Monitoring nitrogen deposition in typical forest ecosystems along a large transect in China

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Abstract The nitrogen (N) deposition fluxes were investigated in eight typical forest ecosystems along the North–South Transect of Eastern China (NSTEC; based on the ChinaFLUX network) by ion-exchange resin (IER) columns from May 2008 to April 2009. Our results demonstrated that the method of IER columns was both labor cost saving and reliable for measuring dissolved inorganic nitrogen (DIN) deposition at the remote forest stations. The deposition of DIN in the throughfall ranged from 1.3 to 29.5 kg N ha^{-1} a^{-1}, increasing from north to south along NSTEC. The relatively high average ratio of ammonium to nitrate in deposition (1.83) indicated that the N deposition along the NSTEC in China mostly originated in farming and animal husbandry rather than in industry and vehicle activities. For seasonal variability, the DIN deposition showed a single peak in the growing season in the northern part of NSTEC, while, in the southern part, it exhibited double-peaks in the early spring and the mid-summer, respectively. On the annual scale, the DIN deposition variations of the eight sites could be mainly explained by precipitation and the distances from forest stations to provincial capital cities.

Keywords Dissolved inorganic nitrogen (DIN) · Ion-exchange resin (IER) · Forest ecosystem · North–South Transect of Eastern China (NSTEC) · ChinaFLUX

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Introduction

Atmospheric nitrogen (N) deposition has increased sharply in recent decades as a result of consumption of fossil fuel, emission of industrial waste gases, excessive application of fertilizers, and rapid development of animal husbandry (Galloway et al. 2008). The current global atmospheric deposition of N is about 25–40 Tg N a^{-1} (Neff et al. 2002), and it is expected to double in the next 25 years (Lamarque et al. 2005). China, as a rapidly developing country, contributes about 20 % N mobilization to the global total, and its contribution is expected to increase significantly in the near future (Galloway et al. 1996). The increase in reactive N emissions contributes to heavy regional deposition, which has been observed in China (Liu et al. 2006).

Atmospheric N deposition is one of the most important N sources in forest ecosystem, and its sharp increase might lead to remarkable effects on the forest ecosystem functioning. There has been widespread concern about the effect of increasing N deposition on natural forest ecosystems (Fangmeier et al. 1994; Klopatek et al. 2006), because of the high sensitivity of biodiversity and productivity of these ecosystems to N input (Aber and Magill 2004).

Experiments simulating N addition have shown that N deposition would affect foliar N concentration (Mo et al. 2008), N mineralization and nitrification (Fang et al. 2008), N leaching (Fang et al. 2008), N$_2$O efflux, and CH$_4$ consumption (Zhang et al. 2008) in China's forest ecosystems. However, research on N deposition in China mainly focused on heavily polluted areas (Chen and Mulder 2007; Fang et al. 2008), and only few measurements have been conducted to clarify the spatial and temporal variations of N deposition in the forest ecosystems across a large geographical region. Therefore, it is critical to estimate N deposition in forest ecosystems in an effort to quantify the effects of N amendment to the biogeochemical cycle. Unfortunately, so far, there has not been any long-term observation network to monitor N deposition dynamic continuously in China's typical forest ecosystems in a unified way. Previously reported N deposition data were difficult to be compared because of the composition complexity, large spatial-temporal variability, and different measurement methods.

The measurement of N deposition in forest ecosystems was conventionally conducted by collecting throughfall and calculated by multiplying the aqueous volume by N concentration. However, this liquid collecting way is time consuming and labor intensive, for aqueous samples must be collected at multiple points, because of spatial heterogeneity, and withdrawn on events or weekly basis, because of the instability of aqueous samples (Fenn and Poth 2004). Moreover, many forest long-term ecological research stations in China are located in remote areas. They lack of precise measuring instruments, and sometimes even do not have the uninterrupted electronic power for the sample keeping. As a result, N deposition fluxes could hardly be measured accurately and continuously through conventional throughfall collection. Ion-exchange resin (IER) is a useful alternative method that can dramatically reduce the labor required to collect samples of N deposition in the field. This technique also can work well in remote stations where electric power is unavailable or unreliable (Fenn et al. 2002).

ChinaFLUX is an observation and research network (Yu et al. 2006) and this paper reports its first year monitoring result of dissolved inorganic nitrogen (DIN) deposition in the typical forest ecosystems in China. Our findings are based on collection over a 1-year period at eight forest stations along the North–South Transect of Eastern China (NSTEC). This is the first detailed measurement of the DIN deposition flux in forest ecosystems across a large geographical region and the first DIN deposition measurement using IER columns in China. Our unique dataset cannot only contribute to accurate estimation of regional N input budgets, but also provide basic data to model simulation in carbon (C) and N couple cycling and reduce the uncertainty in productivity evaluation under future global change scenarios. The specific objectives of this paper were: (1) to compare the N deposition measurement method of IER columns to conventional throughfall collection, (2) to quantify DIN deposition flux and characterize its seasonal variation in forest ecosystems along NSTEC, and (3) to explain the main reasons for the N depositions in the forest ecosystems.

Materials and methods

Site description

NSTEC provides an ideal platform for studying the C and N cycles of forest ecosystems in East Asia's monsoon region (Peng 2001). NSTEC extends from Hainan Island to China's northern border, ranging from longitude 108–118° E at latitude less than 40° N and

from longitude 118–128° E for latitude equal to or greater than 40° N. The transect embraces 25 provinces and covers near to one third of the China's total terrestrial territory (Fig. 1). A vegetation sequence is distributed along the NSTEC, including the cold-temperate coniferous forest, temperate mixed forests, warm-temperate deciduous broad-leaved forest, subtropical evergreen coniferous forest, evergreen broad-leaved forest, and tropical rainforest from north to south (Yu et al. 2006).

