Annual Variation Characteristics of Atmospheric Reactive Nitrogen Dry and Wet Deposition in Dryland Farming Areas of Taiyuan City, Shanxi Province (Postprint)

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Abstract

Given the important impact of atmospheric nitrogen deposition on the entire ecosystem, China has successively carried out research on atmospheric nitrogen dry and wet deposition at different scales in recent years, but there is little data from multi-year continuous monitoring in agricultural areas. This study conducted a 4-year monitoring experiment using the DELTA system, passive samplers, and rain gauges in the dryland farming area of He Village, Yangqu County, on the outskirts of Taiyuan City, Shanxi Province, to observe the temporal variation of atmospheric nitrogen dry and wet deposition. The results showed that: from April 2011 to March 2015, the average deposition fluxes of atmospheric reactive nitrogen NH3, HNO3, NO2, particulate NO3 (pNO3), and particulate NH4+ (pNH4+) in He Village over the four years were 4.50 kg(N) · hm 2 · a 1, 3.54 kg(N) · hm 2 · a 1, 2.56 kg(N) · hm 2 · a 1, 1.62 kg(N) · hm 2 · a 1, and 2.75 kg(N) · hm 2 · a 1, respectively, with total atmospheric nitrogen dry deposition ranging from 12.38~18.95 kg(N) · hm 2 · a 1, peaking in 2011 and reaching its lowest in 2014. There was a significant positive correlation between monthly nitrogen dry deposition and ammonia deposition from April 2011 to March 2015, with correlation coefficients ranging from 0.809 8~0.937 1, indicating that reactive nitrogen deposition in this region is mainly influenced by agricultural ammonia emissions. The average concentrations of NO3 and NH4+ in rainwater in He Village over the four years were 3.20 mg(N) · L 1 and 2.43 mg(N) · L 1, respectively, with atmospheric nitrogen wet deposition ranging from 11.67~41.31 kg(N) · hm 2 · a 1. There were substantial interannual variations in nitrogen wet deposition, with the highest annual wet deposition occurring in 2012 and the lowest in 2014, with atmospheric nitrogen wet deposition accounting for more than 50% of total nitrogen deposition each year. Furthermore, over

the four years, not only was there a significant linear or quadratic correlation between NO3 -N and NH4+-N in wet deposition, but both also showed significant linear or quadratic relationships with precipitation amount, indicating that precipitation has a substantial influence on the wet deposition of NO3 -N and NH4+-N. This study demonstrates that in the dryland farming areas of Taiyuan City, interannual variation in nitrogen wet deposition is greater than that of dry deposition, and the total deposition amount is relatively high. Although it is a dryland farming area, nitrogen dry deposition in this region is slightly lower than wet deposition. The research results provide a theoretical basis for farmland nitrogen fertilizer application and nitrogen cycle monitoring in this region.

Full Text

Temporal Variation of Atmospheric Wet/Dry Reactive Nitrogen Deposition in Taiyuan Rainfed Farming Area of Shanxi Province

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Abstract: Atmospheric nitrogen (N) deposition exerts significant impacts on entire ecosystems. Although numerous studies on atmospheric N deposition have been conducted across various scales in China in recent years, multi-year continuous monitoring data from agricultural regions remain scarce. This study employed the DELTA system, passive samplers, and rain gauges to conduct a four-year monitoring experiment (April 2011–March 2015) in Hecun Village, a rainfed agricultural area in Yangqu County, Taiyuan City, Shanxi Province, to investigate temporal variation in atmospheric N deposition. The results showed that over the four-year period, the average annual deposition rates of reactive nitrogen species NH , HNO , NO , particulate NO , and particulate NH were 4.50 kg(N) · hm ^{2} · a ^{1}, 3.54 kg(N) · hm ^{2} · a ^{1}, 2.56 kg(N) · hm ^{2} · a ^{1}, 1.62 kg(N) · hm ^{2} · a ^{1}, and 2.75 kg(N) · hm ^{2} · a ^{1}, respectively. Total atmospheric N dry deposition ranged from 12.38 to 18.95 kg(N) · hm ^{2} · a ^{1}, peaking in 2011 and reaching its minimum in 2014. Monthly dry N deposition exhibited significant positive correlations with ammonia deposition across all years (April 2011–March 2015), with correlation coefficients ranging from 0.8098 to 0.9371, indicating that reactive N deposition in this region is primarily influenced by agricultural ammonia emissions. The average concentrations of NO -N and NH -N in rainwater were

