Wet and dry nitrogen deposition in the central Sichuan Basin of China

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HIGHLIGHTS

- Wet and dry N deposition was quantified in central Sichuan Basin for 5 years.
- N forms in N deposition showed significant seasonal and inter-annual variations.
- All reduced N forms contributed 50.9% of total N deposition.
- N deposition (averaged 30.8 kg N ha\(^{-1}\) yr\(^{-1}\)) is a significant N nutrient input.
- Long term and standardized research is urgent for evaluate N deposition.

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Abstract

Reactive nitrogen (Nr) plays a key role in the atmospheric environment and its deposition has induced large negative impacts on ecosystem health and services. Five-year continuous in-situ monitoring of N deposition, including wet (total nitrogen (\(\mu\)TN), total dissolved nitrogen (\(\mu\)TDN), dissolved organic nitrogen (\(\mu\)DON), ammonium nitrogen (\(\mu\)AN) and nitrate nitrogen (\(\mu\)NN)) and dry (\(\mu\)NH\(_3\), \(\mu\)HNO\(_3\), \(\mu\)pNH\(_4\)\(^+\), \(\mu\)pNO\(_3\), \(\mu\)pNO\(_2\)) deposition, had been conducted since August 2008 to December 2013 (wet) and May 2011 to December 2013 (dry) in Yan-ting, China, a typical agricultural area in the central Sichuan Basin. Mean annual total N deposition from 2011 to 2013 was 30.8 kg N ha\(^{-1}\) yr\(^{-1}\), and speculated that of 2009 and 2010 was averaged 28.2 kg N ha\(^{-1}\) yr\(^{-1}\), respectively. Wet and dry N deposition accounted for 76.3% and 23.7% of annual N deposition, respectively. Reduced N (\(\mu\)AN, \(\mu\)NH\(_3\) and \(\mu\)pNH\(_4\)) was 1.7 times of oxidized N (\(\mu\)NN, \(\mu\)pNO\(_3\), \(\mu\)pNO\(_2\) and \(\mu\)pNO\(_2\)) which accounted for 50.9% and 30.3% of TN, respectively. Maximum loadings of all N forms of wet deposition, gaseous NH\(_3\), HNO\(_3\) and particulate NH\(_4\)\(^+\) in dry deposition occurred in summer and minimum loadings in winter. Whether monthly, seasonal or annual averaged, dissolved N accounted for more than 70% of the total. N deposition in the central Sichuan Basin increased during the sampling period, especially that of ammonium compounds, and has become a

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serious threat to local aquatic ecosystems, the surrounding forest and other natural or semi-natural ecosystems in the upper reaches of the Yangtze River.

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1. Introduction

Atmospheric nitrogen (N) deposition is an important component of N biogeochemical cycling (Galloway et al., 2004), with recent studies providing evidence for a positive relationship between increased bulk N deposition and anthropogenic reactive N emissions (NH₃ and NOₓ) (Liu and Zhang, 2009; Liu et al., 2013). The main sources of N deposition are emissions from increasing N fertilizer consumption in agriculture, industry, the export trade (Zhang et al., 2013) and fossil fuel combustion (Compton et al., 2011). Nitrogen fertilizer produced by the Haber-Bosch process feeds almost 50% of the ever increasing world population (Erisman et al., 2008), but NH₃ and NOₓ emissions from the overuse of N fertilizer and other anthropogenic sources have increased atmospheric N deposition such that it now plays a key role in air pollution around the world. They have also caused large negative impacts on ecosystem health and services such as biodiversity loss (Stevens et al., 2004; Clark and Tilman, 2008), eutrophication (Bergström and Jansson, 2006), changes in N transformation process (Hart et al., 1994; Pilkington et al., 2005) and global acidification (Gao et al., 2013; Bates and Peters, 2007).

China produced 37.1 Tg N fertilizer and agriculture consumed 28.1 Tg N fertilizer in 2010, accounting for >30% of total world production and consumption, and exceeding the combined N fertilizer use in North America (11.1 Tg N) and the European Union (10.9 Tg N) in 2009 (Zhang et al., 2013), but less than half of the N applied was taken up by crops (Zhang et al., 2008a). The Sichuan Basin (2.6 × 10⁵ km²) is one of the most intensive agricultural regions in Southwest China, comprising 7% of China’s cropland, producing 10% the country’s total agricultural feed and food, and accounting for 60% of Sichuan’s total grain output (Zhu et al., 2009). Fertilizer N was applied as ammonium bicarbonate and urea at average rates of 108, 218, 283 and 323 kg N ha⁻¹ yr⁻¹ in 1980, 1990, 2000 and 2010 (Sichuan Statistical Yearbook, 1981–2011), respectively, reflecting a continuously increasing trend. Ammonium bicarbonate is highly susceptible to NH₃ loss (Cai et al., 1986), and even though it is being replaced by urea, agriculture is the dominant source of NH₃ emissions when compared with industry, vehicle emissions and human waste (Bouwman et al., 1997). The relatively dispersed agricultural production and decentralized agricultural management, lack of scientific fertilizer application guide and partly the result of topography, causes farmers to apply N fertilizer in accordance with their own subjective ideas, leading to over-fertilization. The continual increases in agricultural production and fertilizer use, industrial development, and population growth are likely to cause increasing reactive N emissions from this area, where the so called ‘Purple Soil’ (a local Chinese name, and for entisols (USA classification)) (Zhu et al., 2009) with its high pH (6.0–8.5) occurs widely. Nitrogen pollution is a serious issue in the Sichuan Basin, even in the upper reaches of the Yangtze River, with its widely distributed natural and semi-natural ecosystems. Liu and Zhang (2009) have shown that bulk N deposition (i.e. wet plus some dry deposition, measured in open collectors) in the Sichuan Basin is >25 kg N ha⁻¹ yr⁻¹. High N deposition will not only provide N useful for crop growth in agro-ecosystems but also that will negatively affect natural and semi-natural ecosystems (Liu et al., 2011).

Previous research in the Sichuan Basin measured only ammonium and nitrate N in bulk deposition (e.g. Liu and Zhang, 2009; Yang and Sun, 2008), and Xu et al. (2015) only evaluated total inorganic N deposition nationally. No systematic research has been conducted to provide for detailed information on atmospheric deposition that includes both wet and dry deposition of inorganic and organic N species in the Sichuan Basin. Therefore we continuously monitored in-situ N deposition in Yan-ting County of the Sichuan Basin, with the objectives being 1) to provide continuous monitoring data for N deposition for national evaluation; 2) to quantify forms and fluxes, seasonal and inter-annual variations of N deposition in order to better understand the current status of N deposition and impacts in this area.