Along NSTEC, we chose eight forest sites to characterize DIN deposition in China's typical forest types, included Huzhong (HZ), Genhe (GH), Maoershan (MES), Changbaishan (CBS), Dagangshan (DGS), Huitong (HT), Qiangyanzhou (QYZ), and Dinghushan (DHS) (Fig. 1). The specific characteristics of individual stations are described in Table 1.

Sample collection

Nitrate and ammonic ions in precipitation or throughfall can be exchanged by anion IER and cation IER and locked by functional groups with opposite electronic charges firmly. The nitrate and ammonic ions could be captured from precipitation or throughfall

without collecting the aqueous samples. IER has been placed in nylon mesh bags (Klopatek et al. 2006; Susfalk and Johnson 2002) or ABS pipe (Fenn et al. 2002; Simkin et al. 2004) for adsorption of ions from atmospheric deposition.

In this study, nitrate and ammonic ions in precipitation and throughfall were captured monthly, from May 2008 to April 2009, using the IER columns (Fig. 2). The IER columns consist of a funnel attached to PVC tubes. The funnel (7-cm inner diameter) is covered with gauze to prevent intrusion of leaves and insects. The IER columns are inserted into the inner PVC tube (2-cm inner diameter). The inner tube is sealed with absorbent cotton at the bottom, and the extracted liquid could run out. The inner PVC tube is contained with an outer PVC tube (4-cm inner diameter), to which bottom a PVC sieve is attached. The IER columns were stabilized by stakes and the funnel mouths were ensured to higher than 1 m from the ground level (to avoid the effect of splashing soil). At each site, we randomly placed five IER columns in an open area to measure the N deposition in precipitation, and five others under the canopy to measure the N deposition in throughfall, according to Thimonier's design (Thimonier 1998).

Fig. 1 Forest types along North–South Transect of Eastern China (*NSTEC*) and locations of the research stations
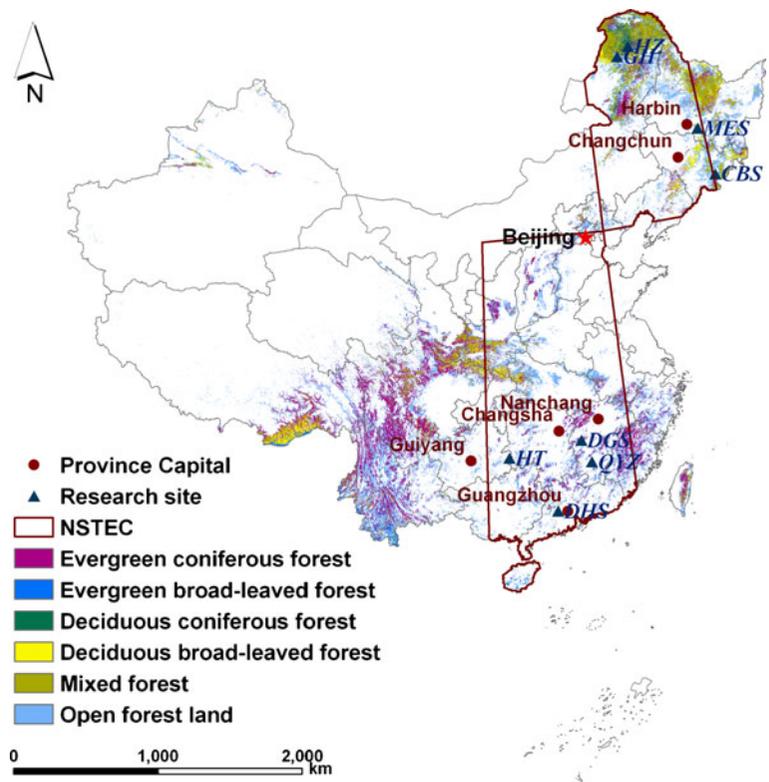
Table 1 Characteristics of the sampling stations

Site	Coordinates	Elevation (m)	Mean annul temperature (°C)	Mean annual precipitation (mm)	Forest type
Huzhong	51.3° N, 122.7° E	898	−5.6	525.8	Deciduous coniferous forest
Genhe	50.8° N, 121.5° E	805	−5.1	466.7	Deciduous coniferous forest
Maoershan	45.4° N, 127.5° E	430	1.1	663.6	Mixed conifer and broadleaved forest
Changbaishan	42.4° N, 128.1° E	736	3.0	714.1	Mixed conifer and broadleaved forest
Dagangshan	27.5° N, 114.5° E	652	17.0	1,633.7	Evergreen broadleaved forest
Huitong	26.7° N, 109.4° E	427	15.6	1,393.6	Artificial coniferous forests
Qianyanzhou	26.1° N, 115.1° E	110	17.1	1,675.1	Artificial coniferous forests
Dinghushan	23.2° N, 112.5° E	275	22.1	1,771.1	Monsoon evergreen broadleaved forest

The resin column is a mixed-bed of cation and anion IER, with 10 g of each. The ionic bonds between nitrate or ammonic 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). Based on previous N deposition studies in China and the ion exchange capacity of the resin, we determined that 20 g of the mixed-bed IER would have been excessive for our purpose (Sheng et al. 2010). We replaced the inner PVC tubes monthly. We sealed the used inner PVC tubes with plastic caps and then transported them to the Key Laboratory of Ecosystem Network Observation and Modeling in Beijing. There, the cations or anions were released back into solution from the resin columns through the use of 100 mL of 0.2 mol L^{-1} KCl solution. This procedure was repeated three times for each column. The KCl extracts were frozen until the time of analysis (Klopatek et al. 2006).

Because the forest stations were closed in winter in the north part of NSTEC due to the serious weather condition, we only got one value of DIN deposition in the whole non-growing season at the stations of HZ, GH, MES, and CBS. At the GH, QYZ, and HT forest research stations, we in parallel used a conventional rainwater-collection method synchronously to compare the results of the two methods for observing DIN deposition flux from precipitation.

