

Nitrogen Deposition and Its Spatial Pattern in Main Forest Ecosystems along North-South Transect of Eastern China

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Abstract: A continuous three-year observation (from May 2008 to April 2011) was conducted to characterize the spatial variation of dissolved inorganic nitrogen (DIN) deposition at eight main forest ecosystems along the north-south transect of eastern China (NSTEC). The results show that both throughfall DIN deposition and bulk DIN deposition increase from north to south along the NSTEC. Throughfall DIN deposition varies greatly from 2.7 kg N/(ha·yr) to 33.0 kg N/(ha·yr), with an average of 10.6 kg N/(ha·yr), and bulk DIN deposition ranges from 4.1 kg N/(ha·yr) to 25.4 kg N/(ha·yr), with an average of 9.8 kg N/(ha·yr). $\text{NH}_4^+\text{-N}$ is the dominant form of DIN deposition at most sampling sites. Additionally, the spatial variation of DIN deposition is controlled mainly by precipitation. Moreover, in the northern part of the NSTEC, bulk DIN deposition is 17% higher than throughfall DIN deposition, whereas the trend is opposite in the southern part of the NSTEC. The results demonstrate that DIN deposition would likely threaten the forest ecosystems along the NSTEC, compared with the critical loads (CL) of N deposition, and DIN deposition in this region is mostly controlled by agricultural activities rather than industrial activities or transportation.

Keywords: forest ecosystem; nitrogen deposition; $\text{NH}_4^+\text{-N}$; $\text{NO}_3\text{-N}$; eastern China

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1 Introduction

Atmospheric nitrogen (N) deposition has cascaded in recent decades due to extensive use of fossil fuels in industry and transportation, heavy application of fertilizers in agriculture, and expansion of animal husbandry (Galloway *et al.*, 2004; 2008). The current global atmospheric N deposition is about 25–40 Tg N/yr (Neff *et*

al., 2002), and it is expected to double in the next 25 years (Lamarque *et al.*, 2005). In China, total NO_x emission increased from 8.4 Tg N/yr in 1990 to 11.3 Tg N/yr in 2000, and total NH_3 emission increased from 10.8 Tg N/yr to 13.6 Tg N/yr over the same time period (Lu and Tian, 2007). Elevated N deposition to terrestrial ecosystems may lead to N saturation and cause a great deal of ecological risk (Matson *et al.*, 2002), such as

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eutrophication of water bodies (Gao *et al.*, 2007), soil acidification (Bouwman *et al.*, 2002), plant nutrient imbalances, and undesirable changes in biodiversity (Stevens *et al.*, 2010).

Forest ecosystems are commonly considered to be N deficient and sensitive to sharply increased N deposition (Aber and Magill, 2004). There has been widespread concern worldwide about the effect of increasing N deposition on forest ecosystems, and researchers have confirmed that elevated N deposition to forest ecosystems has resulted in N saturation in many sites (MacDonald *et al.*, 2002; Kristensen *et al.*, 2004; Dise *et al.*, 2009). However, research on N deposition of forest ecosystems in China has mainly focused on heavily polluted areas (Fan *et al.*, 2009; Fang *et al.*, 2011), and little information about forest ecosystems across a wide geographical region following the same protocol is available. Moreover, the observation of N deposition has been conducted conventionally by collecting rainfall, but this method has many disadvantages. Ion-exchange resin (IER) is a useful alternative method to collecting N deposition, which can work well in remote stations (Fenn and Poth, 2004). Sheng *et al.* (2012) reported the initial results for dissolved inorganic nitrogen (DIN) deposition in main forest ecosystems along the north-south transect of eastern China (NSTEC) using the samples collected by the IER method. The results showed that N deposition had significant seasonal variability and increased from north to south. However, there were some uncertainties due to the short-term nature of the monitoring. In the current study, DIN deposition of eight forest ecosystems along the NSTEC was measured continuously for three years (from May 2008 to April 2011). This study therefore presents a more comprehensive assessment of DIN deposition in Chinese forest ecosystems.

Our objectives were: 1) to characterize the spatial pattern of DIN deposition and its components in throughfall and precipitation, and 2) to examine the change of DIN deposition in precipitation (bulk DIN) after interaction with the canopy. We discuss how DIN deposition and its components observed in main forest ecosystems along the NSTEC differed from those in other researches. On the basis of these findings, we have estimated and discussed total atmospheric N deposition, which includes wet deposition and dry deposition. Quantifying atmospheric N deposition is important for

understanding the N status of forest ecosystems and searching for suitable strategies for N pollution abatement.