2. Materials and methods

2.1. Site descriptions

The experimental site (N31°16’, E105°28’ at an altitude of 400–600 m) was located at the ‘Yan-ting Agro-Ecological Station of Purple Soil’, a member station of the Chinese Ecosystem Research Network (CERN), Chinese Academy of Sciences, in the central Sichuan Basin, southwestern China (Fig. 1). It has a moderate subtropical monsoon climate with annual mean precipitation of 836 mm, distributed 5.9%, 65.5%, 19.7% and 8.9% in the spring, summer, autumn and winter, respectively. The average air temperature is 17.3 °C. The calcareous ‘Purple Soil’, which has a pH > 8, is widely distributed in this region. The site is located in a typical intensive agricultural area with mostly rice-wheat, rice-oilseed rape and maize-wheat cropping systems and an average synthetic N fertilizer application rate of >280 kg N ha⁻¹ yr⁻¹.

2.2. Measurements of wet and dry deposition

2.2.1. Sampling and analysis for wet deposition

Wet deposition was collected and quantified at Yan-ting from August 2008 to December 2013. Precipitation after each event was collected using wet-dry automatic collectors (UNS 130/E & DSC HD/PE, produced by Eigenbrodt, Germany). The cover of the collection funnel opened automatically without delay when the precipitation sensor was activated and closed automatically when precipitation ceased and no water remained on the sensor surface. A thermostatic device in the sensor controlled evaporation in order to ensure the cover closed quickly once precipitation ceased. Precipitation was collected in a 5000 ml sample bottle during the event and taken to the laboratory within 2 h of each event. After each sampling, the container was cleaned with deionized water. All samples were refrigerated at 4 °C and analyzed within 48 h.

Each sample was filtered through a 0.45 μm membrane filter (Tianjin, China), and 50 ml of filtrate stored in plastic bottles. The species and concentrations of inorganic N (IN) including NH₄⁺-N (AN) and NO₃⁻-N (NN) were measured using an Automatic Analyzer (AA3 continuous-flow analyzer, Seal Corporation, Germany). The detection limit of N for this instrument was 3 μg N L⁻¹. It should be noted that NO₃⁻-N was converted to NO₂⁻-N during the analysis, so NO₂⁻-N was included in our analysis, and the ‘NO₃⁻-N’ recorded equal to the sum of NO₂⁻-N and NO₃⁻-N. Total N (TN) and total
dissolved N (TDN) were measured via oxidation by alkaline potassium persulfate and measured using UV spectrophotometry (UV-1810 PC, Persee Corporation, China). Dissolved organic N (DON) was calculated as DON = TDN-DIN.

2.2.2. Sampling and analysis for dry deposition

Dry deposition was collected and quantified from May 2011 to December 2013. Gaseous NH₃, HNO₃ and particulate NH₄⁺ and NO₃⁻ (pNH₄⁺ and pNO₃⁻) were collected using a DELTA system, designed by the Center for Ecology and Hydrology, Edinburgh, UK. The particle cut-off size for the DELTA system was estimated to be 4–5 μm (Tang et al., 2009). The DELTA system comprised a denuder filter sampling train, which contained two long glass denuders (each 15 cm long) and two short glass denuders (each 10 cm long) to collect gaseous HNO₃ and NH₃, respectively, and two aerosol filters of diameter 37 mm to capture pNH₄⁺ and pNO₃⁻. A mini air pump (6 V) provided low sampling flow rates of 0.2–0.4 L min⁻¹, with a high sensitivity dry gas meter to record the sampled volume. When a laminar air stream passes through the denuder, coated on the inside with acid or alkaline solution, HNO₃ or NH₃, respectively, were captured, while aerosols pass through to be captured by the filters. The mixed air inlet is set at 1.6 m above the ground (Tang et al., 2009; Luo et al., 2013).

The denuder filter sampling train was collected on the last day of each month. It was immediately refrigerated at 4 °C and analyzed within one week. 10 ml 0.05% H₂O₂ solution were used to extract the denuders for HNO₃ and the filters for pNO₃⁻, and 10 ml high purity water were used to extract the denuders for NH₃ and the filters for pNH₄⁺. Ammonium and nitrate in the solutions were measured with an Automatic Analyzer, as described above.

Atmospheric NO₂ was collected using Gradko passive diffusion tubes as in the UK Environmental Change Network (http://www.ecn.ac.uk). The samplers comprised polyacrylic tubes, two caps, and stainless steel mesh disks. Two dry disks were placed in a gray cap and 30 μl 20% triethanolamine pipetted into the caps, which were then placed on the tubes; the other end of each tube was capped with a white cap; three tubes were prepared for each month. The samplers were fixed 1.5 m above the ground, the white caps removed, and the tubes exposed for one month. At the end of each month, the white caps were replaced for transport and the disks extracted with a solution containing sulphanilamide, H₃PO₄ and N-1-Naphthylethylene-diaminedihydrochloride. NOₓ was analyzed using visible spectrophotometry with a colorimetric method at a wavelength of 542 nm (Luo et al., 2013).
2.3. Calculation and statistical analysis

Monthly wet N deposition was obtained by multiplying the concentration of N in an event by the amount of precipitation in that event as measured in a standard rain gauge, using the equation:

\[ C = \left( \sum_{i=1}^{n} C_i P_i \right) / \left( \sum_{i=1}^{n} P_i \right) \]

\( P_i \) is the amount (mm) of precipitation in event i, \( C_i \) is the measured concentration (mg N L\(^{-1}\)) of N in that event, and \( n \) is the number of precipitation events in a month.

The monthly, seasonal and annual wet depositions were calculated according to the equation:

\[ D_w = 10^{-2} \sum_{i=1}^{n} C_i P_i \]

\( P_i \) is the amount (mm) of precipitation in event i, \( C_i \) is the measured concentration (mg N L\(^{-1}\)) of N forms in that event, \( n \) is the number of precipitation events in the monthly, seasonal or annual period, and \( 10^{-2} \) is a unit conversion factor.