Analytical methods

These extracts were then analyzed by a continuous flow analyzer (BRAN+LUEBBE, AACE3, Germany) to determine the flux of ions over the period of resin column exposure. Catalyzed by the Cu_2^+, NO_3^- was reduced by hydrazine sulfate into NO_2^-, and then it reacted with sulfanilamide in hydrochloric acid to produce a diazo compound. The diazo compound reacted on NEDD to produce a rose dye, whose concentration was detected at the wavelength of 550 nm. NH_4^+ reacted with sodium salicylate and DCI to form a blue compound, whose concentration was detected at the wavelength of 660 nm. A 0.2 mol L^{-1} KCl

Fig. 2 Designs of the ion-exchange resin (IER) columns

solution was used as the standard diluents and sampler cleaning liquid and the detection rate is 50 samples h^{-1}. The procedure detection limits for both NH_4^+ and NO_3^- were below 0.05 mg N L^{-1} with a relative error of 1 %.

Laboratory test

This study used #717 anion-exchange resin (Shanghai, China) and #732 cation-exchange resin (Shanghai, China; similar to Amberlite IRA-400 and Amberlite IR-20, respectively) to exchange the cations and anions in the precipitation or throughfall solution. Because these ions had never been reported in the study of N deposition observation, we performed laboratory test with resin columns that were preloaded with a simulated deposition solution. This allows us to test the absorption efficiency of the mixed resins and the recovery efficiency of the preloaded resins. Five columns were preloaded with the simulated solution and the experiment was repeated with similar results. The maximum simulated solution is double more than the maximum N deposition rate as recorded in the scientific literature. In the laboratory test, more than 99 % of the nitrate and ammonium was absorbed from the simulated solution (Table 2). The recovery efficiency of the preloaded resins was 90.3–95.5% for the nitrate and 90.9–100% for the ammonium (Table 3). Well, the average recovery efficiencies were above 95% for the both nitrate and the ammonium below the concentration of 2.0 mg N L^{-1}, which is the most probable NO_3^-–N or NH_4^+–N concentration in the precipitation and throughfall. It proves that our materials and methods are reliable.

We also determined the background levels of nitrate and ammonium in the IER, and the total NO_3^-–N

extracted from the blank resin column is $3.4\pm1.4\times10^{-2}$ mg N kg^{-1} mix resin and total NH_4^+–N is $4.7\pm0.3\times10^{-1}$ mg N kg^{-1} mix resin.

Data analysis

We calculated the monthly DIN deposition flux by using the following equation (Sheng et al. 2010):

$$D_{IER} = \frac{C_{ex} \times V_{ex}}{100A}$$

Where D_{IER} is the DIN deposition flux per month (in kg N ha^{-1} $month^{-1}$), C_{ex} is the N (including NO_3^-–N and NH_4^+–N) concentration in the KCl extracted solution (in mg N L^{-1}), V_{ex} is the volume of KCl solution (in L), A is the area of funnel mouth (in m^2), and 100 is the conversion factor of unit.

Statistical analyses

Sample differences of DIN deposition among sampling sites were tested with the one-way analysis of variance. Comparisons of means were conducted using the Tukey's HSD test. Regression analysis was used to examine the relationship between monthly DIN deposition fluxes from conventional method and from the IER columns, and the relationship between the annual DIN deposition flux and the distance from the research station to the nearest big city. All the analyses were conducted by SPSS software package. Statistically significant difference was set as p value of <0.05 unless stated otherwise.

Results and discussion

Method comparison

We measured the DIN deposition flux in precipitation by the conventional method as well as the IER columns at GH, HT, and QYZ forest stations, respectively. There was a significant linear relation between the monthly DIN fluxes observed by the two methods (p< 0.001), and the fitting curve was nearly to the 1:1 line (Fig. 3). Therefore, we concluded that the #717 and #732 mixed IER columns would work well in characterizing DIN deposition flux in the monthly field measurements.

Table 2 Absorption efficiency of the IER under a concentration series

Ion concentration (mg N L^{-1})	Absorption efficiency (%)	
	NO_3^-–N	NH_4^+–N
6.0	99.0±0.1	99.1±0.3
4.0	99.1±0.2	99.5±0.2
2.0	99.2±0.3	99.2±0.8
1.0	99.4±0.5	99.3±0.6
0.5	100.0±0.1	99.2±1.0

Mean (±1 SE), n=5

Table 3 Recovery efficiency of the preloaded IER extracted by 0.2 mol L^{-1} KCl

Concentration (mg N L^{-1})	NO_3^-–N recovery efficiency (%)			NH_4^+–N recovery efficiency (%)		
	After first extraction	After second extraction	After third extraction	After first extraction	After second extraction	After third extraction
6.0	33.2±0.3	77.5±0.1	93.0±0.1	67.7±0.5	76.9±0.1	90.9±0.7
4.0	32.6±0.4	77.1±0.1	92.9±0.1	67.9±0.3	77.6±0.6	92.8±0.7
2.0	31.9±0.4	76.9±0.4	96.0±2.1	70.8±0.6	84.3±0.6	95.2±2.7
1.0	31.3±1.5	76.0±0.1	95.3±2.5	77.0±1.7	93.3±5.9	98.5±1.5
0.5	34.2±1.5	76.5±0.5	95.5±0.5	86.5±4.0	94.0±0.4	100.0±0.0

Mean (±1 SE), $n=5$

DIN deposition flux

Annual DIN deposition flux of the forest ecosystems along NSTEC generally showed increasing trend from north to south (Fig. 4). Annual rainfall DIN deposition flux increased from 1.8 to 16.5 kg N ha^{-1} a^{-1} along NSTEC. Rainfall N deposition was significantly lowest at HZ, GH, and CBS ($p < 0.05$), all of which were located in the northern part of NSTEC while N deposition at DHS was remarkably higher than that at any other forest site along NSTEC ($p < 0.05$). Annual throughfall deposition flux at the HZ forest ecosystem was only 1.3 kg N ha^{-1} a^{-1}, while annual throughfall

deposition flux at DHS was 29.5 kg N ha^{-1} a^{-1}, which was 20 times more than that of HZ. The spatial pattern of throughfall deposition was similar to that of the rainfall deposition. Different from the rainfall deposition, the throughfall deposition flux at MES was significantly lower than that at any forest site in the southern part of NSTEC ($p < 0.05$), although the value was also obviously higher than that at other sites in the northern part of NSTEC ($p < 0.05$).

