Postprint: Responses of Leaf Nitrogen Content and 15N Values of Typical Deciduous Broad-Leaved Trees in Beijing to Atmospheric Nitrogen Deposition

Authors: Liu Chaoming, Tang Meiqing, Ma Kun, Liu Xingyun, Yu Han, Zhang Ying

Date: 2017-04-18T00:00:00+00:00

Abstract

To investigate plant responses to atmospheric nitrogen deposition and the indicative role of this nitrogen source, this study collected leaf samples from typical deciduous broadleaf tree species Populus and Salix at 198 sampling sites in the Beijing area, and measured their nitrogen content and 15N values. The results showed that leaf nitrogen content of Populus in Beijing ranged from 16.5–38.6 g/kg, with an average of (24.0±4.0) g/kg; leaf nitrogen content of Salix ranged from 17.2–36.2 g/kg, with an average of (25.9±4.1) g/kg. Within the study area, leaf nitrogen content of both Populus and Salix exhibited a diagonal distribution pattern of low in the northwest and high in the southeast, which aligns with the spatial variation of atmospheric nitrogen deposition in this region. Since there was no significant variation in climatic factors within the study area, the changes in leaf nitrogen content reflected the influence of atmospheric nitrogen deposition on plant elemental stoichiometric characteristics and plant responses to atmospheric nitrogen deposition. Leaf 15N values of Populus in Beijing ranged from -3.95‰ to 8.10‰, with an average of (1.15±2.48)‰; leaf 15N values of Salix ranged from -3.04‰ to 9.73‰, with an average of (2.31±2.60)‰. Leaf 15N values of both Populus and Salix exhibited a spatial distribution pattern of high in the northwest, high in the central region, and low in the southeast, which is opposite to the spatial distribution trend of leaf nitrogen content. The relatively high 15N values in the central urban area reflected the impact of traffic pollution on the increase of atmospheric nitrogen compounds; the relatively high 15N values in the northwest indicated that this region was less affected by anthropogenic emission sources, with natural nitrogen cycling being the main reason for its higher 15N values; the relatively low 15N values in the southeast were likely the result of combined effects from agricultural activities and traffic.

Full Text

Preamble

Acta Ecologica Sinica, ChinaXiv Partner Journal
Vol. 37, No. 7, Apr. 2017
DOI: 10.5846/stxb201511202348

Responses and Indications of Foliar Nitrogen Contents and 15N Values to Atmospheric Nitrogen Deposition in Beijing, China

LIU Chaoming, TANG Meiqing, MA Kun, LIU Xingyun, YU Han, ZHANG Ying
School of Nature Conservation, Beijing Forestry University, Beijing 100083, China

Abstract

To investigate plant responses to atmospheric nitrogen deposition and their indication of nitrogen sources, we collected and analyzed leaf samples of typical deciduous broadleaf trees (*Populus* and *Salix*) across the Beijing area. Foliar nitrogen contents and 15N values were measured to assess spatial patterns and their relationship with atmospheric nitrogen deposition.

The foliar nitrogen content of *Populus* species in Beijing ranged from 16.5 to 38.6 g/kg, with a mean of (24.0 ± 4.0) g/kg. *Salix* species showed a similar range of 17.2–36.2 g/kg, with a mean of (25.9 ± 4.1) g/kg. Both genera exhibited a distinct spatial pattern of lower nitrogen contents in the northwest and higher contents in the southeast, forming a diagonal gradient that corresponds to the spatial variation of atmospheric nitrogen deposition in the region. Climate factors showed no significant variation across the study area, indicating that the observed changes in foliar nitrogen content reflect the influence of atmospheric nitrogen deposition on plant elemental stoichiometry and demonstrate plant responses to this external nitrogen input.

For 15N values, *Populus* leaves ranged from -3.95‰ to 8.10‰, with a mean of (1.15 ± 2.48)‰, while *Salix* leaves ranged from -3.04‰ to 9.73‰, with a mean of (2.31 ± 2.60)‰. In contrast to the nitrogen content patterns, 15N values showed higher values in the city center and northwest, and lower values in the southeast. The elevated 15N values in central urban areas reflect the impact of traffic pollution on atmospheric nitrogen compounds, while the higher values in the northwest indicate minimal influence from anthropogenic emission sources, where natural nitrogen cycling is the primary driver of the enriched isotopic signature. The lower 15N values in the southeast likely result from combined effects of agricultural activities and traffic emissions.