The monthly fluxes (F) of gaseous and particulate dry deposition N forms are the product of the concentration of N species at a specific height (\( C(z) \)) and the deposition velocity (\( V_d \)):

\[ F = -C_2 V_d \]

As it is difficult to measure the real \( V_d \), well-tested \( V_d \) models with empirical parameters cited from the literature, combined with the on-site meteorological data, were used to estimate \( V_d \). For Nr gases, the dry deposition velocity is calculated as the reciprocal of the sum of three different deposition resistances according to the big-leaf resistance analogy model (Wesely and Hicks, 1977) as

\[ V_d = 1/(R_a + R_b + R_c) \]  

\( R_a \) is the aerodynamic resistance, \( R_b \) is the quasi laminar boundary layer resistance, and \( R_c \) is the canopy resistance. \( R_a \) is parameterized according to Erisman and Draaijers (1995) as

\[ R_a(z) = (k u_r)^{-1} \left[ \ln \left( \frac{z - d}{z_0} \right) + \psi_h \left( \frac{z - d}{z_0} \right) + \psi_h \left( \frac{z_0}{L} \right) \right] \]

\( z \) is the measuring sensor height, \( k \) is the von Karman constant (0.41), \( u_r \) is the friction velocity, \( d \) is the zero-plane displacement height, \( z_0 \) is the roughness length, \( \psi_h \) is the integrated stability function for entrained scalars and \( L \) is the Monin-Obukhov length. \( d \) and \( z_0 \) were set to be 0 m and 0.01 m respectively for bare soil and 0.67 and 0.1 times of the plant height, respectively, when the soils were fully covered with plants (Li et al., 2000; Zhang et al., 2003b).

\( R_b \) is also parameterized according to Erisman and Draaijers (1995) as

\[ R_b = (2/(k u_r)^{2/3}) (S_c/P_i)^{2/3} \]

\( k \) is the von Karman constant, \( u_r \) is the friction velocity, \( P_i \) is the Prandtl number, and \( S_c \) is the Schmidt number. \( R_c \) is calculated according to Wesely (1989) as

\[ R_c = \left[ \frac{1}{R_s + R_m} + \frac{1}{R_l} + \frac{1}{R_d} + \frac{1}{R_c} + \frac{1}{R_a} + \frac{1}{R_g} \right]^{-1} \]

\( R_s \) is stomatal resistance, \( R_m \) is the leaf mesophyll resistance, \( R_l \) is the leaf cuticles and otherwise the outer surfaces in the upper canopy resistance, \( R_d \) is the gas-phase transfer resistance affected by buoyant convection in canopies, \( R_c \) is the leaves, twig, bark or other exposed surfaces resistance in the low canopy, \( R_a \) is the transfer resistance that depends only on canopy height and density, and \( R_g \) is the soil, leaf litter, etc., at the ground surface.

For particulate N species, the dry deposition velocity is parameterized according to Slinn (1982) using the following equation:

\[ V_d = \frac{1}{R_a + R_{surf}} + V_g \]  

\( R_a \) is the aerodynamic resistance, \( R_{surf} \) is the surface resistance, and \( V_g \) is the gravitational settling (or sedimentation) velocity. Details of the parameterization of \( R_{surf} \) and \( V_g \) can be found in Zhang et al. (2001).

Seasonal N deposition was calculated as the sum of wet and dry N deposition per month, summed for the appropriate three-month period. The seasons were divided into spring (March to May), summer (June to August), autumn (September to November) and winter (December to February). All statistical analyses were made using SPSS 13.0 (SPSS Inc., USA) and figures drawn using SPSS 13.0 and Office Standard 2010 (Microsoft, USA). A one-way analysis of variance (ANOVA) was used to describe seasonal variation of N forms (\( \alpha = 0.05 \)).

3. Results

3.1. Precipitation

Annual precipitation from 2008 to 2013 averaged 1026.4 mm, and increasing gradually over the experimental period, it was 23% higher than the long-term average. The most intense rain events occurred in the rainy season from May to September. Precipitation in the rainy season accounted for 76.4%, 81.0%, 85.6%, 83.9%, 84.4% and 89.3% of total annual precipitation from 2008 to 2013, respectively.

3.2. Wet deposition

During the monitoring period from August 2008 to December 2013, concentrations of total N (CWTN), total dissolved N (CWTN) and NO\(_3\) -N (CWNN) in all wet deposition averaged 2.90, 2.55, 1.48 and 1.05 mg N L\(^{-1}\), respectively (Fig. 2A); CWAN was 1.4 times that of CWNN. Monthly amounts of total N (WTN) and total dissolved N (WTDN) in wet deposition averaged 1.85 and 1.57 kg N ha\(^{-1}\), respectively (Fig. 2C). On average, monthly WTDN accounted for 86.4% of WTN, and was the dominant component of monthly loadings. Monthly amounts of ammonium (\( \omegaN \)) and nitrate (\( \omegaNN \)) in wet deposition averaged 0.87 and 0.52 kg N ha\(^{-1}\), respectively (Fig. 2C). Monthly amounts of total dissolved organic N in wet deposition (\( \omegaDON \)) averaged 0.18 kg N ha\(^{-1}\) (Fig. 2C), accounting for 11.5% of WTN. Seasonal average WTN, WTDN, WAN, \( \omegaNN \) and \( \omegaDON \) in spring were 5.42, 4.42, 2.48, 1.54 and 0.40 kg N ha\(^{-1}\), respectively (Fig. 3A), in summer were 10.48, 8.98, 5.37, 2.46 and 1.15 kg N ha\(^{-1}\); in autumn were 5.19, 4.34, 2.09, 1.66 and 0.59 kg N ha\(^{-1}\), and in winter were 1.37, 1.22, 0.50, 0.59 and 0.13 kg N ha\(^{-1}\). Seasonal WTDN accounted for 85.0% of WTN on average; and WAN was about 1.5 times that of \( \omegaNN \). All components of wet N deposition had significant seasonal differences. N loadings in summer were significantly higher than that in other seasons, and the least loadings were usually in winter.
3.3. Dry deposition

3.3.1. Gaseous dry deposition (NH₃, HNO₃, NO₂)

During the monitoring period from May 2011 to December 2013, concentrations of gaseous NH₃ (CDNH₃), HNO₃ (CDHNO₃) and NO₂ (CDNO₂) averaged 3.71, 0.56 and 2.62 μg N m⁻³, respectively (Fig. 2B). CDNH₃ was 6.6 times that of CDHNO₃. Monthly dry depositions of gaseous ammonia (DNH₃), gaseous nitric acid (DHNO₃) and nitrogen dioxide (DNO₂) averaged 0.31, 0.11 and 0.07 kg N ha⁻¹, respectively (Fig. 2D). Seasonal average DNH₃, DHNO₃, DNO₂ in spring were 0.67, 0.32 and 0.27 kg N ha⁻¹, in summer were 0.45, 0.28 and 0.10 kg N ha⁻¹, and in winter were 0.19, 0.17 and 0.19 kg N ha⁻¹. DNH₃ in summer was significantly higher than those in other seasons; DHNO₃ in winter and DNO₂ in autumn were significant lower than those in other seasons.

