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Atmospheric organic nitrogen deposition: Analysis of nationwide data and a case study in Northeast China

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ABSTRACT

The origin of atmospheric dissolved organic nitrogen (DON) deposition is not very clear at present. Across China, the DON deposition was substantially larger than that of world and Europe, and we found significant positive correlation between contribution of DON and the deposition flux with pristine site data lying in outlier, possibly reflecting the acute air quality problems in China. For a case study in Northeast China, we revealed the deposited DON was mainly derived from intensive agricultural activities rather than the natural sources by analyzing the compiled dataset across China and correlating DON flux with NH_4^+–N and NO_3^-–N. Crop pollens and combustion of fossil fuels for heating probably contributed to summer and autumn DON flux respectively. Overall, in Northeast China, DON deposition could exert important roles in agro-ecosystem nutrient management and carbon sequestration of natural ecosystems; nationally, it was suggested to found rational network for monitoring DON deposition.

1. Introduction

Due to dramatic increase of atmospheric nitrogen (N) deposition since the industrial revolution (Galloway et al., 2004, 2008; Vitousek et al., 1997), N, the most limiting nutrient for plant growth and metabolism (Aerts and Chapin, 2000; LeBauer and Treseder, 2008), has been causing substantial environmental impacts and even becoming harmful to many ecosystems (such as soil and water acidification, eutrophication, and nutrient imbalances) (Jiang et al., 2010; Liu et al., 2011; Matson et al., 2002). Thus, quantifying the nitrogen deposition and delineating its anthropogenic sources are the key first step for taking further measure to reduce the harmful influences. N-containing compounds from rainwater include NH_4^+–N, NO_3^-–N and dissolved organic N (DON). NH_4^+–N and NO_3^-–N are easier to be determined, and their anthropogenic sources and deposition patterns have also been better understood. The major sources of NH_4^+–N is animal excretion and fertilizer, and NO_3^-–N is primarily derived from the combustion of fossil fuels (Cornell et al., 2003; Hertel et al., 2006). In contrast, the sources of DON still remain poorly understood because the composition of DON is very complex, originating from anthropogenic or natural sources (Cape et al., 2011; Neff et al., 2002). In order to clearly distinguish the sources of deposited DON, previous researches have employed various methods, such as integrating analysis of large geographical data (Cornell, 2011; Neff et al., 2002; Zhang et al., 2012), correlating DON flux with other ions in long-term time scale (Ham and Tamiya, 2007; Kieber et al., 2005), comparing differences of season variation between DON and NH_4^+–N or NO_3^-–N (Cape et al., 2004; Keene et al., 2002) and stable isotope method (Kelly et al., 2005; Miyazaki et al., 2010).

North America, Europe and East Asia are the three hotspots of N deposition around world (Galloway et al., 2004). National Acid Deposition Program in the U.S., the European Monitoring and Evaluation Program and a recent research across China (Lu and Tian, 2007) have mostly concentrated on inorganic N deposition. However, DON is a ubiquitous and quantitatively significant component of deposited N, accounting for 11–56% of total dissolved N (TDN) across the world (Cape et al., 2011; Cornell, 2011). In addition, the bioavailability of DON showed almost the same importance as inorganic N for both terrestrial and aquatic ecosystems (Bronk et al., 2007; Nasholm et al., 2009). Therefore, ignoring this component could potentially underestimate the N input and subsequently leads to serious uncertainty when evaluating the ecological risks of N deposition.

In recent years, more attentions have focused on DON deposition. From an analysis of global DON deposition data, Neff et al.

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(2002) summarized that the proportional contribution of DON to total deposition was essentially the same in both polluted and unpolluted environments. However, China is facing acute air quality problems (Cornell, 2011). Across the nation, the patterns of DON deposition and its relative importance have not been explored. Zhang et al. (2012) presented a comprehensive assessment of DON deposition in different regions of China. Nevertheless, the monitoring sites were mostly located in developed regions (east and middle of China) and around Beijing area. Northeast China is an important base of agricultural production and heavy industry area in China. So far, DON deposition status in this region is still scarce to the best of our knowledge.

The lower Liaohe River Plain (LRP) is located at central part of Liaoning Province (Fig. 1), with highly intensive agricultural activity, large population density and high level of economic development in Northeast China. Thus, LRP is an ideal area to survey DON deposition and analyze its causes, through which we could fill the data gap in China. In this paper, we firstly synthesized the national DON deposition data from published papers. Then we characterized seasonal variation of DON deposition in LRP from a 5-year observation (2004–2008) and explained the causes based on the national result. Lastly, we evaluated the potential significances of deposited DON for both agricultural and natural ecosystem in Northeast China and presented recommendations of DON deposition monitoring in the country.

2. Materials and methods

2.1. Database of national DON deposition

We compiled a literature database of DON deposition, covering 52 monitoring sites widely distributed over China (Appendix 1). These sites were mostly located at big cities and economically developed regions (east and middle of China), and data in northeast China, northwest China and the Tibetan plateau was relatively fewer. The reference sources included English-language journals and Chinese-language journals. Yearly DON deposition and proportion of DON to TDN were used in our analysis.

