, the tolerance level of most benthic organisms; SEL represents serious effect level, under which harmful impacts to benthic organisms may occur. When the concentration is lower than LEL, we consider it to have no ecological risk. When the concentration is between LEL an SEL, we deem it low ecological risk. When the concentration is higher than SEL, we deem it high ecological risk.

Table 1 Ecological risk assessment standards for nutrients in sediments

Standard value	TN (mg/kg)	TP (mg/kg)
LEL	550	600
SEL	4800	2000

Results and discussion

Particle size distribution

Particle size distribution is a critical factor that influences the mobility and portion of pollutants in runoff (Kim and Sansalone 2008; Lee et al. 2014). The particle size distribution results for different impervious surfaces are shown in Fig. 1. It can be concluded from the graph that the particle size of roaddeposited sediments is mostly concentrated around 250-500 μ m (25.76%) and > 800 μ m (29.06%), and the particle size of roof-deposited sediments is mostly concentrated around <125 µm (35.75%) and 250–500 µm (32.13%). The proportion of road-deposited sediments with particle size larger than 800 µm is much higher than that for the other particle size ranges. Our sampling point was located in a village area, and according to Zhao et al. (2010), the result of our study indicates the low efficiency of street sweepers in urban villages, and that these area are seldom swept. Due to the poor management of these areas, the proportion of coarse-particlesize dust is much higher than in other areas. Meanwhile, we found that the proportion of roof-deposited sediments is relatively higher when the particle size is less than 125 µm compared to that of road-deposited sediments. This result is similar to Shen et al. (2016), who found that particles from the roof were predominantly fine particles (120 µm, 65%). This is mainly because the main source of roof-deposited sediments is atmospheric deposition, and the primary particle size of dust-fall ranges from 2.5 to 40 µm (Shaltout et al. 2016). The factors that caused these differences between road- and roof-deposited sediments are likely the differences in the constructed surface material, sediment source, and the influence of human activities, including traffic density and sweeping (Charters et al. 2016; Khanal et al. 2014).

Variation of TN and TP concentrations

Variation of TN and TP concentrations in road-deposited sediments and roof-deposited sediments along the grain size gradient is presented in Fig. 2 panels a and b, respectively. The concentration of TN in roof-deposited sediments is much higher than the concentration of TN in road-deposited sediments along all grain sizes (nearly two times, but not more than three times). The reason for this phenomenon may be the differences in cleaning methods and frequency, pollution sources, the surrounding environment, and human activities (Khanal et al. 2014; Yu et al. 2014). The variation in TN concentration in road-deposited sediments is that the concentration decreases when the particle size increases, but the decline is slight. While for roof-deposited sediments, the concentration of TN first increases and then quickly declines when the grain size increases. The range of variation of TN in roof-deposited sediments is much larger than that of TN in





road-deposited sediments. The variation of TN in roaddeposited sediments is relatively steady.

As shown in Fig. 2b, outside the particle size of $90-250 \mu m$, the concentration of TP in road-deposited sediments is generally higher than that in roof-deposited sediments, and the maximum gap is nearly two times. The variation in TP concentration in road-deposited sediments is that the concentration of TP will decrease when the particle size increases. Meanwhile, we found that the variation characteristic of TP concentration in roofdeposited sediments presented a trend where the concentration first increases and then decreases. But high concentrations of TP still concentrate in small-grain-size fractions. These results are similar to Sartor and Boyd (1972), which indicated that the concentrations of TN and TP in dust with smaller grain sizes are generally higher than those in dust with larger grain sizes. This means that dust with small grain sizes has more potential pollution capacity than dust with larger grain sizes. A previous study in Beijing demonstrated that the 62-105-µm-grain-size fraction has the greatest influence on the overall contamination of road-deposited sediments (Zhao et al. 2010). In this study, we analyzed from the perspective of nutrient pollution and found that particles with grain size of < 125 µm were the most important mass component for both road and roof. Many previous studies support our results, as others have found that finer particles generate the highest pollutant concentrations (Deletic and Orr 2005; Sansalone and Ying. 2008; Zhou et al. 2015).