3.3.2. Particulate dry deposition (pNH₄⁺, pNO₃⁻)

Concentrations of pNH₄⁺ (CDpNH₄⁺) and pNO₃⁻ (CDpNO₃⁻) averaged 3.15 and 1.15 μg N m⁻³ respectively (Fig. 2B). The mean CDpNH₄⁺ was 2.7 times that of CDpNO₃⁻. Monthly amounts of particulate ammonium (DPNH₄⁺) and particulate nitrate (DPNO₃⁻) deposited averaged 0.15 and 0.05 kg N ha⁻¹, respectively (Fig. 2D). Seasonal average DpNH₄⁺ and DpNO₃⁻ were 0.33 and 0.14, 0.52 and 0.11, 0.40 and 0.16, and 0.37 and 0.18 kg N ha⁻¹ in spring, summer, autumn and winter, respectively (Fig. 3B). DpNH₄⁺ and DpNO₃⁻ did not vary significantly through the seasons.

3.3.3. Annual N deposition

Annual N deposition in 2009 and 2010 comprised only wet deposition, which in 2011, 2012 and 2013 included wet and dry deposition. From 2011 to 2013, annual N deposition was 29.6, 33.9 and 28.8 kg N ha⁻¹ yr⁻¹, respectively (Fig. 4). Wet N deposition was 22.5 kg N ha⁻¹ yr⁻¹ on average, respectively; dry N deposition was 6.0, 7.8 and 8.0 kg N ha⁻¹ yr⁻¹ from 2011 to 2013, averaged 7.3 kg N ha⁻¹ yr⁻¹. According to the average amount of dry N deposition from 2011 to 2013, it was speculated that annual total N deposition in 2009 and 2010 were 30.5 and 25.9 kg N ha⁻¹ yr⁻¹, respectively. Annual average WTDN, WAN, WNN, WDN, pNH₃, pHNO₃, pNH₄⁺, pNO₃⁻ and pNO₂ were 19.0, 10.4, 6.3, 2.3, 3.3, 1.1, 1.6, 0.6 and 0.8 kg N ha⁻¹ yr⁻¹, respectively. The main form of N deposition was inorganic, with ammonia-containing substances (WAN, NH₃ and pNH₄⁺) and nitrate-containing substances (WNN, HNO₃, NO₂ and pNO₃⁻) accounting for 50.9% and 30.3% of TN, respectively. Annual total DON accounted for 11.9% of WTDN or 8.4% of TN, respectively.

4. Discussion

4.1. Reduced N compared with oxidized N in wet and dry deposition

Concentrations of AN and monthly, seasonal and annual amounts of AN in wet deposition were 1.4, 1.7, 1.5 and 1.7 times those of NN, respectively, and the ratios of the concentrations and monthly, seasonal and annual amounts of gaseous NH₃ to HNO₃ in
dry deposition were 6.6, 2.8, 2.5 and 2.9, respectively, analogous ratios of pNH₄⁺ and pNO₃/CO were 2.7, 3.0, 2.9 and 2.8, respectively. Reduced N was more than oxidized N. The ratio of all reduced N (WAN, DNH₃ and DpNH₄⁺) to oxidized N (WNN, DHNO₃, DNO₂ and DpNO₃/C₀) in total N deposition was about 1.7, higher than that at Chengdu, which was measured 1.3 in 2008 (Wang and Han, 2011). It was also higher than those of rural areas of China in recent years, e.g. 1.5 in bulk deposition in rural sites in North China in 2011 (Luo et al., 2013), and 1.5 in wet deposition in the Tai Lake region (Table 2) (Xie et al., 2008). In general, concentrations of AN were usually higher than those of NN (Table 1), no matter what the ecosystem and when it was measured. Recently, some studies have confirmed that urban areas also have local NH₃ sources, contradicting the traditional view that NH₃ is transported to urban areas from agricultural regions (Pan et al., 2016). Concentrations of all components of dry deposition (DNH₃, DHNO₃, DpNH₄⁺, DpNO₃/C₀ and pNO₂)
\( \text{NH}_3 \text{ and particulate NH}_4^+ \) of means of all single measured values per months during monitoring period. Concentrations of monthly \( \text{pNH}_4^+ \) this alkaline soil. The concentration of gaseous \( \text{NH}_3 \) then decreased, volatilization from the ammonium bicarbonate and urea applied to April to July, which are the months with higher temperatures and in this study were much lower than those measured in North China from 2006 to 2008 (Shen et al., 2009) or 2010 to 2012 (Luo et al., 2013).

Concentrations of \( \text{WAN} \) and \( \text{WNN} \) in wet deposition (Fig. 5A) decreased from January to May, and then increased from October to December. Precipitation washes out pollutants from the atmosphere, and concentrations in precipitation depend on pollutant load and the amount of precipitation. We measured minimum values in July, August or September, which are the rainy months. It should also be noted that crops are planted and fertilized in the Yan-ting area from late May to June, and again in October to November, when concentrations of all ammonium and nitrate forms in wet N deposition rapidly increased.

Gaseous \( \text{NH}_3 \) in dry deposition (Fig. 5B) increased rapidly from April to July, which are the months with higher temperatures and in which fertilizers are applied, leading to considerable ammonia volatilization from the ammonium bicarbonate and urea applied to this alkaline soil. The concentration of gaseous \( \text{NH}_3 \) then decreased, but increased a little in October when fertilizer is again applied but at relatively lower temperatures. Concentrations of monthly \( \text{pNH}_4^+ \) in dry deposition (Fig. 5B) varied slightly through the year, indicating that emission sources or the formative environment for aerosols were stable and that precipitation did not wash out \( \text{pNH}_4^+ \) effectively. Pan et al. (2012) also showed that, in northern China, the proportion of all samplers was only 45% which \( \text{pNH}_4^+ \) changed as the changes of the rainfall. Concentrations of \( \text{HNO}_3 \) and \( \text{pNO}_3^- \) increased from January to March (Fig. 5C), then decreased gradually from April to September. When the rainy season ended, concentrations of these two oxidized pollutants increased again because these oxidized pollutants are washed out effectively. Our results contrast with those of Northern China (Pan et al., 2012), where amounts of \( \text{WNO}_3 \) were not significantly correlated with rainfall. Strong local anthropogenic sources (industry and high density transportation) in Northern China could cause these very different results.