Keywords: Deciduous broadleaf trees; foliar nitrogen content; 15N; atmospheric nitrogen deposition; Beijing area

Introduction

Atmospheric nitrogen deposition refers to the process by which reactive nitrogen compounds return from the atmosphere to terrestrial or aquatic surfaces [1]. Human activities have dramatically increased emissions of nitrogenous compounds to the atmosphere, leading to elevated nitrogen deposition globally [1-3]. Eastern China has become one of the regions with the highest atmospheric nitrogen deposition in the world since the mid-20th century, alongside the central-eastern United States and western Europe [4-5]. This reactive nitrogen deposited on Earth’ s surface may profoundly affect terrestrial and aquatic ecosystems [6-8]. For terrestrial vegetation, 50%-70% of atmospheric deposition is absorbed by plant canopies [9], and most plants can directly absorb nitrogen oxides through leaf stomata, exhibiting corresponding increases in foliar nitrogen content [10-12]. Thus, foliar nitrogen levels can serve as a biological indicator of atmospheric nitrogen deposition [13-14].

Liu et al. [15] demonstrated that increased atmospheric nitrogen deposition in China has led to higher foliar nitrogen contents in plants, though climate factors such as precipitation also influence leaf nitrogen content at large scales [16-17]. However, changes in foliar nitrogen content show significant positive correlations with corresponding increases in atmospheric nitrogen deposition [15]. Stable isotope techniques are indispensable in modern ecological research, particularly for studying ecosystem biogeochemical cycles [18-19]. Different sources of reactive nitrogen exhibit distinct 15N signatures. Ammonia volatilization (NH3) from fertilizer application and livestock excreta typically shows 15N values between -48‰ and -36.3‰ [20-21], while NOx emissions from fossil fuel combustion and vehicle exhaust show different isotopic compositions. Felix et al. [21] reported 15N values of -56‰ to -23.1‰ for ammonia volatilization samples from fertilizer use collected with passive samplers in the United States. Atmospheric particulate matter sampled near livestock sources showed 15N values of -4‰ to 22‰ [22-23]. Coal combustion emissions typically have 15N values of 2‰-15‰ [22-23], while vehicle exhaust emissions show a wider range. Ye et al. [24] found that 15N-NO3- from vehicle emissions ranges from -13‰ to -2‰ under high-speed driving with complete fuel combustion, but from 3.9‰ to 12‰ under low-speed driving with incomplete combustion [25].

Since plant leaves can intercept and absorb most atmospheric nitrogen deposition [14], their 15N values have biological significance for indicating both the flux and sources of atmospheric nitrogen deposition [26-27]. In principle, foliar 15N can be used to identify and distinguish nitrogen sources in atmospheric deposition [28-29].

Approximately 60% of Beijing’ s area lies in the North China Plain, a region of globally typical high nitrogen deposition, with total deposition reaching 100 kg N hm ^{2}, including both organic and inorganic nitrogen species from wet and dry deposition [30]. Due to its relatively small latitudinal and longitudinal range, Beijing’ s multi-year average precipitation and temperature show little variation, but atmospheric nitrogen deposition exhibits a clear pattern of lower values in the north and northwest, and higher values in the south and southeast [31]. This study conducted grid-based sampling of typical deciduous broadleaf trees (*Populus* and *Salix*) in Beijing to measure foliar nitrogen content and 15N values, thereby verifying the response of foliar nitrogen content to atmospheric nitrogen deposition independent of climate factors, and assessing its indication of nitrogen deposition sources.

1 Study Area

Beijing is located on the northern edge of the North China Plain (39°28 -41°05 N, 115°25 -117°30 E), characterized by a warm temperate semi-humid and semi-arid monsoon continental climate. The terrain extends from northeast to southwest in a diagonal distribution of mountainous and plain areas. The western region comprises the Taihang Mountains, the north features the Yanshan Mountains, and the central and southeastern areas belong to the North China Plain. Land use in the northwestern mountainous area is dominated by forests with some grasslands, while the southern region is primarily farmland. Influenced by the southern North China Plain, the wet nitrogen deposition brought by precipitation in the plain area averages 34 kg N hm ^{2}, with organic nitrogen deposition of about 7 kg N hm ^{2}, and dry nitrogen deposition also reaching high levels [26,27]. Areas dominated by urban and agricultural land show higher atmospheric nitrogen deposition, while regions with forest and grassland distribution in the north and northwest exhibit lower deposition [29].