DIN deposition in the forest ecosystems along NSTEC except DHS, was close to the deposition level of forest ecosystems in western Canada (5 to 11 kg N ha^{-1} a^{-1}) (Kochy and Wilson 2001), northeastern of United Stated (4–8 kg N ha^{-1} a^{-1}) (Klopatek et al. 2006), and Japan (3.5–10.5 kg N ha^{-1} a^{-1}) (Mitchell et al. 1997) but was lower than that in most forest ecosystems in Europe, where the deposition value ranges from 12.3 to 61.0 kg N ha^{-1} a^{-1} (Tietema and Beier
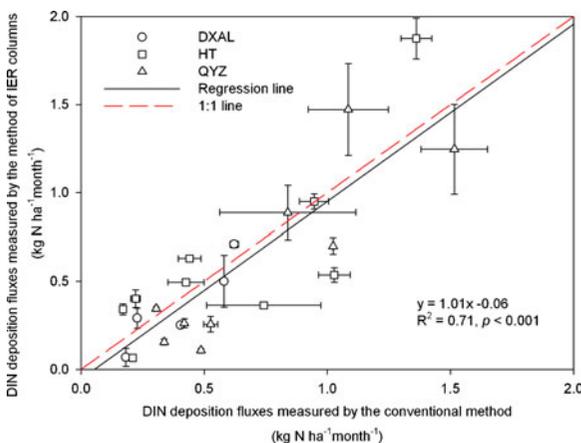
Fig. 3 Results comparison between the DIN fluxes measured by the conventional method and DIN flux by the method of IER columns. The *error bars* along abscissa axis indicate standard errors ($n=3$) of mean DIN flux measured by conventional method, and the *error bars* along vertical axis indicate standard errors ($n=5$) of mean DIN flux measured by IER columns. The *dashed line* is the 1:1 line. The parallel measurements for the period from May 2008 to September 2008 came from the GH forest station, and for the period from July 2008 to March 2009, came from the forest stations of HT and QYZ
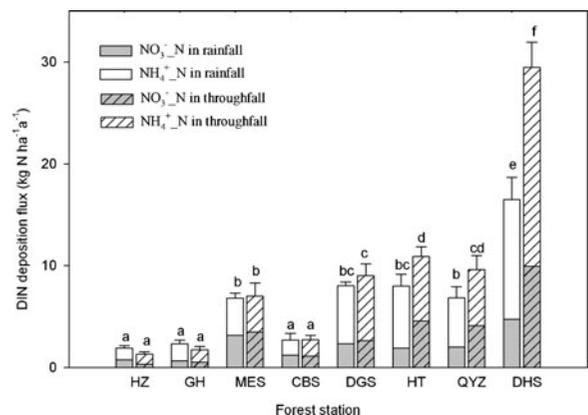
Fig. 4 DIN deposition flux in rainfall and throughfall in the forest ecosystems along NSTEC. *Error bars* indicate standard errors of the DIN deposition flux ($n=5$). The latitude of the forest station declined from left to right

1995). Moreover, even the highest value of DIN deposition in the forest ecosystems along NSTEC (observed in DHS) was just close to the middle level of DIN deposition in Europe's forest ecosystems (Tietema and Beier 1995), but was similar to the maximum value of the N deposition observation in Swiss Long-Term Forest Ecosystem Research plots (Thimonier et al. 2005). The comparisons implied that N input through throughfall in forest ecosystems in East China had exceeded that in North America and ranked the second in the world.

An ecosystem defined as N saturated when available N is in excess of total plant and microbial nutritional demand (Aber et al. 1998), resulting in above-normal sustained N losses from the ecosystem. Most studies agrees with that the forest ecosystem will retain more than 90 % of the input nitrogen when DIN deposition is below 10 kg N ha^{-1} a^{-1} while outputs are substantial when inputs are above 25 kg N ha^{-1} a^{-1} (Dise and Wright 1995; Herrmann, et al. 2005). Hereby, the forest ecosystems in the southern end of NSTEC already exposed to a risk of N saturation.

Composition of DIN deposition

Annual DIN deposition in forest ecosystems along NSTEC mainly consisted of NH_4^+–N (Fig. 4), and the annual average ratio of NH_4^+–N/NO_3^-–N ranged from 1.01 to 2.70 in different forest ecosystems. But there was no obvious trend in NH_4^+–N/NO_3^-–N along the changing of latitudes of NSTEC. The highest value of NH_4^+–N/NO_3^-–N was observed at HZ, which was located far from a large city, and the lowest value occurred at MES, a site proximate to the provincial capital city.