### Table 1: Concentrations of AN and NN in bulk N precipitation at various sites in China.

<table>
<thead>
<tr>
<th>Observation period</th>
<th>Ecosystem-type</th>
<th>City</th>
<th>Location</th>
<th>AN (ng N L(^{-1}))</th>
<th>NN (ng N L(^{-1}))</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>2008.03–2008.10</td>
<td>Urban</td>
<td>Sichuan, Chengdu</td>
<td>30°39.2’N,104°0.7’E</td>
<td>3.03 ± 2.60</td>
<td>2.36 ± 1.99</td>
<td>(Wang and Han, 2011)</td>
</tr>
<tr>
<td>2006.01–2006.12</td>
<td>Urban</td>
<td>Beijing</td>
<td>39°56’N,116°23’E</td>
<td>3.10 ± 2.08</td>
<td>1.54 ± 1.11</td>
<td>(Xu and Han, 2009)</td>
</tr>
<tr>
<td>2006.01–2006.12</td>
<td>Urban</td>
<td>Guangdong, Guangzhou</td>
<td>–</td>
<td>0.99 ± 0.16</td>
<td>0.75 ± 0.12</td>
<td>(Cao et al., 2009)</td>
</tr>
<tr>
<td>1998–2000</td>
<td>Urban</td>
<td>Xiang, Lhasa</td>
<td>–</td>
<td>0.20 ± 0.27</td>
<td>0.10 ± 0.15</td>
<td>(Zhang et al., 2003)</td>
</tr>
<tr>
<td>2005.01–2005.12</td>
<td>Urban</td>
<td>Shanghai</td>
<td>31°18’N,121°30’E</td>
<td>1.91 ± 0.20</td>
<td>1.09 ± 0.15</td>
<td>(Huang et al., 2008)</td>
</tr>
<tr>
<td>1999.01–2004.09</td>
<td>Suburban</td>
<td>Beijing, Haidian</td>
<td>39°57’N,116°18’E</td>
<td>4.67 ± 1.33</td>
<td>2.24 ± 0.54</td>
<td>(Liu et al., 2006)</td>
</tr>
<tr>
<td>2004–2008</td>
<td>Suburb</td>
<td>Shenyang, Liaoning</td>
<td>41°32’N,123°2’E</td>
<td>1.82 ± 0.51</td>
<td>0.89 ± 0.21</td>
<td>(Yu et al., 2011)</td>
</tr>
<tr>
<td>2005.09–2007.10</td>
<td>Suburb</td>
<td>Zhejiang, Hangzhou</td>
<td>30°18’N,119°04’E</td>
<td>0.81 ± 0.20</td>
<td>0.59 ± 0.10</td>
<td>(Li et al., 2010)</td>
</tr>
<tr>
<td>2006.04–2007.10</td>
<td>Suburb</td>
<td>Heilongjiang, Wuchang</td>
<td>44°44’N,127°36’E</td>
<td>1.39 ± 0.31</td>
<td>0.55 ± 0.06</td>
<td>(Li et al., 2010)</td>
</tr>
<tr>
<td>2003.04–2004.02</td>
<td>Rural</td>
<td>Xinjiang, Urumqi river valley</td>
<td>–</td>
<td>0.32 ± 0.26</td>
<td>0.44 ± 0.82</td>
<td>(Zhao et al., 2008)</td>
</tr>
<tr>
<td>2009.06–2010.07</td>
<td>Rural</td>
<td>Hubei, Zigui</td>
<td>30°38’N,31°11’N,110°18’N -111°0’E</td>
<td>1.44 ± 0.76</td>
<td>1.53 ± 1.44</td>
<td>(Wu and Han, 2015)</td>
</tr>
<tr>
<td>2008.04–2008.06</td>
<td>Rural</td>
<td>Jiangxi,Yingtian</td>
<td>28°12’N,116°55’E</td>
<td>2.58 ± 3.19</td>
<td>0.83 ± 0.65</td>
<td>(Cui et al., 2011)</td>
</tr>
<tr>
<td>2009.01–2012.12</td>
<td>Rural</td>
<td>Sichuan,Xiaying</td>
<td>31°16’N,105°28’E</td>
<td>1.48 ± 1.05</td>
<td>1.05 ± 0.91</td>
<td>(This study)</td>
</tr>
<tr>
<td>2012.01–2012.12</td>
<td>Natural</td>
<td>Yunnan, Lijiang</td>
<td>100°07’–100°10’ E, 27°10’–27°40’N</td>
<td>0.29 ± 0.03</td>
<td>0.01 ± 0.01</td>
<td>(Niu et al., 2014)</td>
</tr>
<tr>
<td>2010.04–2011.05</td>
<td>Natural</td>
<td>Sichuan, Jiu Zhaigou</td>
<td>33°02’N,103°56’E</td>
<td>0.19 ± 0.18</td>
<td></td>
<td>(Qiao et al., 2015)</td>
</tr>
</tbody>
</table>

Fig. 5. Monthly concentration of reduced and oxidized N forms in wet and dry deposition. (A) concentrations of \( \text{NH}_4^+ \)-N and \( \text{NO}_3^- \)-N in wet deposition, (B) concentrations of gaseous \( \text{NH}_3 \) and particulate \( \text{NH}_4^+ \) in dry deposition, and (C) concentrations of gaseous \( \text{HNO}_3 \) and particulate \( \text{NO}_3^- \) in dry deposition. Error bar in each column denotes standard deviation (error) of means of all single measured values per months during monitoring period.