2.2. Research site

Rainwater was sampled in Shenyang Experimental Station (41°32′ N latitude, 123°23′ E longitude and at an altitude of 31 m above sea level), Chinese Academy of Sciences (CAS), located at the central part of LRP (Fig. 1). This station belongs to Chinese Ecosystem Research Network (CERN), concentrating on continuous observation of environmental change and conducting long-term manipulated experiments in agro-ecosystem. This site is mainly affected by warm-temperate continental monsoon climate with distinct rainy season from June to September. The average annual precipitation is about 520 mm. The main land-use types around the monitoring site are paddy field and dry farmland with corn or soybean. The cropping system is single harvest per year. This site is also embraced by typical heavy industry cities of Northeast China, about 35 km south of Shenyang and Fushun, and 40 km north of Liaoyang, Benxi and Anshan (Fig. 1).

2.3. Sampling and chemical analysis

The experimental station did not operate due to the severe weather condition in winter. Thus, we did not sample precipitation in January, February and December of every year. However, this scheme of sampling collection had very small influence on quantifying annual N deposition because the precipitation in the above dry period only accounted for about 5% of annual value.

At the monitoring site, a manual rain gauge (SDM6, Tianjin Weather Equipment Inc., China) made of stainless steel was installed to collect rainwater samples. The rain gauge was placed at open areas without surrounding obstacles. Between the rain events, the funnel-shaped top and the inner collecting tube of the rain gauge were cleaned using acid wash of 10% HCl solution, and then cleaned with deionized water to avoid contamination (Anderson and Downing, 2006). The sample was collected immediately after each rain event. The amount of rainwater was recorded, and then the samples were thoroughly mixed. Samples with visible contamination (e.g., by bird droppings) were discarded. The samples were stored at the refrigerator of −20 °C before analysis, avoiding transformation of inorganic N to organic N by microorganisms (Cornell et al., 2003). Samples were filtered through 0.45 μm glass fiber filters before analysis. The dissolved inorganic nitrogen (DIN) concentrations including NH_4^+–N and NO_3^-–N in rainwater was determined following standard procedure with a continuous flow analyzer (TRAACS 2000, Bran-Luebbe Inc., Germany). TDN was analyzed by a modified Kjeldahl procedure (Lu, 1999). In this method, iron powder was used as a reducing agent in the pretreatment procedure to convert NO_3^-–N to NH_4^+–N (Liao, 1981), and TDN in samples was then quantified by Kjeldahl analysis. DON was calculated by the difference of TDN and DIN. Because the rain collector is a standard rain gauge rather than a wet-only, event based system, the N deposition we quantified was the bulk deposition.

2.4. Calculation and data analysis

The volume-weighted concentrations (C (mg N L^{-1})) were calculated using the equation:

$$C' = \left(\sum_{i=1}^{n} C_i P_i\right) \Big/ \left(\sum_{i=1}^{n} P_i\right)$$

where P_i is the rainwater amount (mm) of an individual precipitation event i, C_i is the measured N concentration (mg N L^{-1}), and n is the number of precipitation event in the corresponding monthly or annually scale.

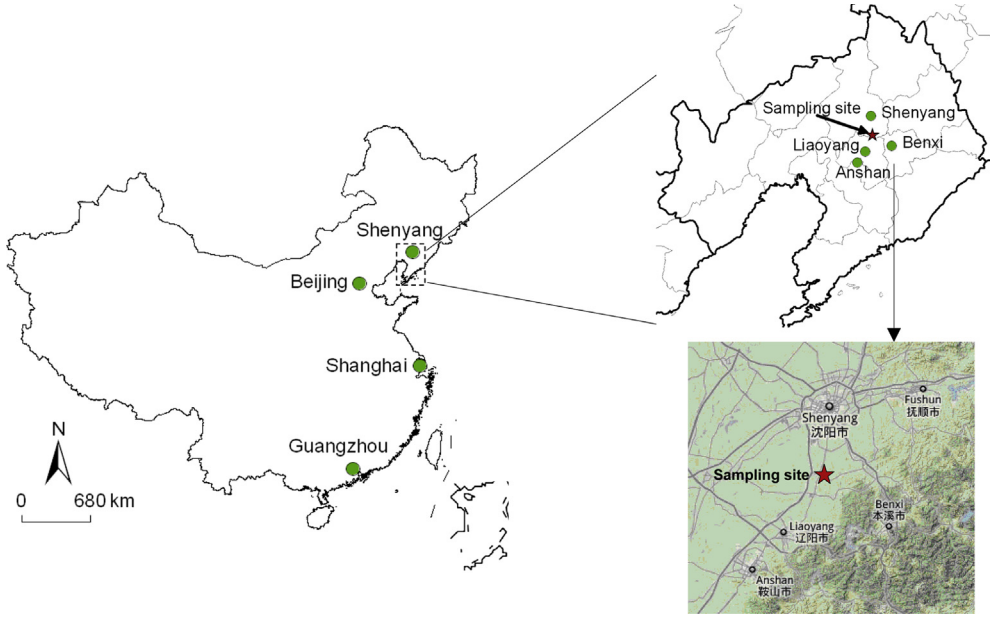

Fig. 1. Location map of the sampling site (Shenyang Experimental Station).