2 Materials and Methods

2.1 Site description

Located in the East Asian monsoon region, the NSTEC provides an ideal platform for studying the spatial pattern of DIN deposition in Chinese forest ecosystems, because this region lies in a concentrated distribution area of forest (Peng *et al.*, 2002) and spans a wide range of environmental conditions. The NSTEC stretches from Hainan Island to the northern border of China, with a spatial distance of more than 3700 km, ranging from 108°E to 118°E for latitude less than 40°N and from 118°E to 128°E for latitude equal to or greater than 40°N (Fig. 1). The NSTEC embraces 25 provincial level administrative unites, and covers nearly 1/3 of the territory of China (Fig. 1). Due to the influence of the East Asian monsoon, the climate of the NSTEC has apparent latitudinal gradients for temperature and precipitation, and the great spatial variations in climate are the driving forces for the diverse distribution of forest ecosystems along the transect. Zonal forest ecosystems along the NSTEC include cold-temperate coniferous forest, temperate mixed forest, warm-temperate deciduous broad-leaved forest, subtropical evergreen broad-leaved forest, and tropical monsoon rainforest from north to south (Zhang and Yang, 1995).

In this study, eight monitoring sites along the NSTEC were chosen to quantify DIN deposition in throughfall and precipitation, including Huzhong (HZ) of Heilongjiang Province, Genhe (GH) of Inner Mongolia, Mao'ershan (MES) of Heilongjiang Province, Changbaishan (CBS) of Jilin Province, Daganshan (DGS) of Jiangxi Province, Huitong (HT) of Hunan Province, Qiangyanzhou (QYZ) of Jiangxi Province, and Dinghushan (DHS) of Guangdong Province, of which HZ, GH, MES, and CBS lie in the northern part of the transect, and the others belong to the southern part of the transect. Details for all the sites are listed in Table 1, and geographical distribution of the sites is mapped in Fig. 1.

2.2 Sampling

Sheng *et al.* (2012) measured the DIN deposition in precipitation by the conventional method as well as the IER

Table 1 Site characteristics along north-south transect of eastern China (NSTEC)

Site	Longitude (E°)	Latitude (N°)	MAT (°C)	MAP (mm)	Forest ecosystem
HZ	123.0	51.8	-5.6	525.8	Deciduous coniferous forest
GH	121.5	50.8	-5.1	466.7	Deciduous coniferous forest
MES	127.5	45.4	1.1	663.6	Mixed conifer and broadleaved forest
CBS	128.1	42.4	3.0	714.1	Mixed conifer and broadleaved forest
DGS	114.5	27.5	17.0	1633.7	Evergreen broadleaved forest
HT	109.4	26.7	15.6	1393.6	Evergreen coniferous forest
QYZ	115.1	26.1	17.1	1675.1	Evergreen coniferous forest
DHS	112.5	23.2	22.1	1771.1	Evergreen broadleaved forest

Notes: HZ, Huzhong; GH, Genhe; MES, Mao'ershan; CBS, Changbaishan; DGS, Dagangshan; HT, Huitong; QYZ, Qiangyanzhou; and DHS, Dinghushan. MAT, Mean annual temperature; MAP, Mean annual precipitation

