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Contribution of atmospheric nitrogen deposition to diffuse pollution in a typical hilly red soil catchment in southern China

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ABSTRACT

Atmospheric nitrogen (N) deposition is currently high and meanwhile diffuse N pollution is also serious in China. The correlation between N deposition and riverine N export and the contribution of N deposition to riverine N export were investigated in a typical hilly red soil catchment in southern China over a two-year period. N deposition was as high as 26.1 to 55.8 kg N/(ha·yr) across different land uses in the studied catchment, while the riverine N exports ranged from 7.2 to 9.6 kg N/(ha·yr) in the forest sub-catchment and 27.4 to 30.3 kg N/(ha·yr) in the agricultural sub-catchment. The correlations between both wet N deposition and riverine N export and precipitation were highly positive, and so were the correlations between NH_4^+-N or NO_3^--N wet deposition and riverine NH_4^+-N or NO_3^--N exports except for NH_4^+-N in the agricultural sub-catchment, indicating that N deposition contributed to riverine N export. The monthly export coefficients of atmospheric deposited N from land to river in the forest sub-catchment (with a mean of 14%) presented a significant positive correlation with precipitation, while the monthly contributions of atmospheric deposition to riverine N export (with a mean of 18.7% in the agricultural sub-catchment and a mean of 21.0% in the whole catchment) were significantly and negatively correlated with precipitation. The relatively high contribution of N deposition to diffuse N pollution in the catchment suggests that efforts should be done to control anthropogenic reactive N emissions to the atmosphere in hilly red soil regions in southern China.

Introduction

It is well known that diffuse source pollution typically comes from unlicensed sources and dispersed land-use activities (Environmental Agency, 2007). Major sources of diffuse water pollution include contaminated run-off from roads, drainage from housing estates, surplus nutrients from farmland, livestock wastes, as well as atmospheric deposition mainly due to the

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emissions of reactive nitrogen gases into the air from transport, industry and agriculture (Vitousek et al., 1997; Liu et al., 2013).

Former studies had shown that nitrogen (N) deposition onto a watershed contributed a large proportion to riverine N input to the downstream lakes or bays (Paerl, 1997; Boyer et al., 2002; Howarth, 2006). For example, in the study of N load to Chesapeake Bay by Howarth (2006), atmospheric N deposition to the Bay includes direct N deposition to the water surface of the Bay and indirect N deposition by input to the Bay from deposition onto watersheds. Using monitoring data of the Environmental Protection Agency of United States for N deposition and the retention rate of N deposition cited from a forest watershed, the author estimated that N deposition contributed 26% of the total N load to the Bay. Considering that the cited N deposition was underestimated and deposition retention rate overestimated, the author also calculated the total input to the Bay from deposition based on the scenarios that N deposition increased to 15.5 kg N/(ha·yr) and the retention rate for N deposition was 70%. Then the contribution of N deposition to the N load to Chesapeake Bay increased to 32% and even to 49%, showing very high values.

Recently Liu et al. (2013) reported that annual bulk N deposition increased by 60% over China during the 1980s (13.2 kg N/ha) and 2000s (21.1 kg N/ha), largely due to the increasing NH_3 emissions from cropland N fertilization and livestock waste and NOx emissions from industry and transport accompanied with the strong economic growth in the recent decades. Meanwhile, diffuse pollution has also been increasingly more serious in China since the 1980s. For example, Billen et al. (2013) investigated the distribution of net anthropogenic N input at the scale of world's watersheds and found very high (50 to 75 kg N/(ha·yr) or even higher) N input in major watersheds (*e.g.*, the Yangtze River, the Yellow River and the Pearl River watersheds) in China. The high N input to watersheds in China could be one of the major causes for the frequent occurrence of algae blooming in lakes or coasts in China in the recent years (Duan et al., 2009).