The average value of NH_4^+–N/NO_3^-–N in N deposition in forest ecosystems along NSTEC was 1.83, which was opposite to the research in European and North America forest ecosystems, where the value of NH_4^+–N/NO_3^-–N is 0.81 in average (Watmough et al. 2005). The NH_4^+–N/NO_3^-–N ratio in the DIN deposition could reflect the level of industrial development (Yu et al. 2011), because anthropogenic source of NH_4^+-N is mainly deriving from volatilizing of fertilizers and excrements of human and animal in the agro-ecosystems, while the major anthropogenic sources of NO_3^-–N originate from fossil fuel combustion of power plants and automobiles (Galloway et al. 2004; Jenkinson 1990). China is the largest ammonia emitter in the world with 15.2 Tg a^{-1}, and fertilizers and animal husbandry are the two main components of ammonia emission (Philippe et al. 2011). Ammonia volatilization derived from fertilizers takes up a great proportion in the loss of N from agro-ecosystems (Olivier et al. 1998). Accurately, more than 80 % of the volatilized ammonia could be transformed to NH_4^+ and deposited back into terrestrial ecosystems through precipitation in some agro-ecosystems under favorable conditions in China (Xu 1996). Therefore, different from developed countries, most forest ecosystems along NSTEC were less affected by NO_3^-–N, from fossil fuel consumption, than NH_4^+–N, from volatilizing of fertilizers and animal husbandry. However, across China, the mass ratio for NH_4^+–N/NO_3^-–N in forest ecosystems we surveyed was lower than that in agriculture stations (Liu et al. 2006).

Seasonal variation

DIN fluxes at HZ and GH during the growing season showed remarkable seasonal fluctuation, with a peak occurring in the middle of the growing season. (Fig. 5a, b). Because those two forest ecosystems are in remote mountain areas, DIN deposition might be mainly from natural release. For example, reactive N was released most intensely from the most active microbe and highly decomposed rate of animal and plant residues in summer (Ludwig et al. 2001).

DIN deposition fluxes at MES and CBS were higher in the beginning of the growing season but lower in the latter half part of the growing season (Fig. 5c, d). This might be the result of agricultural management in the around crop fields. At the beginning of the growing season, spring wheat or summer maize was planted with ground fertilizer and top-dressed about a month later. The volatilization of fertilizer might account for the higher N deposition observed at the beginning of the growing season. DIN deposition in the winter (from November to next April) accounted for 50.1 % of DIN deposition at the MES, but it accounted less than 20 % at other sites in the northern part of NSTEC (Fig. 5). The distance between MES and Harbin (the biggest city in northeastern China) is no more than 100 km, and the combustion of fuel for winter heating might be the reason for heavy deposition in winter.

DIN deposition fluxes in the four tropical forest ecosystems exhibited a double-peaks curve in 1 year's observation, with the peaks appeared in the early
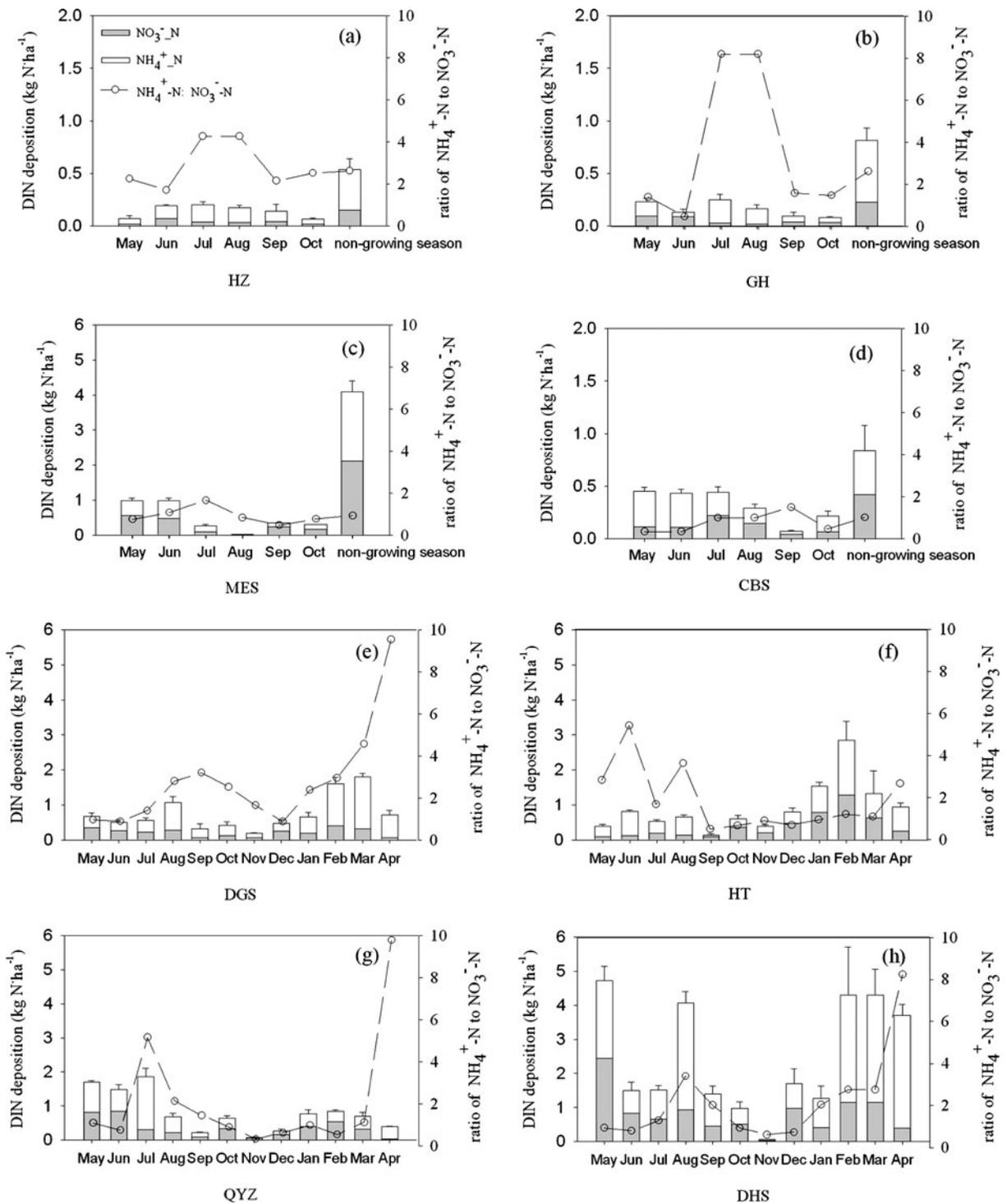
Fig. 5 Monthly variation of DIN deposition in throughfall and the ratio of NH_4^+–N to NO_3^-–N in DIN deposition in the forest ecosystems along NSTEC. The DIN deposition sampled only once in the non-growing season at the forest stations of HZ, GH, MES, and CBS. *Error bars* indicate standard errors of the monthly DIN deposition flux ($n=5$)
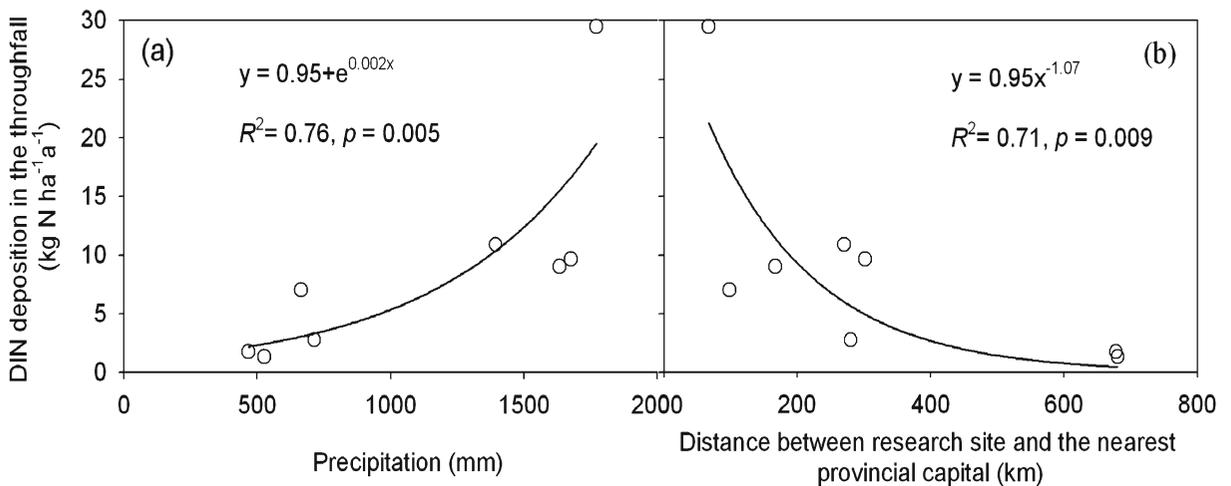
Fig. 6 The relationships between the DIN deposition flux and its impact factors in the forest ecosystems along NSTEC