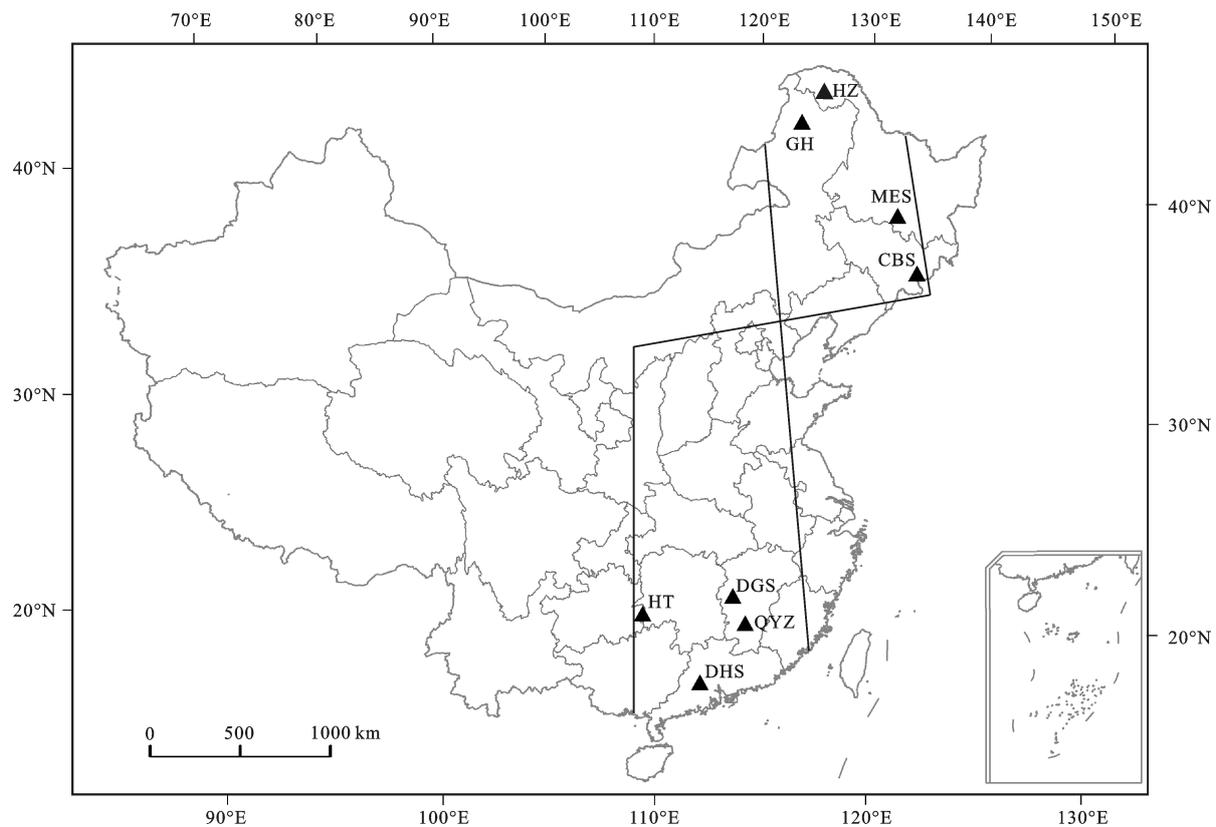


Fig. 1 Distribution of monitoring sites along north-south transect of eastern China (NSTEC). Abbreviations are same to those in Table 1. Region lies in two lines is area of NSTEC

columns, and the results showed that there was a significant linear relationship between the monthly DIN deposition observed by the two methods, and the fitting curve was nearly to the 1 : 1 line. Therefore, we concluded that the IER technique could work well in monitoring DIN deposition in field observations. Additionally, the IER technique is superior to conventional collection methods (i.e., traditional rainfall collection), which are labor intensive and analytically expensive to

implement at broad scales (Fenn and Poth, 2004; Simkin *et al.*, 2004).

In our study, IER columns were used to evaluate DIN deposition in throughfall and precipitation at eight forest ecosystems along the study transect. Information about the design of IER columns was illustrated in detail in the prior study of Sheng *et al.* (2012). At each forest monitoring site, five IER columns were randomly placed under the canopy to measure throughfall DIN deposition

and five others in an open area to collect bulk DIN deposition. Meanwhile, two IER columns with both ends sealed were installed to determine the background N in the ion resin (Fenn and Poth, 2004). Background N in the resin, although minimal, should be subtracted from the deposition data to determine the actual DIN deposition. The ionic bonds between nitrate or ammoniac ions and the charged exchange sites on the resin produce a sample that is chemically more stable than that which would have been produced by ions in solution. This allows for monthly rather than event-based sampling (Fenn *et al.*, 2002); therefore, the columns were retrieved and new ones were installed monthly from May 2008 to April 2011. All the IER columns were placed 1 m high above the ground to avoid litter, debris, or other influences from the ground. Moreover, it should be noted that, due to serious weather conditions, one value of DIN deposition could be obtained during the non-growing season at the stations at HZ, GH, MES and CBS. It is well known that ion-exchange resin will lose its potency in cold winter weather; therefore, a conventional rainwater-collection method was conducted at the four sites in winter. We used a plastic bucket to collect rainfall, or snow in winter, from November to April of the following year. At each site, five plastic buckets were randomly placed under the canopy and five others in an open area. All the plastic buckets were stabilized by stakes and placed 1 m high above the ground like the IER columns.

2.3 Chemical analyses

At the end of each field sampling, the resin columns were unscrewed from the funnel assembly, sealed at both ends, and returned to the laboratory. The resin from each column was well mixed and was extracted three times with 100 mL 0.2 mol/L KCl solution. The preliminary laboratory test with resin columns that were preloaded with a simulated deposition solution was performed by Sheng *et al.* (2012) to test the absorption efficiency of the mixed resins and the recovery efficiency of the preloaded resins. The results showed that the absorption efficiency was more than 99.0% for $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$, and the recovery efficiency was 90.9%–100.0% for $\text{NH}_4^+\text{-N}$ and 90.3%–95.5% for $\text{NO}_3^-\text{-N}$ after three KCl extractions. Therefore, this procedure was repeated three times for each column. All the extracts and snow samples were frozen at 20°C before analysis

and then analyzed by a continuous flow analyzer (BRAN+LUEBBE, AACE3, Germany). The detection limits of $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ were below 0.05 mg N/L, and the relative standard deviation was within 1%.