Zhang et al. [31] simulated nitrogen deposition in the North China Plain using the FRAME model, showing that areas with high atmospheric nitrogen deposition are primarily distributed in the southern and southeastern parts of Beijing, with total deposition up to 50 kg N hm ^{2}, including approximately 27 kg N hm ^{2} of inorganic nitrogen. The spatial distribution of atmospheric nitrogen deposition in Beijing shows significant variation [29].

[Figure 1: see original paper] Land-use map and 5 km atmospheric N deposition map of Beijing area

2 Plant Sample Collection and Processing

A 5 ×5 latitude-longitude grid was used to divide the study area, with samples collected near grid intersections in topographically suitable locations. Due to mountainous terrain, some points were inaccessible. Leaf sampling was conducted in late August during peak vegetation growth and maximum biomass. At each sampling site, five mature, healthy leaves were collected from sun-exposed branches of five *Populus* and *Salix* individuals. When insufficient trees were available, the number of sampled individuals was reduced accordingly, but sufficient leaf material was ensured for analysis. After collection, samples were transported to the laboratory on the same day, oven-dried at 105°C for 30 minutes for enzyme deactivation, then dried at 65°C for 48 hours to constant weight.

Samples of each species at each site were combined and ground to pass a 1-mm sieve for analysis.

Nitrogen content was determined using the Kjeldahl digestion method, and 15N values were measured using a continuous-flow isotope ratio mass spectrometer (Thermo MAT253). Sample masses of 0.4–0.5 g were used for nitrogen content analysis, and 1.5–2 mg for stable isotope analysis.

3 Data Analysis

Statistical analysis was performed using SigmaPlot 10.0. Spatial distributions of foliar nitrogen content and 15N values were interpolated using the Inverse Distance Weighted (IDW) method in ArcGIS 10.0 (ArcMap) to generate distribution maps for the study area.

[Figure 2: see original paper] Location of sampling sites of typical deciduous broadleaf trees, *Populus* and *Salix*, in Beijing area

1 Overall Characteristics of Foliar Nitrogen Content and 15N Values in *Populus* and *Salix* in Beijing

Foliar nitrogen content in *Populus* ranged from 16.5 to 38.6 g/kg, with a mean of (24.0 ± 4.0) g/kg. *Salix* showed a range of 17.2–36.2 g/kg, with a mean of (25.9 ± 4.1) g/kg, significantly higher than *Populus*. For 15N values, *Populus* ranged from -3.95‰ to 8.10‰, with a mean of (1.15 ± 2.48)‰, while *Salix* ranged from -3.04‰ to 9.73‰, with a mean of (2.31 ± 2.60)‰, significantly higher than *Populus*.

Combined across both genera, foliar nitrogen content ranged from 16.5 to 38.6 g/kg with a mean of (24.7 ± 4.1) g/kg, while 15N values ranged from -3.95‰ to 9.73‰ with a mean of (1.56 ± 2.57)‰.

Foliar nitrogen contents and 15N values of *Populus* and *Salix* in Beijing area

Genus	Sample size (n)	N content (g/kg)	Range	15N (‰)	Range
Populus		24.0 ± 4.0 a	16.5–38.6	1.15 ± 2.48 a	-3.95–8.10
Salix		25.9 ± 4.1 b	17.2–36.2	2.31 ± 2.60 b	-3.04–9.73
Total		24.7 ± 4.1	16.5–38.6	1.56 ± 2.57	-3.95–9.73

Note: Different letters within the same column indicate significant differences in foliar nitrogen content or 15N values between genera ($P < 0.01$).