4.2. Temporal variation of N deposition in the central Sichuan Basin

4.2.1. Annual variation of wet N deposition

Precipitation and average concentrations of \( \text{WAN} \) and \( \text{WNN} \) in wet deposition from 2002 to 2012 and the ratio of the concentrations of \( \text{WAN} \) and \( \text{WNN} \) decreased from 2.3 in 2002–2004 (Yang and Sun, 2008) to 1.4 in 2008–2013 (our data) (Fig. 6A). Amounts of \( \text{WAN} \) and \( \text{WNN} \) were 5.8 and 2.7 kg N ha\(^{-1}\) yr\(^{-1}\) at Yan-ting from 2002 to 2004 (Yang and Sun, 2008), which accounted for 44.8% and 20.8% of wet N deposition, respectively, and had increased to 10.4 and 6.3 kg N ha\(^{-1}\) yr\(^{-1}\) from 2009 to 2013, accounting for 47.0% and 27.8% of wet N deposition, respectively.
However, the ratio of ammonium N to nitrate N decreased over that period, despite the amounts of both \( w_{AN} \) and \( w_{NN} \) having increased (Fig. 6B). This shows that emissions of \( w_{AN} \) and \( w_{NN} \) were relatively stable or increasing, those of reduced forms more than oxidized forms. Thus agricultural, transport and industrial sources of N in the Yan-ting area increased, but those of transport and industry increased faster than those of agriculture. This was because, after the earthquake of May 2008, the local construction industry (brickyard, cement works and timber mills) developed rapidly.

N deposition in the Sichuan Basin has increased rapidly and Sichuan has become one of the most polluted areas in China. Mean wet N deposition at our monitoring site from 2002 to 2004 was 13.1 kg N ha\(^{-1}\) yr\(^{-1}\) (Yang and Sun, 2008) and 22.5 kg N ha\(^{-1}\) yr\(^{-1}\) from 2009 to 2013 (our data), i.e. an increase of >70% in the last 10 years. Large quantities of studies have shown that N deposition has been increasing recently (Liu et al., 2013), but over a very long period. Wet N deposition at Rothamsted Experimental Station in 1855 was 5.2 kg N ha\(^{-1}\) yr\(^{-1}\) but increased to 18.0 kg N ha\(^{-1}\) yr\(^{-1}\) in 1980 then decreased gradually (Goulding et al., 1998). Total N deposition in Northern China averaged 60.6 kg N ha\(^{-1}\) yr\(^{-1}\) from 2008 to 2010 (Pan et al., 2012) and 76.4 kg N ha\(^{-1}\) yr\(^{-1}\) from 2010 to 2012 (Luo et al., 2013). Highly significant (\( P < 0.001 \)) increases in bulk N deposition have also been found in southwest China, with an annual average increment rate of 0.4 kg N ha\(^{-1}\) yr\(^{-1}\) from 1980 to 2010 (Liu et al., 2013). From 2011 to 2013, the inter-annual variation coefficient of wet N deposition, \( w_{AN} \) and \( w_{NN} \) was large at 28.1%, 29.3% and 43.7%, respectively. Yang et al. (2015) showed that in East Asia, the inter-annual variations of aerosols were dominated by variations in meteorological conditions, and the decadal trends of this variation were driven by variations of anthropogenic emissions. Such high inter-annual variations in our study show that long-term, continuous and uniform standard research is necessary and important to better understand the changes of N deposition.

Liu et al. (2013, 2016) concluded that NH\(_4\)-N from agricultural sources still dominates wet/bulk N deposition but the contribution has decreased drastically between the 1980s and 2000s. This contrasts with the N deposition pattern in the US, where reduced N was of increasing importance compared with oxidized N (Li et al., 2016). The absolute amount of NH\(_4\)-N deposited is still increasing. In addition to agricultural sources there are emissions from cities. Ammonia is a key to the formation of secondary particles, being easily absorbed by acid materials such as H\(_2\)SO\(_4\) and HNO\(_3\). However, the importance of ammonium N in the Sichuan Basin is not appreciated. Indeed in China, where the annual emission of ammonia was 8.4 Tg from 2005 to 2008 (Paulot et al., 2014), ammonia induced pollution, especially that from agriculture, is not yet taken into account in air pollution legislation.

4.2.2. Seasonal and inter annual variation of N deposition

Concentrations and amounts deposited of all N forms in wet deposition (\( w_{TN}, w_{TDN}, w_{AN}, w_{NN} \) and \( w_{DON} \)) in summer were significantly higher than in other seasons and their minima always occurred in winter (Fig. 3A), because abundant rainfall in summer washes out pollutants. The inter-annual coefficients of variation of wet N deposition (\( w_{TN}, w_{TDN}, w_{AN}, w_{NN} \) and \( w_{DON} \)) were 12.8%, 12.9%, 9.9%, 23.4% and 35.0%, respectively. \( w_{AN} \) varied the least but \( w_{NN} \) and \( w_{DON} \) varied the most indicating that, in Yan-ting, agriculture sources were relatively stable, but industrial and traffic sources variable, perhaps because of their rapid development. Amount of \( p_{NH_3} \) was 5.0 times that of \( p_{HNO_3} \) and 8.4 times that of \( p_{NO_2} \) in summer. \( p_{NH_3} \) in summer was significantly higher than in winter, spring and autumn (Fig. 3B), in agreement with research in Northern China. Thus summer fertilization is a dominant factor influencing the composition of N in the atmosphere in this area. \( p_{NH_3} \) showed no significant seasonal variation, but the highest value also occurred in summer, showing the importance of fertilizer use, especially as livestock emissions and human excretion are relatively stable over the whole year. This is contrary to research in Northern China (Pan et al., 2012), where \( p_{NH_3} \) was higher in winter than in other seasons not only because this is the dry season, but also because of the complex local emissions from industry or transportation. \( p_{HNO_3} \) in summer was higher than in other seasons and significantly different to that in winter. The inter-annual coefficients of variation of \( p_{NH_3}, p_{HNO_3}, p_{NH_4}, p_{NO_3} \) and \( p_{NO_2} \) were 11.6%, 10.9%, 17.2%, 39.9% and 23.6%, respectively. The variations of reduced N in dry deposition were lower than those of oxidized N because annual industrial and traffic sources varied considerably, in agreement of variations in wet N deposition.