However, the contributions of atmospheric N deposition to diffuse N pollution in major watersheds of China are less examined at present. In this study, a case study was conducted in a typical hilly red soil catchment in Central South China to systematically investigate the contribution of N deposition to diffuse N pollution. The objectives of this study were: (1) to quantify atmospheric dry and wet N depositions in a catchment scale; (2) to find out the correlation between N deposition and riverine N export from the catchment; and (3) to estimate the contribution of atmospheric deposition to riverine N export from the catchment.

1. Materials and methods

1.1. Study area

This study was conducted in the 135-km^2 Jinjing Catchment of the Xiangjiang River Watershed System in Changsha, the capital city of Hunan Province, China. The catchment is 70 km northeast of the city centre and has a population of 45,000. It is a typical hilly agricultural catchment in subtropical central China and has forest, paddy field and tea field as the three primary land use types, which account for 65.5%, 26.5% and 2.4% of the total catchment area, respectively. The other minor land uses in the catchment include reservoir/pond, residential area, river and road, accounting for 5.6% of the total catchment area. The climate in the catchment is subtropical monsoon and humid, with an average annual rainfall of 1330 mm (1955–2010), an annual mean air temperature of 17.5°C (1955–2010) and a prevailing wind from north and northwest in the whole year. Three sampling sites, named Xishan, Huinong and Feiyue, were chosen to represent the three main land use types, *i.e.*, forest, paddy field and tea field, respectively, in the Jinjing Catchment (Fig. 1) to conduct atmospheric N deposition monitoring. Besides, one forest sub-catchment (named Fuling; area: 0.6 km^2) and one agricultural sub-catchment (named Tuojia; area: 56.9 km^2) were chosen to conduct riverine/ditch N export monitoring at the outlets of the sub-catchments. As a mirror of the Jinjing Catchment, the agricultural sub-catchment had a similar land use pattern as the Jinjing Catchment with forest, paddy field, tea field and other land uses accounting for 58.3%, 31.7%, 4.3% and 5.7% respectively, of the total sub-catchment area.

1.2. Sampling

1.2.1. Atmospheric wet and dry N deposition monitoring

In this study, the year-round wet and dry N deposition rates were monitored at the three sampling sites (Feiyue, Huinong and Xishan) from September 2010 to August 2012. At each site, daily (8:00 am to 8:00 am next day) rainfall or snowfall samples were collected by using wet-only samplers installed at a height of 1.5 m above the ground. For the dry N deposition sampling, the concentrations of five main N_r species (ammonia — NH_3, nitrogen dioxide — NO_2, nitric acid — HNO_3, particulate ammonium — pNH_4^+ and particulate nitrate — pNO_3^-) were measured monthly at each site. NH_3, HNO_3, pNH_4^+ and pNO_3^- were observed using the DELTA (DEnuder for Long-Term Atmospheric) sampling system designed by the Centre for Ecology and Hydrology, Edinburgh, UK. Detailed information on measuring the wet and dry N depositions at the sampling sites can be found in Shen et al. (2009, 2013).

1.2.2. Riverine N export monitoring

A real-time hydrological monitoring system and water quality sampling point were set up at each outlet of the two selected sub-catchments. Instantaneous discharge data were automatically collected in every 10 min and were used to calculate the daily cumulative runoff. Water samples were periodically collected at a 10-day interval at the outlet of each sub-catchment. The collected water samples were immediately transported to the laboratory and stored at −18°C till analysis. More detailed information on riverine N export monitoring can be seen in Wang et al. (2014).

1.3. Analytical procedures

The collected wet deposition and riverine N export samples were stored in the polypropylene bottles at −18°C and were usually analyzed at one-month intervals. After thawing, the samples were filtered using 0.45 μm filter membranes. The NH_4^+-N and NO_3^--N contents in the filtrates were analyzed with a flow-injection auto-analyzer (Tecator FIA Star 5000 Analyzer, Foss Tecator, Sweden). The total dissolved N content in the filtrate was measured by the potassium persulfate oxidation method (Zhang et al., 2008), with the transformed NO_3^- also analyzed by the flow-injection auto-analyzer. The dissolved organic N content (DON) was calculated as the difference between the total N and the inorganic N contents.