2 Spatial Distribution of Foliar Nitrogen Content and 15N Natural Abundance in *Populus* and *Salix* in Beijing

IDW interpolation revealed that foliar nitrogen content in both genera showed an overall diagonal distribution pattern of low values in the northwest and high values in the southeast. For *Populus*, higher nitrogen contents were found in the northeast and south, while lower contents occurred in the west and southwest. *Salix* showed a similar northwest-to-southeast gradient, but with higher nitrogen contents primarily in the east and south, and lower contents in the west, north, and northeast. The spatial distribution of foliar nitrogen content across both genera demonstrated higher values in the east and south, and lower values in the west and north.

In contrast, spatial interpolation of 15N values showed an opposite trend. Higher 15N values were distributed in the north, city center, and southwest, while lower values occurred in the northeast and southeast. Both *Populus* and *Salix* exhibited this northwest-high, southeast-low pattern, which is opposite to the spatial distribution of foliar nitrogen content.

[Figure 3: see original paper] Spatial distribution of foliar N contents of *Populus* and *Salix* in Beijing area
[Figure 4: see original paper] Spatial distribution of foliar 15N values of *Populus* and *Salix* in Beijing area

1 Plant Foliar Nitrogen Content Response to and Indication of Atmospheric Nitrogen Deposition

The mean foliar nitrogen content across all sampling points was (24.7 ± 4.1) g/kg, similar to values reported in other multi-species studies in Beijing and surrounding areas [32], but significantly higher than the national average for deciduous woody and broadleaf woody plants [16,33]. At large scales, foliar nitrogen content is inversely proportional to mean annual temperature and precipitation, and directly proportional to latitude [17,33]. Beijing lies in the mid-high latitude zone of China, where typical deciduous broadleaf species have higher foliar nitrogen contents than the national average, consistent with zonal characteristics.

While climate and soil factors also influence foliar nitrogen content [16-17], these show little variation across Beijing’ s relatively small geographic range. The significant spatial variation in foliar nitrogen content is therefore likely associated with external nitrogen input from atmospheric deposition [13,27]. Approximately 60% of Beijing’ s area lies within the North China Plain, a region of globally typical high nitrogen deposition with total deposition up to 100 kg N hm ^{2} [28-30]. Simulations show that areas outside the plain, influenced by mountainous terrain and covered by forests and grasslands, have significantly lower nitrogen deposition than the eastern and southern parts of the city [31]. The spatial distribution of foliar nitrogen content in *Populus* and *Salix* aligns well with this pattern of atmospheric nitrogen deposition, indicating that plant leaves are sensitive to external nitrogen from atmospheric deposition and can indicate atmospheric nitrogen compound concentrations.

2 Plant Foliar 15N Values as Indicators of Atmospheric Nitrogen Deposition Sources

Lower 15N values in *Populus* and *Salix* leaves occurred in the northeastern and southeastern regions, which are also areas of high nitrogen deposition. These 15N signatures indicate major sources of atmospheric nitrogen compounds. When 15N-NH is negative, it is primarily associated with ammonia volatilization from fertilizer application [34-35]. In the North China Plain' s major agricultural areas, 15N-NH and 15N-NO in wet nitrogen deposition average (-1.2 ± 4.5)‰ and (-2.5 ± 3.7)‰, respectively, particularly during the fertilization season [28]. Although 15N-NO is also a nitrogen source, negative 15N-NO values are mainly related to emissions from high-speed vehicle traffic [24].

The northeastern and southeastern regions are Beijing' s primary agricultural areas. Despite higher elevation in the northeast, the Pinggu area' s fruit and forestry industry makes it an important agricultural zone. The negative 15N values in these regions are therefore closely related to agricultural activities. The eastern and southern areas, with flat terrain, serve as major traffic corridors into Beijing. Traffic pollution emissions contribute to the negative 15N values observed. Walter et al. [24,36] showed that three-way catalytic converters in vehicle exhaust systems significantly reduce and fractionate 15N-NO . Their research indicated that vehicles with catalytic converters emit NO with significantly higher 15N values than those without. Due to logistics supply and restrictions on freight trucks entering the 5th Ring Road, suburban areas become primary routes for vehicles lacking catalytic converters. The negative 15N values in eastern and southern Beijing are thus closely related to emissions from these vehicles. Additionally, the region around Capital Airport shows negative 15N values from both ground transportation connections and aircraft emissions, with aircraft NO 15N ranging from -7.7‰ to 0.6‰ [36].