spring and the mid-summer, respectively (Fig. 5e–h). The value of NH_4^+–N/NO_3^-–N was dramatically high in the spring at most research stations. This could be explained by the ammonia volatilization caused by top-dressing in the spring rice fields (Zheng et al. 2004). In rice fields, the average ammonia loss rate from top-dressed urea is up to 36 %, and the ammonia emission factor for ammonium bicarbonate is approximately 1.5 times greater that from urea (Yan et al. 2003). The second DIN deposition peak in the four tropical forest ecosystems could be the result of natural releasing of the forest ecosystem by the organic decomposition and the nitrification/denitrification processes (Ludwig et al. 2001). Soil warming stimulates decomposition and mineralization of soil organic matter resulting in releasing more reactive nitrogen. Study shows that the increased

soil temperature in the mid summer would significantly promote higher concentration of nitrate and ammonium to release (Lükewille and Wright 1997), which would contribute the heavier DIN deposition.

Factors contribution DIN deposition

The precipitation is an important factor affecting the DIN deposition flux. Previous study has reported that NH_4^+–N or NO_3^-–N concentration increased with the decrease of precipitation amount, indicating that there is cleaning effect of rainfall on suspended particle in the air (Yu et al. 2011), and this effect was controlled by below-cloud processes in the seriously polluted areas and by in-cloud processes in slightly polluted areas (Hertel et al. 2006). In our dataset, annual

Table 4 Information of the nearest capital cities of the monitoring stations

Monitoring station	Nearest capital city			Distance(km)
	Name	Location	Population (mil.)	
HZ	Harbin	126.7° E, 45.8° N	3.43	525.8
GH	Harbin	126.7° E, 45.8° N	3.43	446.7
MES	Harbin	126.7° E, 45.8° N	3.43	98.5
CBS	Changchun	125.3° E, 43.9° N	2.65	279.9
DGS	Changsha	113.0° E, 28.2° N	1.86	167.3
HT	Guiyang	106.7° E, 26.6° N	1.57	270.2
QYZ	Nachang	115.9° E, 28.7° N	1.73	301.6
DHS	Guangzhou	113.2° E, 23.1° N	6.45	67.4

Population statistics came from China Population Statistics Yearbook in 2008

precipitation accounted for 76 % of the DIN change in the throughfall along NSTEC (Fig. 6a).

The DIN deposition in forest ecosystem was also remarkable affected by the distance to big city (Table 4). We found that the distance between the research station and the provincial capital city could explain more than 70 % variance in annual DIN deposition flux (Fig. 6b). DIN deposition flux at MES and at DHS, where were proximate to big cities, was significantly higher than that at any other site in the northern or southern part of NSTEC, respectively ($p < 0.05$). Take DHS for example, the DIN deposition in its nearest capital city, Guangdong, is up to more than 40 kg N ha^{-1} a^{-1} (Huang et al. 2010), much larger than that in DHS forest ecosystem.

Besides the N oxide emission from factorier and vehicles, the dramatically increasing numbers and scales of intensive livestock and poultry farms around the big cities also contribute to the N emission in recent decades in China (Su 2006). It has been reported that ammonia emmision from livestock and poultry farms has been the most important ammonia source and could account for more than half of the globle emmsions (Schlesinger and Hartley 1993). In some European countries, livestock sector has been considered responsible for the increased emissions of ammonia which results in substantial ammonia deposition. These overfull N depostions greatly exceeds the critical loads for N and other acidifying compounds in a large part of the country (Lekkerkerk 1998).