4.3. N deposition in Yan-Ting compared with other agricultural regions

Annual N deposition at Yan-Ting, including wet and dry deposition, was 30.8 kg N ha\(^{-1}\) yr\(^{-1}\) on average, higher than in some agricultural areas such as Changsha, Shenyang and the Tai Lake region, but lower than in Fengqiu, Jiangsu, Ying-tan and Northern China (Table 2). Xu et al. (2015) reported annual N deposition in rural areas of the Northeast Plain, the North China Plain including the Weihe Plain and the Sichuan Basin of 40.1, 56.4 and 35.4 kg N ha\(^{-1}\) yr\(^{-1}\), respectively, in these major agricultural regions
of China. Our data are slightly lower than the average values for the Sichuan Basin, indicating that the Yan-ting area experiences moderate N depositions for China. Wet and dry N deposition accounted for 76.3% and 23.7% of annual N deposition, respectively. Dentener et al. (2006) used a multi-model to evaluate and predict dry N deposition on a regional scale in 2000 and 2030. Their results showed that NHX and NOy dry deposition in East Asia were 7.4 and 3.0 kg N ha\(^{-1}\) yr\(^{-1}\) in 2000, respectively, increasing to 8.0 and 3.7 kg N ha\(^{-1}\) yr\(^{-1}\) in 2030. Our results for NHX and NOy in dry deposition from 2011 to 2013 were 4.8 and 2.5 kg N ha\(^{-1}\) yr\(^{-1}\), respectively, lower than Dentener et al. (2006) modelled average values for 2000. NHx deposition in our study was approximately the same as in other major agricultural regions around the world in the same year, which in Southeast Asia, South Asia and the East European Plain were 4.6, 4.8 and 4.9 kg N ha\(^{-1}\) yr\(^{-1}\), respectively, but higher than in South America (2.2 kg N ha\(^{-1}\) yr\(^{-1}\)), the USA (3.0 kg N ha\(^{-1}\) yr\(^{-1}\)) and Western Europe (3.9 kg N ha\(^{-1}\) yr\(^{-1}\)). Thus, although its deposition is not the highest of the agricultural areas, the Sichuan Basin is still a hot spot of NHX emission.

On average, the amounts of AN and NN contributed 62.9% and 37.1% of inorganic N in wet deposition, in contrast to that at Ying- tan Ecological Experimental Station (Jiangxi, South China), at which AN and NN contributed almost exactly the reverse at 38.8% and 61.2%, respectively, from 2004 to 2005 (Fan et al., 2009). Our results showed that DON deposition on 2009 to 2013 was 2.3 kg N ha\(^{-1}\) yr\(^{-1}\) on average, ranging from 1.1 to 3.3 kg N ha\(^{-1}\) yr\(^{-1}\). Previous studies in Sichuan (sampling sites were in Wenzhang, Jianyang and Chengdu) from 2005 to 2009 measured annual DON deposition at 4–5 kg N ha\(^{-1}\) yr\(^{-1}\) (Zhang et al., 2012), higher than in our study. Previous research also showed that DON accounted for 20.8% of N deposition over ten monsoon seasons at Ying-tan from 2003 to 2012 (Cui et al., 2014), so DON is not a negligible component of N deposition.

### 4.4. Some ecological effects of N deposition in the upper reaches of the Yangtze River

Precipitation is the only way by which surface water and ground water are replenished. Rainwater containing high concentrations of N is bound to affect surface and ground water quality. Also critical loads for N deposition of the main lakes and reservoirs in China are small and those of some lakes and reservoirs have been exceeded in recent years. So, even if N deposition is the only source of pollution, some lakes and reservoirs will become eutrophic (Ye et al., 2002). In the Environmental Quality Standards for Surface Water (GB3838-2002) in PR China, surface waters have been divided into five categories based on features and ecological functions. Agricultural water and general landscape water is in the fifth category, which requires concentrations of TN, AN and NN of ≤2, 2 and 20 mg N L\(^{-1}\), respectively. Unfortunately 67.0% of TN and 26.3% of AN in the wet deposition sampled in our study exceeded these requirements. This shows that, although the contribution of NN from precipitation to surface water is small and it will be diluted when entering surface and groundwater, the contribution of AN cannot be ignored. In addition, the secondary protection zones for centralized surface water for drinking, the third category, has required concentrations of TN, AN and NN of ≤1, 1 and 10 mg N L\(^{-1}\), respectively. 91.6% of TN and 61.5% of AN in the wet N deposition sampled in our study exceeded this standard. Research has shown that AN concentrations in precipitation in China attain the required standards of the third category only in nature reserves or sparsely populated areas (Table 1), although some urban and rural areas such as Lhasa and Hangzhou achieved the standards in the years before agriculture was greatly intensified (Table 1). NN in all areas can meet the standard for the third category.

The concentration of gaseous NH3 in the central Sichuan Basin was 3.7 µg N m\(^{-3}\) on average and ranged from 0.43 to 13.15 µg N m\(^{-3}\). 87.5% of samples were >1 µg N m\(^{-3}\) and 31.3% of samples >4 µg N m\(^{-3}\). Cape et al. (2009) showed that the annual critical level of NH3 for higher plants was 2–4 µg N m\(^{-3}\) and only 1 µg N m\(^{-3}\) for some sensitive species such as mosses and lichens. Thus gaseous NH3 concentrations at our site exceed the critical level. Indeed, previous researches have showed that Betula Luminifera and Pleioblastus Amarus forest, two local dominant tree species in the ecological barrier constructed in the upper reaches of Yangtze River, have been negatively affected by gradually increasing N deposition (Tu et al., 2009; Luo et al., 2010). The hilly area of Sichuan is not only important for agriculture, but also the major ecological barrier area in the upper reaches of the Yangtze River due to its location at the frontier of the Three Gorges Reservoir. Large amounts of N deposition into this area would cause great stress to local ecosystems, especially the fresh water ecosystem and environment of the Three Gorges Reservoir. Fortunately, Eshleman et al. (2013) have found that N deposition reduction could very effectively reduce surface water pollution in a forest watershed. Controls on reactive N emissions could relieve the pollution pressure on fresh water ecosystems in the environment of the Three Gorges Reservoir.

### 4.5. Contrasting methods for measuring dry deposition and uncertainties in N deposition calculations

#### 4.5.1. Comparing measurement methods

Two approaches can be used to measure dry deposition. One is

<table>
<thead>
<tr>
<th>Table 2 Report N deposition at various sites in China.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Location</td>
</tr>
<tr>
<td>---</td>
</tr>
<tr>
<td>Yanting, Sichuan</td>
</tr>
<tr>
<td>Changsha, Hunan</td>
</tr>
<tr>
<td>Changsha, Hunan</td>
</tr>
<tr>
<td>Jiangsu</td>
</tr>
<tr>
<td>Jiu zhai National nature reserve</td>
</tr>
<tr>
<td>Northern China</td>
</tr>
<tr>
<td>Fengxian, Hefei</td>
</tr>
<tr>
<td>Jiangsu</td>
</tr>
<tr>
<td>Shenyang, Liaoning</td>
</tr>
<tr>
<td>Yingtan, Jiangxi</td>
</tr>
<tr>
<td>Tai Lake region</td>
</tr>
<tr>
<td>North China Plain</td>
</tr>
<tr>
<td>Subtropical South China</td>
</tr>
</tbody>
</table>