Higher 15N values in *Populus* and *Salix* leaves appeared in urban areas, the north, and southwest. These elevated values may relate to livestock farming, low-speed vehicle traffic, and coal combustion emissions [21-23]. While some studies show positive 15N values from livestock farms [21], most indicate that 15N-NH from animal waste is negative [20,37], though values become more positive with longer manure storage time as lighter isotopes volatilize first [38]. Beijing' s urban area currently has no crop cultivation or large coal-burning industries. According to Beijing' s land use characteristics, the higher 15N values in urban areas mainly reflect contributions from incomplete fuel combustion emissions under traffic congestion conditions [25]. Notably, while traditional views hold that vehicle emissions are primarily nitrogen oxides, studies show ammonia (NH) is also emitted during low-speed driving with incomplete combustion [39-40]. Whether nitrogen oxides or ammonia, traffic emissions are major pollution sources in Beijing.

In northwestern and southwestern Beijing, lower atmospheric nitrogen deposition coincides with higher 15N values, indicating both lower atmospheric nitrogen compound levels and minimal influence from anthropogenic emission sources. In these forest-covered areas with less human disturbance, leaf litter decomposition through soil nitrogen cycling may contribute to plant nitrogen sources. This factor was not considered for urban and plain areas, where municipal leaf removal and recycling prevent litter accumulation. Although soil samples were not collected due to management restrictions, under high atmospheric nitrogen deposition, plant uptake and utilization of deposited nitrogen is the primary nitrogen source when no other external sources exist.

4 Conclusions

1. Foliar nitrogen contents of typical deciduous broadleaf species *Populus* and *Salix* in Beijing averaged (24.0 ± 4.0) g/kg and (25.9 ± 4.1) g/kg, respectively. These values are similar to those reported in other regional studies but significantly higher than national averages for deciduous broadleaf trees, consistent with zonal characteristics of plant elemental composition.
2. Within the study area, foliar nitrogen content in both genera showed a diagonal distribution pattern of low values in the northwest and high values in the southeast, consistent with spatial variation in atmospheric nitrogen deposition. With no significant climate variation across the region, changes in foliar nitrogen content reflect the influence of atmospheric nitrogen deposition on plant elemental stoichiometry and demonstrate plant responses to atmospheric nitrogen input.
3. Foliar 15N values in both *Populus* and *Salix* showed a spatial pattern of high values in the northwest and urban center, and low values in the southeast. Higher 15N values in central urban areas reflect traffic pollution impacts on atmospheric nitrogen compounds, while higher values in the northwest indicate minimal anthropogenic influence and dominance of natural nitrogen cycling. Lower 15N values in the southeast likely result from combined effects of agricultural activities and traffic emissions.

References

[1] Galloway J N, Dentener F J, Capone D G, Boyer E W, Howarth R W, Seitzinger S P, Asner G P, Cleveland C C, Green P A, Holland E A, Karl D M, Michaels A F, Porter A H, Townsend A R, Vöosmarty C J. Nitrogen cycles: past, present, and future. *Biogeochemistry*, 2004, 70(2): 153-226.

[2] Holland E A, Dentener F J, Braswell B H, Sulzman J M. Contemporary and pre-industrial global reactive nitrogen budgets. *Biogeochemistry*, 1999, 46(1/3): 7-43.

[3] Schlesinger W H. On the fate of anthropogenic nitrogen. *Proceedings of the National Academy of Sciences*, 2009, 106(1): 203-208.

[4] Galloway J N, Townsend A R, Erisman J W, Bekunda M, Cai Z C, Freney J R, Martinelli L A, Seitzinger S P, Sutton M A. Transformation of the nitrogen cycle: recent trends, questions, and potential solutions. *Science*, 2008, 320(16): 889-892.

[5] Dentener F, Stevenson D, Ellingsen K, van Noije T, Schultz M, Amann M, Atherton C, Bell N, Bergmann D, Bey I, Bouwman L, Butler T, Cofala J, Collins B, Drevet J, Doherty R, Eickhout B, Eskes H, Fiore A, Gauss M, Hauglustaine D, Horowitz L, Isaksen I S A, Josse B, Lawrence M, Krol M, Lamarque J F, Montanaro V, Müller J F, Peuch V H, Pitar G, Pyle J, Rast S, Rodriguez J, Sanderson M, Savage N H, Shindell D, Starhan S, Szopa S, Sudo K, van Dingenen R, Wild W, Zeng G. The global atmospheric environment for the next generation. *Environmental Science & Technology*, 2006, 40(11): 3586-3594.

