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RESEARCH ARTICLE

Nitrogen concentration of rainfall in Dalian, China

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Abstract Wet atmospheric deposition samples were collected in rainy days from 2004 to 2008 in Dalian of Liaoning Province, Northeast of China, which were measured by rain gauge and analyzed for total inorganic nitrogen (TIN) concentration. The mean annual volume of rainfall was from 438.25 to 850.94 mm, and the concentration of TIN was 3.47 mg/L, which showed negative correlation with the volume of rainfall. The order of TIN concentration among seasons was: winter > spring > autumn > summer. Increased use of fertilizers in agricultural areas or widespread use of private transportation led to the increase of TIN fluxes. Variations of TIN concentration among years were significantly different; the annual precipitation TIN inputs were positively related to the volume of rainfall and were surprisingly high with the deposition flux ranging from 10.24 to 25.17 kg N/($hm^2 \cdot a$). The maximum mean annual flux was 25.17 kg N/($hm^2 \cdot a$) which is equal to 53.94 kg N/($hm^2 \cdot a$) usage of $CO(NH)_2$ (an amount that could have caused changes in terrestrial life). The fluxes of TIN also showed a different seasonal fluctuation during the course of our study, and we found that majority of TIN deposition occurred in summer (from June to August), which accounted for 56.44% of total annual precipitation and 40.06% of total annual deposition flux. Annual TIN fluxes decreased considerably after the rainy season and reached the lowest level (1.39 kg N/($hm^2 \cdot a$)) in winter.

Keywords nitrogen concentration, rainfall, total inorganic nitrogen (TIN), flux

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1 Introduction

The most abundant element in the atmosphere is nitrogen (N) (78%), which is necessary to sustain life on this planet. Nitrogen is an important limiting nutrient in sensitive ecosystems, and atmospheric nitrogen deposition is a significant component of the ecosystem balance (Bobbink and Lamers, 2002). The process of gas or aerosol moving from the atmosphere to the earth's surface is called deposition. Wet deposition occurs to the earth surface by precipitation, the process of removing gases and particles from the atmosphere by rain, snow, sleet or fog. Sources of deposition are mainly transportation, power plants, industry and agriculture (Driscoll et al., 2003). In order to satisfy a growing global demand of food and energy, anthropogenic use of fossil fuels and agricultural fertilizers has increased dramatically during the last 150 years (Galloway and Cowling, 2002). Increased utilization and emission of N have led to elevated atmospheric N concentrations, long-range atmospheric transport and increased atmospheric deposition of N as determined by wind and precipitation patterns and distances to emission sources (Holland et al., 2005). Consequently, the amount of bioavailable N has increased dramatically in nature (Galloway and Cowling, 2002), and the anthropogenic disturbance of the N cycle is part of the global change concept (Steffen et al., 2004).

The greatest proportion of atmospheric reactive nitrogen is in nitrogen oxides and ammonia. Nitrogen oxides are emitted to the atmosphere in exhaust from burning fossil fuels such as gasoline, diesel, coal and natural gas, and from wildfire and prescribed burning of forests and grasslands. Nitrogen released from livestock and agriculture is primarily in the form of ammonia. When more nitrogen is produced than is taken by plants or the soil, some of the excessive nitrogen may be emitted to the atmosphere as gas (Upadhyay et al., 2008).

We measured atmospheric nitrogen deposition for 5 years from 2004 to 2008. The goals of this study were to calculate the concentrations and fluxes of total inorganic nitrogen (TIN) derived from wet nitrogen deposition and to explore its ecological significance in Dalian, China, thus providing information about nitrogen change in this region.

2 Materials and methods

The study site was located in the greenhouse of Dalian Jiaotong University, where the mean annual temperature was 8°C–11°C, and the annual rainfall was between 550 and 1000 mm. It is in warm temperate and semihumid zone of monsoon and marine climate, and belongs to the urban ecosystem.

Wet atmosphere nitrogen samples were collected and analyzed for wet deposition contents and fluxes of nitrogen during the sampling period. Samples were collected after rain, measured by rain gauge, put into 2 Teflon boxes to minimize contamination, and analyzed for TIN.

3 Results

3.1 Annual contents of TIN in rainfall

The contents of TIN in rainfall in recent years were high (Fig. 1), and all the annual values were beyond 2.50 mg/L with the mean value of 3.47 mg/L. The greatest proportion of atmospheric nitrogen species was in nitrogen oxides and ammonia. Human alteration of the N cycle is the release of NO_x gas from fossil fuel combustion and NH_3 gas from agricultural production to the atmosphere, where they may be converted to nitrate and ammonium, respectively, and then deposited on the land surface as wet and dry deposition (Vitousek et al., 1997). NH_3 is emitted from a large number of sources such as volatilization from animal waste and synthetic fertilizers, biomass burning (including forest fires), soils under native vegetation and agricultural crop emissions from human excreta and fossil fuel combustion (Olivier et al., 1996). Sampling site was near the street where a developed traffic system influenced the contents, and the greenhouse was planted with many kinds of flowers whose fertilizers also impact the contents. The contents of TIN decreased significantly with an increase in the volume of rainfall. For example, the content of TIN was 4.29 mg/L in 2006 whose volume of rainfall was 438.25 mm and decreased to 2.96 mg/L in 2005 when the volume of rainfall was 850.94 mm.