Moreover, the dominant direction of winds is really an important factor to impacting the source of N deposition. However, due to inaccessible data and shortage of knowledge storage on aerodynamics now, the analyses of the impact of winds direction would be a part of future work. Back trajectory analysis and stable N isotope in rainwater analysis are both urgently needed in upcoming study to elucidate where and which mechanisms contributed to the atmospheric N (Yu et al. 2011). So that, we could clarify the amount and sources of reactive N coming from atmosphere to Chinese typical forest ecosystems.

Conclusions

Observation by the IER method was carried out to analyze the DIN deposition in eight forest ecosystems along NSTEC. Our results showed a significant linear relation between the monthly DIN fluxes observed by

the two methods, and demonstrated that IER columns represent a practical and reliable method for measuring DIN deposition at the remote forest stations. The DIN deposition flux ranged from 1.3 to 29.5 kg N ha^{-1} a^{-1}, increasing from north to south along this transect. In the northern part of NSTEC, the highest DIN deposition values appeared in the middle of the growing season at HZ and GH. It appeared in the beginning of the growing season at MES and CBS, due to the ground fertilizer and top-dressing on spring wheat or summer maize. However, in the southern part of NSTEC, where the growing season extends through all the year, N deposition showed peaks in the early spring and the mid-summer, respectively. Different from developed countries, the ratio of NH_4^+–N/NO_3^-–N in DIN deposition in the throughfall was >1, indicating that most forest ecosystems along NSTEC were affected by NH_4^+–N from farming and animal husbandry rather than by NO_3^-–N from fossil fuel consumption. Variation in annual DIN deposition could be explained mainly by annual precipitation and the distance from forest stations to provincial capital cities. Back trajectory analysis and stable N isotope in rainwater are both urgently needed in upcoming study to elucidate where and which mechanisms contributed to the atmospheric DIN deposition in the forest ecosystems in China.

References

Aber, J. D., & Magill, A. H. (2004). Chronic nitrogen additions at the Harvard Forest (USA): the first 15 years of a nitrogen saturation experiment. *Forest Ecology and Management, 196*, 1–5.

Aber, J. D., McDowell, W., & Nadelhoffer, K. J. (1998). Nitrogen saturation in Northern forest ecosystems, hypotheses revisited. *Bioscience, 48*, 921–934.

Chen, X., & Mulder, J. (2007). Indicators for nitrogen status and leaching in subtropical forest ecosystems, South China. *Biogeochemistry, 82,* 165–180.

Dise, N. B., & Wright, R. F. (1995). Nitrogen leaching from European forests in relation to nitrogen deposition. *Forest Ecology and Management, 71,* 153–161.

Fang, Y., Gundersen, P., Mo, J., & Zhu, W. (2008). Input and output of dissolved organic and inorganic nitrogen in subtropical forests of South China under high air pollution. *Biogeosciences, 5,* 339–352.

Fangmeier, A., Hadwiger-Fangmeier, A., Van der Eerden, L., & Jäger, H. (1994). Effects of atmospheric ammonia on vegetation—a review. *Environmental Pollution, 86,* 43–82.

Fenn, M. E., & Poth, M. A. (2004). Monitoring nitrogen deposition in throughfall using ion exchange resin columns: a field test in the San Bernardino Mountains. *Journal of Environmental Quality, 33,* 2007–2014.

Fenn, M. E., Poth, M. A., & Arbaugh, M. (2002). A throughfall collection method using mixed bed ion exchange resin columns. *The Scientific World Journal, 2,* 122–130.

Galloway, J. N., Dianwu, Z., Thomson, V. E., & Chang, L. H. (1996). Nitrogen mobilization in the United States of America and the People's Republic of China. *Atmospheric Environment, 30,* 1551–1561.

Galloway, J. N., Dentener, F. J., Capone, D. G., Boyer, E. W., Howarth, R. W., Seitzinger, S. P., et al. (2004). Nitrogen cycles: past, present, and future. *Biogeochemistry, 70,* 153–226.

Galloway, J. N., Townsen, A. R., Erisman, J. W., Mateete, B., Cai, Z., & Freney, J. R. (2008). Transformation of the nitrogen cycle: recent trends, questions and potential solutions. *Science, 320,* 889–892.

Herrmann, M., Pust, J., & Richard, P. (2005). Leaching of nitrate and ammonium in heathland and forest ecosystems in Northwest Germany under the influence of enhanced nitrogen deposition. *Plant and Soil, 273,* 129–137.

Hertel, O., Skjoth, C. A., Lofstrom, P., Geels, C., Frohn, L. M., Ellermann, T., et al. (2006). Modelling nitrogen deposition on a local scale—a review of the current state of the art. *Environmental Chemistry, 3,* 317–337.

Huang, D., Xu, Y., Zhou, B., Zhang, H., & Lan, J. (2010). Wet deposition of nitrogen and sulfur in Guangzhou, a subtropical area in South China. *Environmental Monitoring and Assessment, 171,* 429–439.

Jenkinson, D. S. (1990). An introduction to the global nitrogen-cycle. *Soil Use and Management, 6,* 56–61.

Klopatek, J. M., Barry, M. J., & Johnson, D. W. (2006). Potential canopy interception of nitrogen in the Pacific Northwest, USA. *Forest Ecology and Management, 234,* 344–354.

Kochy, M., & Wilson, S. D. (2001). Nitrogen deposition and forest expansion in the northern Great Plains. *Journal of Ecology, 89,* 807–817.