[6] Magill A H, Aber J D, Bernston G M, McDowell W H, Nadelhoffer K J, Melillo J M, Steudler P. Long-term nitrogen additions and nitrogen saturation in two temperate forests. *Ecosystems*, 2000, 3(3): 238-253.

[7] Matson P, Lohse K A, Hall S J. The globalization of nitrogen deposition: consequences for terrestrial ecosystems. *Ambio*, 2002, 31(2): 113-119.

[8] Liu X J, Duan L, Mo J M, Du E Z, Shen J L, Lu X K, Zhang Y, Zhou X B, He C, Zhang F S. Nitrogen deposition and its ecological impact in China: an overview. *Environmental Pollution*, 2011, 159(10): 2251-2264.

[9] Lindberg S E, Lovett G M, Richter D D, Johnson D W. Atmospheric deposition and canopy interactions of major ions in a forest. *Science*, 1986, 231(4734): 141-145.

[10] Rondon A, Granat L. Studies on the dry deposition of NO to coniferous species at low NO concentrations. *Tellus*, 1994, 46(B): 339-352.

[11] Lovett G M, Lindberg S E. Atmospheric deposition and canopy interactions of nitrogen in forests. *Canadian Journal of Forest Research*, 1993, 23(8): 1603-1616.

[12] Vallano D M, Sparks J P. Quantifying foliar uptake of gaseous nitrogen dioxide using enriched foliar 15N values. *New Phytologist*, 2008, 177(4): 946-955.

[13] Hicks W K, Leith I D, Woodin S J, Fowler D. Can the foliar nitrogen concentration of upland vegetation be used for predicting atmospheric nitrogen deposition? Evidence from field surveys. *Environmental Pollution*, 2000, 107(3): 367-376.

[14] Caporn S J M, Carroll J A, Dise N B, Payne R J. Impacts and indicators of nitrogen deposition in moorlands: results from a national pollution gradient

study. *Ecological Indicators*, 2014, 45(5): 227-234.

[15] Liu X J, Zhang Y, Han W X, Tang A H, Shen J L, Cui Z L, Vitousek P, Willem E J, Goulding K, Christie P, Fangmeier A, Zhang F S. Enhanced nitrogen deposition over China. *Nature*, 2013, 494: 459-463.

[16] Han W X, Fang J Y, Gou D L, Zhang Y. Leaf nitrogen and phosphorus stoichiometry across 753 terrestrial plant species in China. *New Phytologist*, 2005, 168(2): 377-385.

[17] Han W X, Fang J Y, Reich P B, Woodward I, Wang Z H. Biogeography and variability of eleven mineral elements in plant leaves across gradients of climate, soil and plant functional type in China. *Ecology Letters*, 2011, 14(8): 788-796.

[18] Application of natural abundance method in ecosystem nitrogen cycle research. *Acta Ecologica Sinica*, 1999, 19(3): 408-416.

[19] Craine J M, Elmore A J, Aidar M P M, Bustamante M, Dawson T E, Hobbie E A, Kahmen A, Mack M C, McLauchlan K K, Michelsen A, Nardoto G B, Pardo L H, Penuelas J, Reich P B, Schuur E A G, Stock W D, Temple P H, Virginia R A, Welker J M, Wright I J. Global patterns of foliar nitrogen isotopes and their relationships with climate, mycorrhizal fungi, foliar nutrient concentrations, and nitrogen availability. *New Phytologist*, 2009, 183: 980-992.

[20] Felix J D, Elliott E M, Gish T J, McConnell L L, Shaw S L. Characterizing the isotopic composition of atmospheric ammonia emission sources using passive samplers and a combined oxidation-bacterial denitrifier approach. *Rapid Communications in Mass Spectrometry*, 2013, 27: 2239-2246.

[21] Yeatman S G, Spokes L J, Dennis P F, Jickells T D. Comparisons of aerosol nitrogen isotopic composition at two polluted coastal sites. *Atmospheric Environment*, 2001, 35(7): 1307-1320.