2.4 Calculation of DIN deposition

The monthly DIN deposition of $\text{NH}_4^+\text{-N}$ and $\text{NO}_3^-\text{-N}$ collected by IER columns was calculated according to the following formula (Sheng *et al.*, 2012):

$$DIN_{\text{mon}} = C_{\text{ex}} \times V_{\text{ex}} / 100A \quad (1)$$

where DIN_{mon} is DIN deposition per month (kg N/(ha·month)); C_{ex} is N ($\text{NH}_4^+\text{-N}$ or $\text{NO}_3^-\text{-N}$) concentration (mg N/L) in the extract; V_{ex} is the volume of the extract (L); A is the area of funnel (m^2); and 100 is a unit conversion factor.

DIN deposition in winter was calculated by multiplying the amount of precipitation by N ($\text{NH}_4^+\text{-N}$ or $\text{NO}_3^-\text{-N}$) concentration in winter. This can be described as Equation (2) (Yu *et al.*, 2011):

$$DIN_{\text{win}} = C_{\text{win}} \times P_{\text{win}} / 100 \quad (2)$$

where DIN_{win} is DIN deposition in winter (kg N/(ha·winter)); C_{win} is N ($\text{NH}_4^+\text{-N}$ or $\text{NO}_3^-\text{-N}$) concentration (mg N/L) in winter; P_{win} is the precipitation amount (mm) in winter; and 100 is a unit conversion factor.

2.5 Statistical analysis

One-way analysis of variance (ANOVA) was selected to evaluate the differences of DIN deposition among different sampling sites, and followed by Fisher's Least Significant Difference (LSD) comparisons when the differences were significant. Correlation analysis with a two-tailed significance test was used to examine the relationships between N variables across the study transect. Statistical significance was measured at the 95% confidence level unless otherwise stated. All the analyses were conducted by using SPSS 13.0 statistical software (SPSS Inc., Chicago, IL, USA). Figures were created by using SigmaPlot 13.0 software.

3 Results and Analyses

3.1 Statistics and latitudinal pattern of DIN deposition

Throughfall DIN deposition varied greatly from 2.7 kg N/(ha·yr) to 33.0 kg N/(ha·yr), with an average of 10.6 kg N/(ha·yr), of which $\text{NH}_4^+\text{-N}$ deposition ranged sub-

stantially from 1.9 kg N/(ha·yr) to 22.0 kg N/(ha·yr), whereas NO₃⁻-N deposition varied from 0.9 kg N/(ha·yr) to 11.0 kg N/(ha·yr). Average NH₄⁺-N and NO₃⁻-N deposition were 6.9 kg N/(ha·yr) and 3.7 kg N/(ha·yr), respectively. Bulk DIN deposition ranged from 4.1 kg N/(ha·yr) to 25.4 kg N/(ha·yr), with an average of 9.8 kg N/(ha·yr). NH₄⁺-N deposition ranged from 2.8 kg N/(ha·yr) to 17.9 kg N/(ha·yr), and NO₃⁻-N deposition ranged from 1.3 kg N/(ha·yr) to 7.5 kg N/(ha·yr). Throughfall and bulk DIN deposition both were significantly higher in DHS than those in any other sites, with the lowest values in HZ. Moreover, NH₄⁺-N was the dominant form both in throughfall and bulk DIN deposition at most sampling sites, and NH₄⁺-N/ NO₃⁻-N (mass/mass) was about 2 in forest ecosystems along the NSTEC (Table 2).

The spatial patterns of throughfall and bulk DIN

deposition were similar, and both of them exhibited significant latitudinal patterns, which declined linearly with the increase in latitude (Fig. 2). These patterns were generally similar when different forms of DIN deposition were examined individually (Fig. 2).

3.2 Relationships of different forms of DIN deposition

At most sampling sites, NH₄⁺-N was the dominant form of DIN deposition both in throughfall and precipitation (Table 2, Fig. 3a, 3b). Specifically, NH₄⁺-N accounted for 68% of DIN deposition (Fig. 3a), and variation of NH₄⁺-N could explain 98% of the variability of DIN deposition in throughfall among the sampling sites (Fig. 3a), whereas, NO₃⁻-N accounted for, on average, 32% of DIN deposition, and represented 93% of the variability of DIN deposition in throughfall (Fig. 3a). These trends

Table 2 Mean values of throughfall dissolved inorganic nitrogen (DIN) and bulk DIN deposition (standard errors in parentheses) in main forest ecosystems along north-south transect of eastern China (NSTEC) from May 2008 to April 2011