By comparing the variations of TN and TP, it can be seen that TN and TP in road-deposited sediments both decrease when the grain size fraction increases. And TN and TP in roof-deposited sediments both first increase and then decrease. One law can be described as follows: TN and TP in different impervious surfaces have the same variation regularity. This may be related to the adsorption characteristics of nitrogen and phosphorus, but requires further in-depth study.



Fig. 2 The variation in TN and TP concentrations in grain size fractions of surface-deposited sediment: **a** TN and **b** TP

Pollution loading

Nutrient pollution loading results from road-deposited sediments and roof-deposited sediments are shown in Fig. 3 panels a and b, respectively. These pollution loading calculation results lead us to investigate the loading contribution of particulates in individual size fractions. We found that TN and TP pollution loading from both road-deposited sediments and roof-deposited sediments indicated an asymmetric "w" trend along with grain size fraction as shown in Fig. 3. Roaddeposited sediments in the range of 250–500 μ m (25.55%) and > 800 μ m (19.30%) indicated the highest TN pollution loading, and the grain sizes of $< 90 \ \mu m$ (13.68%) and 250-500 µm (22.99%) indicated the highest TP pollution loading. For roof-deposited sediments, the highest TN pollution loading appeared in the grain sizes of $< 63 \mu m$ (19.08%) and 250-500 µm (29.42%), and the peak TP pollution loading appeared in the grain sizes of $< 63 \mu m (21.15\%), 125-250 \mu m (23.08\%),$ and 250-500 µm (21.03%). A similar asymmetric "w" trend can also be found in our previous study of the pollution loading of heavy metals from roof sediments (Yu et al. 2017). In terms of roof sediments, there may be similarities between the pollution loading of nutrients and the pollution loading of heavy metals, but this too requires further study.

By comparing two different types of impervious surface materials, we find that the TN pollution loading in road-deposited sediments is lower than the TN pollution loading in roof-deposited sediments, provided the particle size is less than 63 μ m. When the particle size is larger than 800 μ m, the TN pollution loading from road-deposited sediments is much

higher than the TN pollution loading from roof-deposited sediments. Both the TN pollution contributions from roaddeposited sediments and roof-deposited sediments are high when the particle size is between 250 and 500 µm. The TP pollution loading from road-deposited sediments is lower than the TP pollution loading from roof-deposited sediments when the particle size is less than 63 µm and the particle size is between 125 and 250 µm, while it is higher than the TP pollution loading from roof-deposited sediments when the particle size is larger than 500 µm. Similarly, when the particle size is between 250 and 500 µm, both road-deposited sediments and roof-deposited sediments have the highest TP pollution contribution. These differences between road surface and roof surface may be because the pollution is a combination of concentration and particle size distribution, and the particle size distribution of road sediments and roof sediments is different (see Fig. 1).

Therefore, controlling TN and TP pollution from surfacedeposited sediments should concentrate on sediments with particle size between 250 and 500 μ m. Previous researchers have found that coarser sediments, such as particles in the range of 250–500 μ m, are less likely to be found in receiving waters, as conventional road sweeping can efficiently remove them (Chang et al. 2005; Herngren et al. 2006). Regardless, we still stress the importance of effective sweeping activities for mitigating pollution loading to the environment. In terms of TN and TP pollution treatment in road-deposited sediments, we should focus on controlling the sediments with particle size less than 90 μ m and with particle size in the range of 250–500 μ m. In terms of TN and TP pollution treatment

Fig. 3 Pollution loading of TN and TP in grain size fractions of surface-deposited sediments: **a** TN and **b** TP (red line indicates the mean value)



from roof-deposited sediments, we should increase efforts in controlling the sediments with particle size less than 500 μ m. Vaze and Chiew (2004) found that, although more than half of the surface pollutants are coarser than 300 μ m, less than 15% of TN and TP are attached to particle size greater than 300 μ m. While we also found that in his study, if the particle size range of 250–500 μ m was considered, nutrient loads are nearly 25% for both TN and TP in this particle size range. It is similar with our findings (see Fig. 3). Furthermore, we can deduce that maybe surface-deposited sediments with particle size in the range of 250–300 μ m have relative high nutrient pollution loading. It is just a suspicion, and we still need a much more specific study to prove this result.