Lamarque, J. F., Kiehl, J., Brasseur, G., Butler, T., Cameron-Smith, P., Collins, W. D., et al. (2005). Assessing future nitrogen deposition and carbon cycle feedback using a multimodel approach: analysis of nitrogen deposition. *Journal of Geophysical Research-Atmospheres, 110,* D19303.1–D19303.21.

Lekkerkerk, L. J. A. (1998). Implications of Dutch ammonia policy on the livestock sector. *Atmospheric Environment, 32,* 581–587.

Liu, X., Ju, X., Zhang, Y., He, C., Kopsch, J., & Zhang, F. (2006). Nitrogen deposition in agroecosystems in the Beijing area. *Agriculture, Ecosystems & Environment, 113,* 370–377.

Ludwig, J., Meixner, F. X., Vogle, B., & Förstner, J. (2001). Soil-air exchange of nitric oxide: an overview of processes, environmental factors, and modeling studies. *Biogeochemistry, 52,* 225–257.

Lükewille, A., & Wright, R. F. (1997). Experimentally increased soil temperature causes release of nitrogen at a boreal forest catchment in southern Norway. *Global Change Biology, 3,* 13–21.

Mitchell, M., Iwatsubo, G., Ohrui, K., & Nakagawa, Y. (1997). Nitrogen saturation in Japanese forests: an evaluation. *Forest Ecology and Management, 97,* 39–51.

Mo, J., Li, D., & Gundersen, P. (2008). Seedling growth response of two tropical tree species to nitrogen deposition in southern China. *European Journal of Forest Research, 127,* 275–283.

Neff, J. C., Holland, E. A., Dentener, F. J., McDowell, W. H., & Russell, K. M. (2002). The origin, composition and rates of organic nitrogen deposition: a missing piece of the nitrogen cycle? *Biogeochemistry, 57,* 99–136.

Olivier, J. G. J., Bouwman, A. F., Van der Hoek, K. W., & Berdowski, J. J. M. (1998). Global air emission inventories for anthropogenic sources of NO_x, NH_3 and N_2O in 1990. *Environmental Pollution, 102,* 135–148.

Peng, S. (2001). Important progress in mechanism research of global change and argoecosystems interaction in eastern China. *China Basic Science, 7,* 18–20 (*in Chinese*).

Philippe, F., Cabaraux, J., & Nicks, B. (2011). Ammonia emissions from pig houses: influencing factors and mitigation techniques. *Agriculture, Ecosystems and Environment, 141,* 245–260.

Schlesinger, W. H., & Hartley, A. E. (1993). A global budget for atmospheric NH_3. *Biogeochemistry, 15,* 191–211.

Sheng, W., Yu, G., Fang, H., Liu, Y., & Hu, Z. (2010). Determination of nitrogen depoisition in throughfall using ion-exchange resins: a field test in planted conigerous forest ecosystem at Qianyanzhou. *Acta Ecologica Sinica, 30,* 6872–6880 (*in Chinese*).

Simkin, S., Lewis, D., Weathers, K., Lovett, G., & Schwarz, K. (2004). Determination of sulfate, nitrate, and chloride in throughfall using ion-exchange resins. *Water, Air, and Soil Pollution, 153,* 343–354.

Su, Y. (2006). Research of countermeasures on waste treating of intensive livestock and poultry farms in China. *Chinese Journal of Eco-Agriculture, 14,* 15–18 (*in Chinese*).

Susfalk, R. B., & Johnson, D. W. (2002). Ion exchange resin based soil solution lysimeters and snowmelt solution collectors. *Communications in Soil Science and Plant Analysis, 33,* 1261–1275.

Thimonier, A. (1998). Measurement of atmospheric deposition under forest canopies: some recommendations for equipment and sampling design. *Environmental Monitoring and Assessment, 52,* 353–387.

Thimonier, A., Schmitt, M., Waldner, P., & Rihm, B. (2005). Atmospheric deposition on Swiss Long-Term Forest Ecosystem Research plots. *Environmental Monitoring and Assessment, 104,* 81–118.

Tietema, A., & Beier, C. (1995). A correlative evaluation of nitrogen cycling in the forest ecosystems of the EC projects Nitrex and Exman. *Forest Ecology and Management, 71*, 143–151.

Watmough, S. A., Aherne, J., Alewell, C., Arp, P., Bailey, S., Clair, T., et al. (2005). Sulphate, nitrogen and base cation budgets at 21 forested catchments in Canada, the United States and Europe. *Environmental Monitoring and Assessment, 109*, 1–36.

Xu, R. (1996). Effect of the NH_4^+ in precipitation to soil acidification in China. *Agro-environmental Protection, 15*, 139–140 (in Chinese).

Yan, X., Akimoto, H., & Ohara, T. (2003). Estimation of nitrous oxide, nitric oxide and ammonia emissions from croplands in East, Sourheast, and South Asia. *Global Change Biology, 9*, 1080–1096.

Yu, G., Wen, X., Sun, X., Tanner, B. D., Lee, X., & Chen, J. (2006). Overview of ChinaFLUX and evaluation of its eddy covariance measurement. *Agricultural and Forest Meteorology, 137*, 125–137.

Yu, W., Jiang, C., Ma, Q., Xu, Y., Zou, H., & Zhang, S. (2011). Observation of the nitrogen deposition in the lower Liaohe River Plain, Northeast China and assessing its ecological risk. *Atmospheric Research, 101*, 460–468.

Zhang, W., Mo, J., Yu, G., Fang, Y., Li, D., Lu, X., et al. (2008). Emissions of nitrous oxide from three tropical forests in Southern China in response to simulated nitrogen deposition. *Plant and Soil, 306*, 221–236.

Zheng, S., Liu, D., Nie, J., Dai, P., & Xiao, J. (2004). Fate and recovery efficiency of controlled release nitrogen fertilizer in flooding paddy soil. *Plant Nutrition and Fertilizer Science, 10*, 137–142 (in Chinese).