Site	Throughfall DIN deposition (kg N/(ha·yr))			Bulk DIN deposition (kg N/(ha·yr))		
	NH ₄ ⁺ -N	NO ₃ ⁻ -N	Total	NH ₄ ⁺ -N	NO ₃ ⁻ -N	Total
HZ	1.9(0.6) ^c	0.9(0.3) ^d	2.7(0.9) ^d	2.8(0.6) ^b	1.3(0.3) ^c	4.1(0.9) ^b
GH	2.0(0.6) ^c	1.1(0.5) ^d	3.1(1.0) ^d	3.0(0.4) ^b	1.8(0.5) ^c	4.8(0.3) ^b
MES	3.7(0.5) ^c	3.7(0.3) ^{bc}	7.4(0.3) ^c	6.7(1.5) ^b	3.8(0.3) ^b	10.5(1.8) ^b
CBS	2.8(1.1) ^c	1.8(0.1) ^{cd}	4.6(1.0) ^{cd}	3.0(0.8) ^b	2.3(0.6) ^{bc}	5.3(1.2) ^b
DGS	6.6(1.3) ^{bc}	2.8(0.1) ^c	9.3(1.4) ^{bc}	6.1(0.4) ^b	2.7(0.4) ^{bc}	8.8(0.8) ^b
HT	7.9(1.1) ^b	3.7(0.5) ^{bc}	11.6(0.7) ^b	7.3(0.7) ^{bc}	2.3(0.2) ^{bc}	9.6(0.8) ^b
QYZ	8.2(1.6) ^b	4.4(0.4) ^b	12.6(1.5) ^b	6.4(1.1) ^b	2.9(0.5) ^{bc}	9.4(1.3) ^b
DHS	22.0(2.3) ^a	11.0(0.4) ^a	33.0(2.3) ^a	17.9(4.0) ^a	7.5(1.4) ^a	25.4(5.1) ^a
Average	6.9(1.4)	3.7(0.7)	10.6(2.0)	6.7(1.1)	3.1(0.4)	9.8(1.5)

Notes: Abbreviations of sites are same to those in Table 1. Different lowercase letters in each column indicate significant differences in NO₃⁻-N, NH₄⁺-N and DIN among forest ecosystems, respectively (one-way ANOVA with Fisher's LSD, *p* < 0.05)

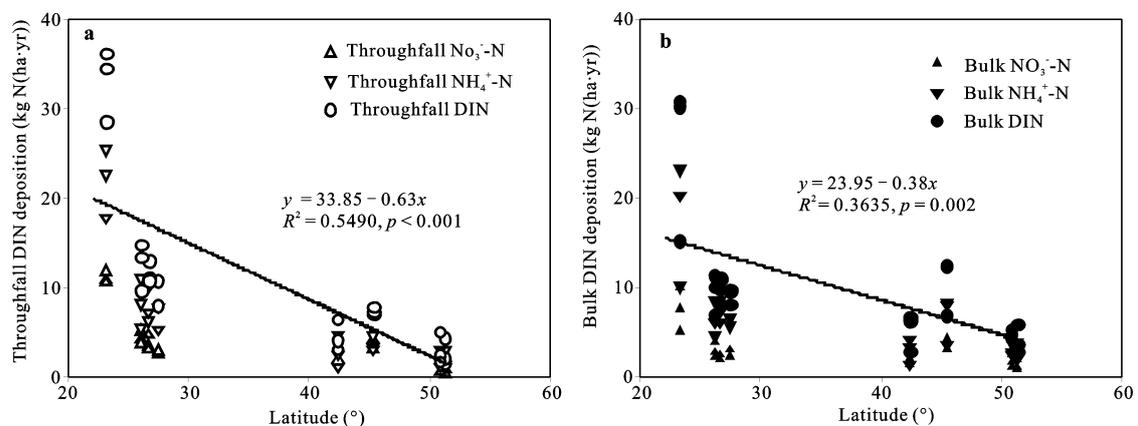


Fig. 2 Latitudinal patterns of throughfall DIN deposition (a) and bulk DIN deposition (b) in main forest ecosystems along north-south transect of eastern China (NSTEC)

were true in analyses using throughfall DIN deposition as well as analyses using bulk DIN deposition. $\text{NH}_4^+\text{-N}$ accounted for 73% of bulk DIN deposition, and $\text{NO}_3^-\text{-N}$ accounted for the remaining 27% (Fig. 3b). Furthermore, $\text{NH}_4^+\text{-N}$ is correlated significantly with $\text{NO}_3^-\text{-N}$ both in throughfall and precipitation (Fig. 3c), suggesting that high $\text{NH}_4^+\text{-N}$ deposition sites usually had high $\text{NO}_3^-\text{-N}$ deposition.

In the northern part of the NSTEC, bulk DIN deposition was, on average, 17% higher than throughfall DIN deposition. In contrast, bulk DIN deposition was 12% lower than throughfall DIN deposition in the southern part of the NSTEC (Fig. 3d). In fact, throughfall DIN deposition increased substantially with the increase of bulk DIN deposition in forest ecosystems along the NSTEC (Fig. 3d), indicating that throughfall DIN deposition is robustly correlated with bulk DIN deposition.