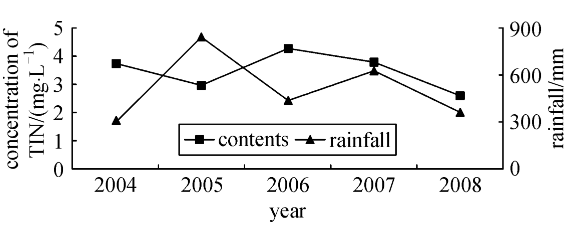

Fig. 1 Annual contents of TIN in rainfall

3.2 Seasonal contents of TIN in rainfall

Figure 2 showed a significant variation in TIN among seasons with the trend as follows: winter > spring > autumn > summer. The relatively high content in spring was resulted from the change of climate conditions such as temperature, farming, fertilization and relevant agricultural activities as well as high frequency of precipitation leaching. The content of TIN in winter was higher than in the other three seasons as a result of the increasing use of fossil fuels in winter for warming, which led to more nitrate nitrogen being released to the atmosphere. The long-term accumulation in the air aggravated the condition of litter rainfall. Lower contents in summer and autumn were related to the dilution effect of strong precipitation.

Recently, the concentration of TIN in rainfall increased, while the frequency of precipitation decreased. For example, the largest value of TIN concentration was 24.35 mg/L when the volume of rainfall was 4.6 mm. Rapid industrialization as well as increased level of life standard and use of automobiles contributed to the increased concentration of nitrate nitrogen in the atmosphere year by year. The character of long transportation accompanied by the change of wind direction in winter led to the increase in particles of nitrogen such as dust which resulted in further change of concentration of NO_x in the atmosphere.

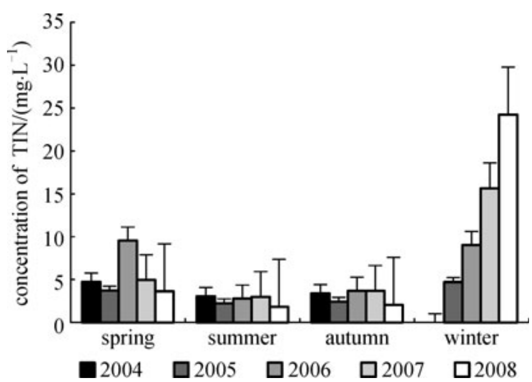

Fig. 2 Variation of concentration of TIN among seasons

3.3 Fluxes of nitrogen by wet deposition and interannual variations

The variations in rainfall among years were significantly different (Fig. 3), with the biggest rainfall of 850.94 mm in 2005, the smallest of 308.50 mm in 2004, and the mean

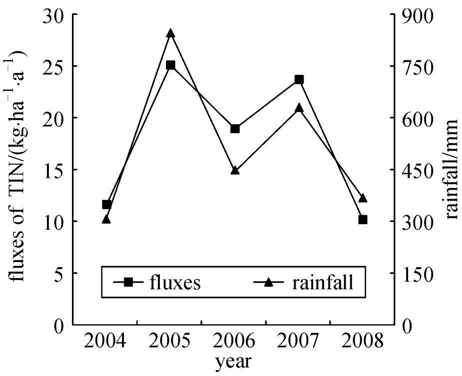

Fig. 3 Change of fluxes of TIN with volume of rainfall among years

value of 517.42 mm. The fluxes of TIN increased with the rainfall (Fig. 4, $R^2 = 0.829$). Fluxes of TIN ranged from 10.24 to 25.17 kg N/(hm^2·a), with a mean value of 17.90 kg N/(hm^2·a) which was lower than that in southern areas in China (Wang et al., 2004; Mei and Zhang, 2007). The average amount of fertilizer used for the agro-ecosystem in Liaoning Province was 150 kg N/(hm^2·a) (Liaoning Province Statistical Bureau, 2006), while the nitrogen caused by atmospheric deposition in our study accounted for 11.93% of the total fertilization which showed a considerable amount. Rapid urbanization has caused large-scale usage of automobiles, which has increased the release of nitrate nitrogen to the atmosphere, and the growth of population promotes the demand for food, which has led to more production of crops as well as more NH_3 being released from increased use of fertilizers. The mean flux of TIN (17.90 kg N/(hm^2·a)) was equal to 38.35 kg N/(hm^2·a) usage of CO (NH) $_2$, while the largest flux 25.17 kg N/(hm^2·a) in 2005 was equal to 53.94 kg N/(hm^2·a) of $CO(NH)_2$. In China, nitrogen utilization efficiencies by dominant crops range from 28% to 41% with a mean value of 35%, so the nitrogen in the rainfall input to the ecosystem is a major source of nitrogen.