w: wet deposition; d: dry deposition; b: bulk deposition.
the surface analysis method, which estimates contaminant accumulation on natural or manmade surfaces, and the other is the atmospheric flux method, which estimates the areal average rate of depletion from the atmosphere to all of the surfaces beneath (Yi et al., 1997); the collection bucket method represents the former, and the DELTA system the latter. Total dry N deposition collected with the DELTA system (DELTA-T) averaged 7.3 kg N ha$^{-1}$ yr$^{-1}$, while that collected by the bucket averaged 6.0 kg N ha$^{-1}$ yr$^{-1}$ in this study (unpublished data of Yang-ting), 21.7% lower than that of the DELTA system (Fig. 7). Tang et al. (2009) showed that the particle cut-off size for the DELTA system was 4–5 μm. Large particulate matter of diameter >10 μm is deposited by gravity, defined as sedimentary deposition (Tang et al., 2006) and will be captured by the bucket. Also, some of the small particles will be captured by the bucket lining through interception, but it is hard to quantify the amount and the cut-off size of this material, although the quantity is very small. In addition, the relative humidity in Yan-ting was 75% (Meteorological data of Yan-ting, auto-observed by Milos 520, VAISALA, made in Finland), and the dissolution ratio of gaseous NH$_3$ in water is 700:1, moreover, HNO$_3$ could be dissolved with water in any proportion, indicating that a lot of dew could form on the bucket lining gaseous NH$_3$ or HNO$_3$ dissolve in it; this is also difficult to quantify. However, because dry deposition was collected once a month, the effect of dew capture could be relatively small because the dew only absorbed limited N content due to much smaller volume of dew compared with that of rainfall or snowfall. In general, dry N deposition collected by the bucket contained particles of diameter >10 μm, some gaseous pollutants and some aerosols but of unknown quantity and cut-off size. The amount of particulate matter collected by the DELTA system (DELTA-P) was much lower than that collected by the bucket (Fig. 7). Thus the bucket is more effective for larger particles and the DELTA system for gaseous material.

4.5.2. Uncertainties of N deposition collection and calculation

Dry deposition fluxes were estimated from measured concentrations and calculated $V_d$ using the meteorological data from Yan-ting and the deposition model. The $V_d$ of NH$_3$, HNO$_3$, NO$_2$ and particulate matter (pNH$_4$ and pNO$_3$) in Yan-ting were estimated at 0.28, 0.77, 0.11 and 0.17 cm s$^{-1}$, respectively. Variations of $V_d$ was large because of differences in the meteorological conditions and the underlying surfaces. Previous research had shown that the $V_d$ of NH$_3$, HNO$_3$, NO$_2$ and particulate matter in agricultural systems in China varied from 0.06 to 0.74, 0.63 to 2.00, 0.05 to 0.59 and 0.03–0.25 cm s$^{-1}$, respectively (Xu et al., 2015; Shen et al., 2009; Pan et al., 2012; Yang et al., 2010). The $V_d$ in Yan-ting was in the lower range. Even so, some uncertainties still exist, such as how the underlying surface parameters (e.g., crop types and growth differences between plots) influenced $V_d$ and the ability of the ground surface to capture dry N deposition (Loubet et al., 2008).

In addition, Sutton et al. (1994) showed that NH$_3$ is both emitted from and deposited to land and water surfaces, i.e. NH$_3$ fluxes near canopies are bi-directional and the net direction of this flux is often uncertain. The compensation point (Farquhar et al., 1980; Sutton et al., 1994) has been used to determine bi-directional fluxes of NH$_3$ near a plant canopy: when NH$_3$ concentrations in air are lower than the NH$_3$ compensation point, emission occurs; if higher, NH$_3$ is deposited. Since bi-directional NH$_3$ exchange was not considered in this study, NH$_3$ deposition may be overestimated because of the relatively high NH$_3$ compensation point of 5 μg N m$^{-3}$ in fertilized cropland (Sutton et al., 1993).

In contrast, total N deposition may be underestimated due to the particle cut-off size of dry deposition. Tang et al. (2009) showed that the particle cut-off size for the DELTA system was 4–5 μm, and so the DELTA sampler does not capture the larger diameter particles. Our results showed that particulate dry N deposition in the bucket was 2.8 times that of the DELTA system. Another complication is that previous research used wet-only (Jiang et al., 2013; Luo et al., 2002) or bulk (Q et al., 2013; Yang et al., 2010) collectors to estimate wet N deposition or total N deposition, and only a few reports distinguished wet and sedimentary dry deposition (Shen et al., 2013; Anatolaki and Tsitouridou, 2007). Thus measurement methods are inconsistent and likely to lead to very different estimations of N deposition. Liu et al. (2006) showed that the ratio of wet-only to bulk deposition was 0.7 on average, and wet-only deposition of inorganic N was 8.3–8.4 kg N ha$^{-1}$ less than that of bulk deposition during 2003 and 2004 in the Dongbei area of Beijing. However, the ratio was different in other areas (Zhang et al., 2008b). In our study, the ratio of wet-only to bulk deposition was 0.8 on average, some large particles were measured in bulk deposition but not contained in wet-only deposition, so if wet-only deposition was used to calculate the total N deposition, it might be underestimated.

5. Conclusions

Five years of continuous observations at Yan-ting showed that dissolved N was the main component of N deposition in this typical agricultural area of the central Sichuan Basin. Reduced N and oxidized N accounted for 50.9% and 30.3% of TN, ammonia induced atmospheric pollution, especially that from agriculture, should be arousing attention. N deposition is increasing and may lead to regional environmental degradation. Rainwater containing high concentrations of N has become a threat to surface water quality and so to the natural and semi-natural ecosystems of the upper reaches of the Yangtze River. It is therefore urgent to improve N management in agro-ecosystems, especially to prevent emissions of gaseous ammonia. Finally, although there has been considerable research into N deposition in China, uncertainties still exist in the current estimation of N deposition, especially in hilly or mountainous areas where topography strongly affects deposition. Nationwide, long-term, continuous and uniform standard research into N deposition needs to be implemented urgently in China.

Acknowledgements

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Eshleman, K.N., Sabo, R.D., Kline, K.M., 2013. Surface water quality is improving due to their cooperation during sample collection and analysis.