3.3 Relationships between mean annual precipitation and DIN deposition

Throughfall DIN deposition and bulk DIN deposition in main forest ecosystems along the NSTEC were closely

related to mean annual precipitation (MAP), and both increased linearly with MAP. MAP could account for 68% variation in throughfall DIN deposition (Fig. 4a), and 48% variation in bulk DIN deposition (Fig. 4b). These trends were true in analyses using DIN deposition as well as analyses using different forms of DIN deposition individually (Fig. 4).

4 Discussion

4.1 Spatial variation of DIN deposition

Remarkable spatial variation of DIN deposition was observed in a number of research studies (Larssen *et al.*, 2006; Chen and Mulder, 2007; Zhang *et al.*, 2008). In the present study, DIN deposition had a significant latitudinal pattern (Fig. 2), and its variation could be explained mainly by precipitation (Fig. 4). Therefore, precipitation is considered of paramount importance to the spatial pattern of DIN deposition. Our findings in this paper were supported by the results proposed by Zhao *et al.* (2009), Yu *et al.* (2011), and so on. In our study, throughfall DIN deposition ranged greatly from 2.7

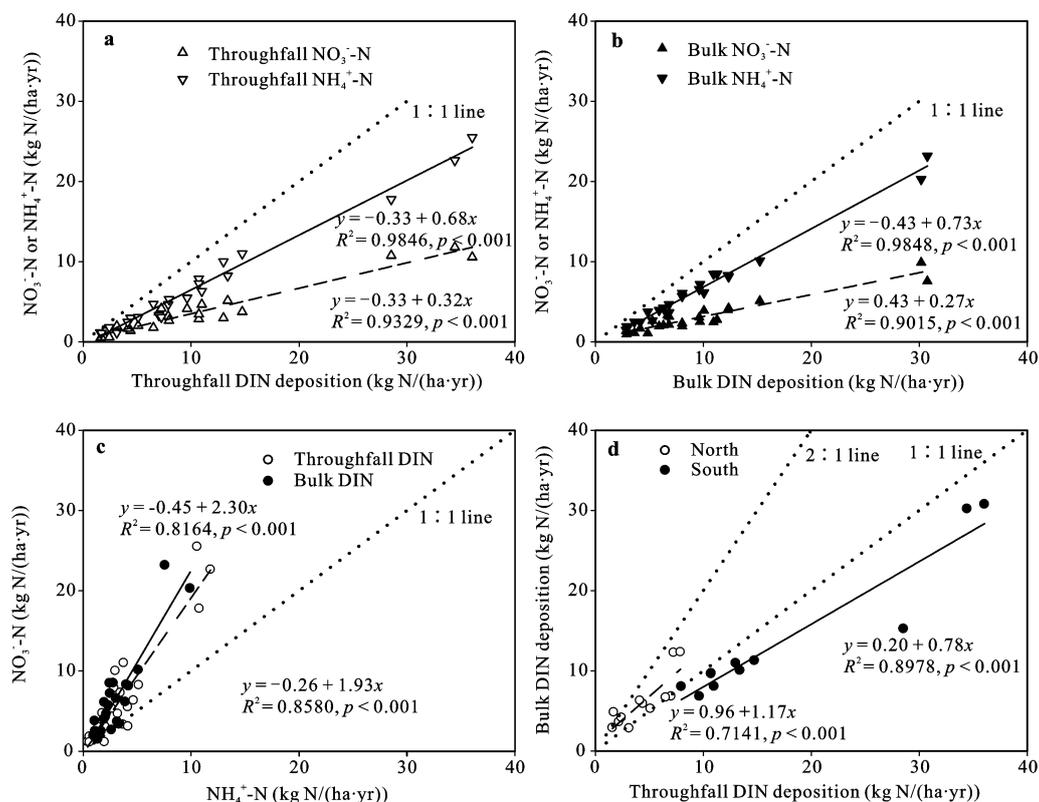


Fig. 3 DIN deposition versus $\text{NO}_3^-\text{-N}$ and $\text{NH}_4^+\text{-N}$ in throughfall (a) and precipitation (b), $\text{NO}_3^-\text{-N}$ versus $\text{NH}_4^+\text{-N}$ in throughfall and precipitation (c), and throughfall DIN deposition versus bulk DIN deposition (d) in main forest ecosystems along north-south transect of eastern China (NSTEC)