[22] Heaton T H E. *Tellus*, 1990, 42(3): 304-307.

[23] Felix J D, Elliott E M, Shaw S L. Nitrogen isotopic composition of coal-fired power plant NO : Influence of emission controls and implications for global emission inventories. *Environmental Science & Technology*, 2012, 46: 3528-3535.

[24] Walters W W, Goodwin S R, Michalski G. Nitrogen stable isotope composition (15N) of vehicle-emitted NO . *Environmental Science & Technology*, 2015, 49: 2278-2285.

[25] Ammann M, Siegwolf R, Pichlmayer F, Soter M, Saurer M, Brunold C. Estimating the uptake of traffic-derived NO from 15N abundance in Norway spruce needles. *Oecologia*, 1999, 118(2): 124-131.

[26] Elliott E M, Kendall C, Wankel S D, Burns D A, Boyer E W, Harlin K, Bain D J, Butler T J. Nitrogen isotopes as indicators of NO source contributions to atmospheric nitrate deposition across the midwestern and northeastern United States. *Environmental Science & Technology*, 2007, 41(22): 7661-7667.

[27] McNeil B E, Read J M, Driscoll C T. Foliar nitrogen responses to elevated atmospheric nitrogen deposition in nine temperate forest canopy species. *Environmental Science & Technology*, 2007, 41(15): 5191-5197.

[28] Zhang Y, Liu X J, Fangmeier A, Goulding K T W, Zhang F S. Nitrogen inputs and isotopes in precipitation in the North China Plain. *Atmospheric Environment*, 2008, 42: 1436-1448.

[29] Zhang Y, Song L, Liu X J, Li W Q, Lü S J, Zheng L X, Bai Z C, Cai G Y, Zhang F S. Atmospheric organic nitrogen deposition in China. *Atmospheric Environment*, 2012, 46: 195-204.

[30] Shen J L, Tang A H, Liu X J, Fangmeier A, Goulding K T W, Zhang F S. High concentrations and dry deposition of reactive nitrogen species at two sites in the North China Plain. *Environmental Pollution*, 2009, 157(11): 3106-3113.

[31] Zhang Y, Dore A J, Liu X J, Zhang F S. Simulation of nitrogen deposition in the North China Plain by the FRAME model. *Biogeosciences*, 2011, 8(11): 3319-3329.

[32] Carbon, nitrogen, and phosphorus stoichiometry of plant leaves in Beijing and surrounding areas. *Acta Scientiarum Naturalium Universitatis Pekinensis*, 2008, 45(5): 855-860.

[33] Stoichiometric characteristics of nitrogen and phosphorus in leaves of 753 plant species along the north-south transect of eastern China. *Acta Botanica Sinica*, 2007, 28(12): 2665-2673.

[34] Gao Y. Atmospheric nitrogen deposition to Barnegat Bay. *Atmospheric Environment*, 2002, 36: 5783-5794.

[35] Russell K M, Galloway J N, Macko S A, Moody J L, Scudlark J R. Sources of nitrogen in wet deposition to the Chesapeake Bay region. *Atmospheric Environment*, 1998, 32: 2453-2465.

[36] Walters W W, Tharp B D, Fang H, Kozak B J, Michalski G. Nitrogen isotope composition of thermally produced NO from various fossil-fuel combustion sources. *Environmental Science & Technology*, 2015, 49: 11363-11371.

[37] Felix J D, Elliott E M. Isotopic composition of passively collected nitrogen dioxide emissions: Vehicle, soil and livestock source signatures. *Atmospheric Environment*, 2014, 92: 359-366.

[38] Högberg P. Tansley review No. 95: 15N natural abundance in soil-plant systems. *New Phytologist*, 1997, 137: 179-203.

[39] Liu T Y, Wang X M, Wang B G, Ding X, Deng W, Lü S J, Zhang Y. Emission factor of ammonia (NH) from on-road vehicles in China: Tunnel tests in urban Guangzhou. *Environmental Research Letters*, 2014, 9(6): 1-8.

[40] Suarea-Bertoa R, Zardini A A, Astorga C. Ammonia exhaust emissions from spark ignition vehicles over the new European driving cycle. *Atmospheric*

Environment, 2014, 97: 43-53.