By comparing the pollution loading of TN and TP from surface-deposited sediments, we conclude that the pollution loading of TP was higher than the pollution loading of TN when the particle size is less than 250 μ m, and lower than the pollution loading of TN when the particle size is bigger than 250 μ m. Thus, the grain size of 250 μ m can be defined as a boundary. When the particle size is small, we should strengthen the control of TP pollution, while when the particle size is large, we should put more effort into reducing TN pollution. These findings suggest that an effective reduction in sediments or total suspended soil load does not equal an effective reduction in TN and TP pollution loading. The results here suggest that an effective treatment should target the sediments with particle size between 250 and 500 μ m.

Ecological risk assessment of nutrient

In this study, we attempt to analysis the ecological risk of nutrients in road-deposited sediments and roof-deposited sediments along a grain size gradient. The results of this assessment are shown in Table 2. We conclude that TN in road-deposited sediments presents low ecological risk in all particle size ranges, but TN in roof-deposited sediments with particle size less than 250 μ m poses high ecological risk. And TP in road-deposited sediments with particle size less than 90 μ m has low ecological risk. When the particle size is larger than

90 μ m, TP in road-deposited sediments has essentially no ecological risk. Meanwhile, only when the particle size is between 63 and 125 μ m, TP in roof-deposited sediments has low ecological risk. If the particle size is lower than 90 μ m or larger than 125 μ m, TP in roof-deposited sediments has essentially no ecological risk. The ecological risk of nitrogenous substances in road-deposited sediments has some differences from the ecological risk of nitrogenous substances in roof-deposited sediments. These may be caused by the differences in underlying surface materials. And there is nearly no difference between the ecological risk of phosphorus substances in road-deposited sediments, as both present almost no ecological risk.

Conclusions and prospects

A characteristics and ecological risk assessment comparison of TN and TP pollution between road- and roof-deposited sediments was conducted in this study. Several important conclusions follow.

We found that sediments with smaller grain size have higher concentrations of TN and TP. Both road-deposited sediments and roof-deposited sediments have the highest TN and TP pollution loading when the particle size is between 250 and 500 μ m. By comparing the pollution loading of TN and TP, we found that the grain size of 250 μ m can be defined as a boundary, such that pollution loading of TP was higher than the pollution loading of TN when the particle size was less than 250 μ m, and lower than the pollution loading of TN when the particle size was larger than 250 μ m.

TN in road-deposited sediments generally has low ecological risk for all grain size fractions, but TN in roof-deposited sediments has high ecological risk when the particle size is less than 250 μ m. TP has almost no ecological risk in all grain size fractions of both road-deposited sediments and roofdeposited sediments.

By analyzing pollution loading and assessing ecological risk, we summarize two specific conclusions: (1) TN in

Table 2Ecological riskassessment of nutrients in surface-
deposited sediment in the north-
ern suburbs of Nanjing

Particle size range (µm)	Ecological risk level			
	TN in road dust	TN in roof dust	TP in road dust	TP in roof dust
< 63	Low	High	Low	None
63–90	Low	High	Low	Low
90–125	Low	High	None	Low
125-250	Low	High	None	None
250-500	Low	Low	None	None
500-800	Low	Low	None	None
> 800	Low	Low	None	None

roof-deposited sediments of grain sizes < 63 and 125–250 μ m not only has high pollution loading, but also has high ecological risk. Surface-deposited sediments within this two grain size ranges have great pollution impact on the urban surface water environment, and they are key to controlling TN pollution. (2) Although TN and TP in the grain size of 250–500 μ m both have little or no ecological risk, the ecological risk assessment standard we used is only based on TN and TP concentration. If the solid mass is considered at the same time, owing to sediments within the range of 250–500 μ m having a higher solid mass fraction for particles in this particle size range, it may be possible to transport much more sediment into water bodies than other particle size ranges, and thus, the nutrient concentrations may continue to rise. As a result, they may still pose relatively high ecological risk.

This study still has several deficiencies. Sampling points were centrally located in the campus area, so the study lacks representativeness. And in this study, we have taken the guidelines for sediment quality for the protection of aquatic life in Ontario, Canada, as the assessment standard and did not consider the differences in nutrient background values in sediments from different areas. Therefore, the ecological risk assessments of this study are only preliminary results and thus require further in-depth study.

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