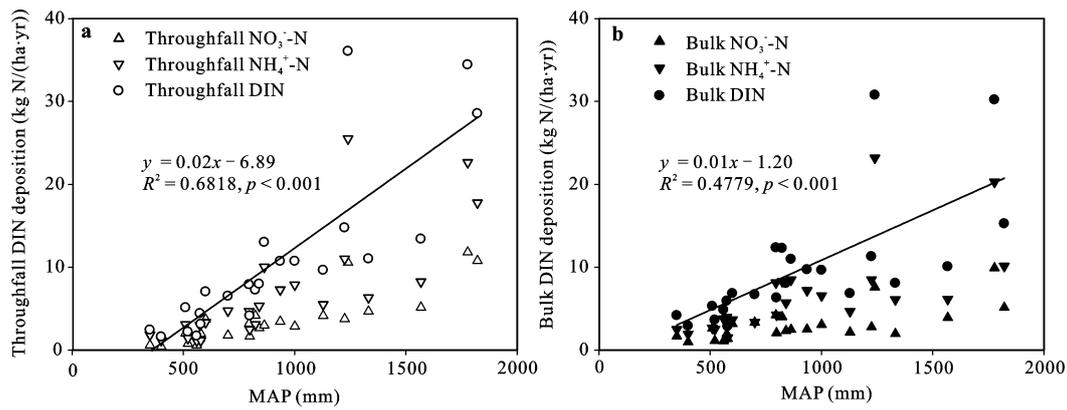


Fig. 4 Relationships between mean annual precipitation (MAP) and throughfall DIN deposition (a) and bulk DIN deposition (b) in main forest ecosystems along north-south transect of eastern China (NSTEC)

kg N/(ha·yr) to 33.0 kg N/(ha·yr), which lies in the range compiled for Chinese forest ecosystems (from 2.8 kg N/(ha·yr) to 71.0 kg N/(ha·yr)) (Fang *et al.*, 2011). Bulk DIN deposition in this study was about 1.7 times that estimated in Japanese forest ecosystems (5.9 kg N/(ha·yr)) (Mitchell *et al.*, 1997), and compared well with the level estimated by Lu and Tian (2007) based on a mapping exercise for the whole of China, but bulk DIN deposition in this study was distinguished by being lower than the previously estimated average value of 16.6 kg N/(ha·yr) based on a compiled analysis in Chinese forest ecosystems (Fang *et al.*, 2011) (Table 3). The higher bulk DIN deposition in Chinese forest ecosystems based on a compiled analysis compared with the estimation in our study may be because of the uneven distribution of investigated forest sites in previous studies that documented many sites in areas where the air was heavily polluted with N.

Throughfall DIN deposition is empirically correlated with bulk DIN deposition, but there is no general knowledge of the relationship in different ecosystems or different regions. Our results showed that bulk DIN

deposition was higher than throughfall DIN deposition in the northern part of the NSTEC, and the trend was opposite in the southern part of the NSTEC (Fig. 3d). Bulk DIN deposition is likely to be taken up by leaves through interactions with forest canopies, particularly at low N deposition sites (Lovett and Lindberg, 1993); therefore, throughfall DIN deposition tended to be lower than bulk DIN deposition in the northern part of the NSTEC. However, in areas with significant fog or particulates, such as the southern part of the NSTEC, forest canopies are efficient traps of gases and particulates from the air; therefore, throughfall DIN deposition is usually substantially larger than bulk DIN deposition (Fenn and Poth, 2004; Fenn *et al.*, 2008).

4.2 Spatial variation of NH₄⁺-N/NO₃⁻-N

DIN deposition is present mainly as NH₄⁺-N and NO₃⁻-N. NH₄⁺-N exists as a result of the dissolution of atmospheric NH₃ and scavenging of NH₄⁺-N aerosols. The main anthropogenic sources include human and animal excrement, volatilizing of fertilizer, and biomass burning, which are closely related with agricultural activities

Table 3 Comparison of bulk DIN deposition reported in different studies

Site	NH ₄ ⁺ -N (kg N/(ha·yr))	NO ₃ ⁻ -N (kgN/(ha·yr))	DIN (kg N/(ha·yr))	Reference
NSTEC	6.7	3.1	9.8	This study
Qinghai of China	2.5	0.4	2.9	Tang, 2000
Shanghai of China	26.7	31.4	58.1	Zhang, 2006
Chinese forest	11.3	5.3	16.6	Fang <i>et al.</i> , 2011
China	7.1	2.8	9.9	Lu and Tian, 2007
Japanese forest	2.9	3.0	5.9	Mitchell <i>et al.</i> , 1997
New York of USA	2.9	4.3	7.2	Golden and Boyer, 2009
Canada	2.4	3.3	5.7	Watmough <i>et al.</i> , 2005

(Prospero *et al.*, 1996). The formation of NO_3^- -N is considerably more complex. NO_3^- -N is an end product of a series of gas-phase photochemical and heterogeneous reactions involving N oxides. The major anthropogenic sources include fossil-fuel combustion by power plants and automobiles (Gao *et al.*, 2007; Pineda Rojas and Venegas, 2010). Thus, the NH_4^+ -N/ NO_3^- -N ratio might reflect the relative contribution of reactive N from industry and transportation, agriculture, and animal husbandry to DIN deposition on the local scale, and can be used as an evaluation of the degree of industrialization (Larssen *et al.*, 2006; Zhao *et al.*, 2009).

