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Evaluation of surface runoff and road dust as sources of nitrogen using nitrate isotopic composition

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ABSTRACT

Stable nitrogen and oxygen isotope ratios of nitrate ($\delta^{15}N$–NO_3 and $\delta^{18}O$–NO_3) have recently been used to identify nitrogen sources in water environments. However, there have been no investigations designed to determine nitrate isotopes in non-point sources in urban areas for evaluating the impact of surface deposits on nitrogen in surface runoff. In this study, we collected rainwater, surface runoff and surface deposits (road dust, roof dust and soil) to evaluate the nitrogen sources in surface runoff using nitrate isotopes. There were no large differences in $\delta^{15}N$–NO_3 among rainwater (−0.3‰ to 1.5‰), surface runoff (−2.7‰ to 0.4‰), leachates from road dust (−5.8‰ to 6.2‰) and soil (−11.5‰ to 0.6‰). In contrast, the $\delta^{18}O$–NO_3 in surface runoff (28.5–47.9‰) was lower than that in rainwater (62.7–78.6‰), and higher than that in leachates from road dust (6.1–27.6‰) and soil (−1.1‰ to 6.6‰). $\delta^{18}O$–NO_3 is a useful indicator for evaluating the NO_3–N sources in surface runoff. Using this indicator, NO_3–N from road dust was estimated to account for more than half of the NO_3–N in surface runoff. This is consistent with a result based on a comparison of their loads per unit surface between rainwater and surface runoff, which also showed that most of the nitrogen in surface runoff was derived from surface deposits.

1. Introduction

Urban surface runoff is contaminated by non-point pollutants such as nitrogen, heavy metals and polycyclic aromatic hydrocarbons derived from traffic activity and has been regarded as an important pathway for the delivery of pollutants to water environments (Garnaud et al., 1999; Zobrist et al., 2000; Haus et al., 2007; Shinya et al., 2002; Murakami et al., 2004; Baun et al., 2006). Non-point pollutants are widely distributed in the atmosphere and on surfaces. Since it is difficult to control these distributed pollutants, pollutant loads from non-point sources have become relatively larger than those from point sources. Consequently, effective strategies are needed to control pollutants from non-point sources.

The sources of non-point derived nitrogen in urban areas include atmospheric particles and surface deposits (e.g. road dust) (Turner et al., 2002; Shinya et al., 2003; Tsai and Cheng, 2004). It is useful to distinguish between the loads imposed by the atmosphere and surface deposits when considering source-specific countermeasures such as road sweeping. A comprehensive investigation of pollutant loads per unit surface in rainwater and surface runoff is a traditional way to distinguish between loads imposed by the atmosphere and by surface deposits (Davis et al., 2001; Sabin et al., 2005). Sabin et al. (2005) collected wet and dry deposition and stormwater within a small, highly impervious urban catchment in Los Angeles and estimated that, according to the ratio of the loads per unit surface in total deposition to that in stormwater, atmospheric deposition potentially accounted for as much as 57–100% of the total trace metal loads in stormwater within the study area. Zobrist et al. (2000) showed that NO_3 loads in wet depositions and the tile roof runoff and the runoff ratio were 2.9 mg N m^{-2}, 3.1 mg N m^{-2}, and 79%, respectively, and the contribution of NO_3 from wet atmospheric deposition to the runoff was therefore calculated to be 93%. However, there were large uncertainties in the estimates of the loads per unit surface, because of the propagation of errors associated with determining the concentration, rainfall amount, and runoff flow rate. Another approach is to use nitrogen stable isotope ratios, because nitrogen stable isotope ratios differ among sources, and this difference would be useful for estimating their sources (Widory et al., 2005; Kohzu et al., 2009; Xue et al., 2009). Recently, it

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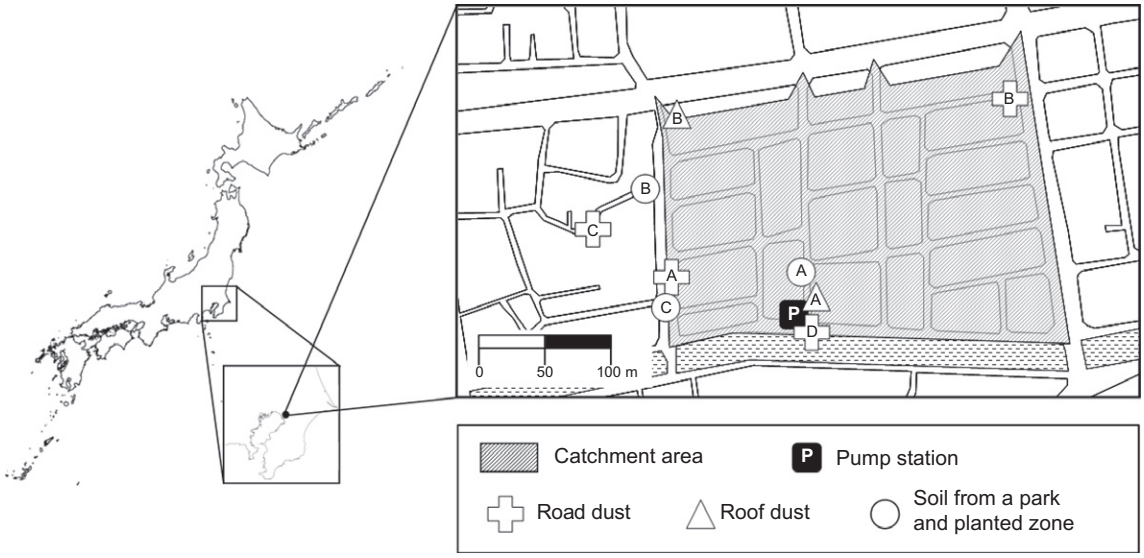