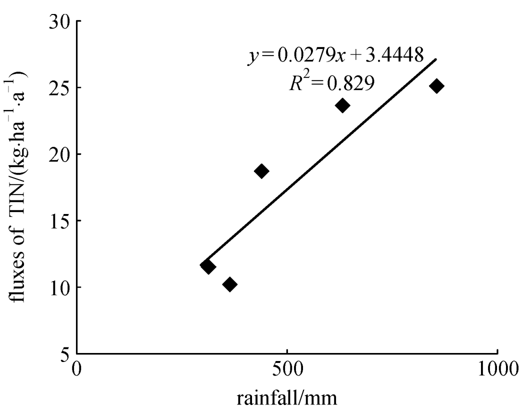

Fig. 4 Relationship between rainfall and fluxes of TIN in wet deposition

3.4 Seasonal variation of rainfall and fluxes of TIN

Rainfalls in spring and summer were bigger than in the other two seasons (Fig. 5) with the peak value in summer and the smallest one in winter. In summer, high mean temperature increased evapotranspiration, and the rainfall increased. The maximum was 398.34 mm in the summer of 2005, while the minimum was 0 mm in the winter of 2004. Summer rainfall accounted for about 56.44% of total annual precipitation. At the large scale, different climate regime (temperature and precipitation) is considered to affect the N cycling rate and, subsequently, the response to anthropogenic N inputs (Hall and Matson, 2003; Lohse and Matson, 2005). In summer, high water fluxes during rain days might favor leaching of deposition N, which led to litter N precipitation in autumn, whereas in winter deposition, N was most likely retained throughout the dry season, which led to more N precipitation in spring. The fluxes of TIN also showed a different seasonal fluctuation during the course of our study (Fig. 6). The highest fluxes were in summer which accounted for about 40.06% of total annual deposition flux, and then decreased considerably after the rainy season and reached the lowest levels in winter. The mean volumes of rainfall in four seasons were 139.37, 292.05, 78.17, and 15.01 mm. The mean fluxes of TIN were 6.37, 7.17, 2.98, and 1.39 kg N/(hm^2·a).

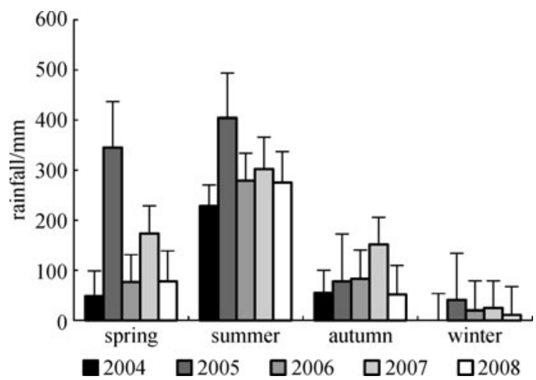

Fig. 5 Variation of rainfall among seasons

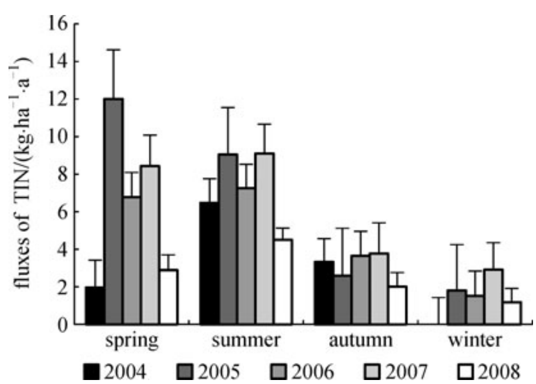

Fig. 6 Variation of fluxes of TIN among seasons

4 Conclusions

Human activities have dramatically increased the mobility and deposition of reactive forms of N (Galloway et al., 2003). Emissions of N pollutants result in N enrichment and altered chemical composition of the atmosphere, hydrologic system, soil, and biota of recipient ecosystems (Fenn et al., 2003). Current information suggests that a critical load of 5–10 kg N/($hm^2 \cdot a$) of total N deposition (both dry and wet deposition combined with all atmospheric N species) would protect the most vulnerable terrestrial ecosystems (heaths, bogs, cryptogams), and values of 10–20 kg N/($hm^2 \cdot a$) would protect forest, depending on soil condition (Krupa, 2003). The concentration of TIN decreases significantly with the increase in rainfall. The annual rainfall volume was 517.42 mm, and the fluxes of TIN increased with the rainfall. The annual mean flux of TIN was 17.90 kg N/($hm^2 \cdot a$), equal to 38.35 kg N/($hm^2 \cdot a$) usage of $CO(NH)_2$, which was a considerable amount of nitrogen fertilizers. Rainfall was mainly centered in spring and summer with a peak value in summer and the lowest one in winter. The maximum was 398.34 mm in the summer of 2005 with a flux of TIN of 8.97 kg N/($hm^2 \cdot a$), and the minimum was 0 mm in the winter of 2004.

N input by deposition plays an important role in N recharge and N compensation to the farmland ecosystem (Wang et al., 2004). Because the inorganic-N depositions by rainfall are all available, and they are easy to be used by plants, we should take into account this part of N in rain (Sun et al., 2006).

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