NH_4^+ -N was the dominant form of DIN deposition, and NH_4^+ -N/ NO_3^- -N varied greatly from 1.0–3.1, with an average of 2.0 in our observation period (Table 2), which compared well with that of a compiled analysis of Chinese forest ecosystems (Fang *et al.*, 2011). In the western China, with a much lower degree of economic development, NH_4^+ -N/ NO_3^- -N is much higher, up to 6.0 (Tang *et al.*, 2000), while NH_4^+ -N/ NO_3^- -N is as low as 0.8 in the more developed regions of East China (Zhang, 2006). Compared to developed countries, the present NH_4^+ -N/ NO_3^- -N ratio in our study is still much larger than those in highly industrialized New York of USA and Canada (less than 1) (Watmough *et al.*, 2005; Golden and Boyer, 2009) (Table 3). The result suggests that NH_4^+ -N from agriculture and human and animal excrement is still the larger portion, as compared with NO_3^- -N from fossil fuel combustion in industry and transportation, and the region is more heavily influenced by agriculture rather than industry or transportation.

4.3 Uncertainty of estimated N deposition

NH_4^+ -N and NO_3^- -N are the main N forms that are readily available for organisms to incorporate into their bodies. Therefore, most researches have concentrated extensively on DIN deposition on local and regional scales, but much confusion has been caused by the use of different measuring methods. In general, DIN deposition measured by different methods had some differences, although they had similar patterns. Moreover, it should be noted that the snowpack might be blown away by wind before melting, which could cause an underestimation of DIN deposition in winter in our study. Therefore, further research is required to use some novel techniques to estimate DIN deposition precisely. Furthermore, besides DIN, organic N is also a ubiquitous

component in atmospheric N deposition. Some studies have found that organic N deposition can account for 10%–30% of wet N deposition (Neff *et al.*, 2002; Violaki *et al.*, 2010). In Chinese forest ecosystems, organic N is 7.7 kg N/(ha·yr), which constitutes 32% of total dissolved N in precipitation (Fang *et al.*, 2011). Additionally, dry N deposition can provide as large an amount of N as the wet deposition or even larger (Anatolaki and Tsitouridou, 2007; Lu and Tian, 2007). In North America, dry deposition contributes only 20%–46% of wet deposition (Ollinger *et al.*, 1993), whereas dry deposition was up to twice wet N deposition in Europe (Kristensen *et al.*, 2004). In China, dry deposition was averaged as 3.0 kg N/(ha·yr) (Lu and Tian, 2007). Consequently, the total N deposition might be greater than 20 kg N/(ha·yr) in forest ecosystems along the NSTEC.

It should be noted that elevated N deposition could cause acidification and eutrophication and considerably burden various ecosystems (Stevens *et al.*, 2004; Bowman *et al.*, 2006). The report on European forest ecosystems demonstrated that, above a threshold loading of approximately 10 kg N/(ha·yr), many sites seemed to be N saturated (MacDonald *et al.*, 2002; Kristensen *et al.*, 2004; Dise *et al.*, 2009). Fang *et al.* (2011) noted that elevated N leaching occurs in forest ecosystems when they receive throughfall DIN deposition of more than 5 kg N/(ha·yr). Available evidence suggests that the critical loads (CL) of N deposition may therefore be nearly identical at approximately 10 kg N/(ha·yr) (Aber *et al.*, 2003). Thus, forest ecosystems, particularly forest ecosystems in the southern part along the NSTEC, are subject to ecological disaster, because excessive N deposition could favor the invasion of nitrophilous plants caused by the decrease of N spatial heterogeneity (Cassidy *et al.*, 2004; Gilliam, 2006). Therefore, in order to minimize the deleterious influences of N deposition on sensitive forest ecosystems, additional reliable measurements of total N deposition are urgently required to develop a better understanding of the N deposition scenarios.

5 Conclusions

The NSTEC, one of the forests concentrated distribution areas in China, showed large spatial variation of DIN deposition, which evidently increased from north to

south. Throughfall DIN deposition and bulk DIN deposition were averaged as 10.6 kg N/(ha·yr) and 9.8 kg N/(ha·yr) respectively, and both were significantly correlated with precipitation. The ranking of DIN deposition was bulk > throughfall DIN deposition in the northern part of the NSTEC, while the trend was opposite in the southern part of the NSTEC, indicating that forest canopy is paramount importance to the spatial variation of DIN deposition. The higher $\text{NH}_4^+\text{-N}/\text{NO}_3^-\text{-N}$ (up to 2.0) compared with more developed regions suggests that DIN deposition in this region was mostly controlled by agricultural activities rather than industrial activities or transportation. This study is valuable to planners and decision-makers in their attempts to curb N emissions to the atmosphere, and to evaluate the effects of consequent N deposition on forest ecosystems.

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