Fig. 1. Sampling locations.

has become possible to measure stable nitrogen and oxygen isotopes of nitrates ($\delta^{15}N$–NO_3 and $\delta^{18}O$–NO_3), and therefore $\delta^{15}N$–NO_3 and $\delta^{18}O$–NO_3 have been used to identify nitrogen sources in agricultural soil, rivers, groundwater, estuarine systems, and aerosols (Mengis et al., 2001; Kaown et al., 2009; Wankel et al., 2009a,b; Ohte et al., 2010). However, to our knowledge, there have been no investigations to determine the nitrate isotopes in non-point sources in urban areas and to evaluate the impact of surface deposits on nitrogen in surface runoff. It is now necessary to investigate $\delta^{15}N$–NO_3 and $\delta^{18}O$–NO_3 from non-point sources in urban areas, and to evaluate the impact of surface deposits on nitrogen in surface runoff using $\delta^{15}N$–NO_3 and $\delta^{18}O$–NO_3.

In this study, we collected rainwater and surface runoff in an urban catchment served by a separate sewer system to measure their nitrogen concentrations, loads per unit surface, $\delta^{15}N$–NO_3 and $\delta^{18}O$–NO_3. In addition, we collected surface deposits (road dust, roof dust and soil from a park and planted zones) and compared the nitrogen concentration, $\delta^{15}N$–NO_3 and $\delta^{18}O$–NO_3 in leachates from the surface deposits with those in surface runoff and rainwater to evaluate the nitrogen sources in the surface runoff.

2. Materials and methods

2.1. Samples

The rainwater, surface runoff and surface deposits were collected in Asahimachi, Chiba City, Chiba Prefecture, Japan, as shown in Fig. 1. The catchment area is about 5.4 ha, and is served by a separate sewer system. The discharge pump station is situated at the end of the sewer system. In this study, road dust, roof dust, and soil from a park and planted zones were considered surface deposits.

The rainwater (bulk water) was collected in cylindrical containers (10 cm in diameter) installed 2 m above the ground. The rainwater sampler collected rainwater together with atmospheric fallout during dry weather periods. Surface runoff was collected at the end of separate sewer pipes by an automatic water sampler (ISCO Co., Ltd.). The automatic water sampler went on line when 1 mm of rainfall was recorded within 2 h. The sampling interval was 10 min. A water level gauge (Grant Co., Ltd.) was also employed to measure the water level of the surface runoff. The flow rate of the surface runoff was calculated with a height-quantity curve.

Table 1 shows information about the rainwater and surface runoff sampling. The rainwater samples were collected in all five events, whereas surface runoff samples were collected in Events 2 and 4. The initial 2-h samples of surface runoff were collected. The flow rate was measured in Event 4. The rainwater and surface runoff samples were filtered with a PTFE membrane filter (Advantec, H045A, pore size: 0.45 μm).

Road dust (collected at sites A–D, Fig. 1), roof dust (A, B) and soil from a park and planted zones (A–C) were also collected in the same catchment area on March 30, 2009. The antecedent dry weather period and rainfall amount were 117 h and 4.5 mm, respectively. The road dust samples were collected from the side of a road using a dry vacuum cleaner (Hitachi CV-100S6). The roof dust and soil samples were collected with pig-bristle brushes and plastic tubes, respectively. The road dust, roof dust and soil were air-dried at room temperature, and then homogenized after being sieved through a 2 mm mesh to remove larger particles. The amounts of road dust collected at Sites A to D were 138, 51,

Table 1
Information on rainwater and surface runoff sampling.

	Date	Total rainfall amount (mm)	Rainfall duration (h)	Maximum rainfall intensity (mm 10 min^{-1})	Antecedent dry weather period (h)	Antecedent rainfall amount (mm)
Event 1	May 12–14, 2008	63.5	21	2.0	31	23.0
Event 2	June 12, 2008	30.5	15	2.5	152	31.5
Event 3	June 21–23, 2008	103.5	47	4.0	200	30.5
Event 4	June 26, 2008	2.5	4	0.5	49	103.5
Event 5	June 29–30, 2008	65.0	31	8.0	54	2.5