3.20 mg(N) · L ¹ and 2.43 mg(N) · L ¹, respectively. Atmospheric N wet deposition ranged from 11.67 to 41.31 kg(N) · hm ² · a ¹, showing substantial interannual variation with the highest annual wet deposition occurring in 2012 and the lowest in 2014. Wet deposition accounted for over 50% of total N deposition each year. Furthermore, both NO -N and NH -N in wet deposition showed significant linear or quadratic relationships with rainfall amount, demonstrating that precipitation strongly influences wet deposition of these N species. These findings reveal that wet N deposition exhibited greater interannual variability than dry deposition in this Taiyuan rainfed farming area, though total deposition levels remained high. Despite being a dryland region, wet deposition slightly exceeded dry deposition. These results provide a theoretical basis for rational N fertilizer application and N cycling monitoring in regional farmland.

Keywords: Reactive nitrogen; Dry deposition; Wet deposition; Annual variation; Rainfed farming area

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Introduction

Excessive anthropogenic emissions of reactive nitrogen [primarily NH , NO , HNO , particulate NH , and NO] have caused atmospheric reactive nitrogen (Nr) pollution and deposition, emerging as a global environmental concern. With China' s rapid socioeconomic development, Nr emissions have increased at an annual rate of 3.7%, reaching 56 Tg (1 Tg = 10^{12} g) by 2010, accompanied by sharp rises in atmospheric NH and NOx emissions. Consequently, some regions of China have become hotspots for high N deposition.

Excessive N application in Chinese croplands is widely recognized as causing serious environmental problems, drawing considerable attention to N balance issues in agricultural ecosystems. Studies indicate that agriculture represents the primary source of atmospheric N deposition in China, with national N deposition increasing by 60% by 2010 compared to the 1980s. N deposition research in China began relatively late, with atmospheric wet deposition monitoring starting in the 1970s, establishment of a national monitoring network in the 1990s, and expanded monitoring across farmland, grassland, forest, and urban ecosystems beginning in 2004.

Current N deposition research in China has focused primarily on wet deposition, with only sporadic reports on dry deposition, most of which address deposition fluxes of specific N compounds rather than providing continuous multi-year monitoring results from typical agricultural zones. As a major energy base in China with extensive rainfed agricultural areas, Shanxi Province exhibits unique characteristics in atmospheric Nr emissions, yet related research remains in its infancy. This study selected Hecun Village in Yangqu County, Taiyuan City—a rainfed agricultural area—as a sampling site to comprehensively monitor atmospheric reactive N pollution and its contributions to dry and wet deposition using the DELTA system and rain gauges. Through four years of continuous

monitoring of atmospheric reactive N in suburban farmland areas of Taiyuan, Shanxi Province, this research quantified local N inputs via dry and wet deposition and elucidated relationships among Nr species, thereby providing baseline data for estimating and predicting atmospheric Nr deposition in agricultural regions across Shanxi and nationwide.

1.1 Sampling Site Description

The monitoring site was established in Hecun Village, Lingjingdian Township, Yangqu County, Taiyuan City, Shanxi Province (112.89°E, 38.05°N), a typical rainfed agricultural area. Located 30 km north of Taiyuan' s suburbs, the site receives an average annual precipitation of 440 mm, with a mean annual temperature of 10.1°C, a frost-free period of 120–140 days, and average wind speed of 2.1 $m \cdot s^{1}$ (predominantly northwesterly winds). The area comprises 451.4 hm^2 of cultivated land with a population density of 58 $persons \cdot km^{2}$, practicing agriculture entirely dependent on natural precipitation. The sampling period extended from April 2011 to March 2015 (with 2011 representing April 2011–March 2012, 2012 representing April 2012–March 2013, and so forth). Dry deposition samples were collected monthly. Both dry and wet deposition instruments were placed at the Hecun rainfed agricultural base, where the primary crop is maize (*Zea mays*) with average fertilizer application rates of 240–275 $kg(N) \cdot hm^{2}$ and 105–130 $kg(P\ O) \cdot hm^{2}$. No other pollution sources existed in the sampling area.

1.2 Dry Deposition Samplers

Dry deposition sampling equipment included one DELTA system and three passive samplers. The DELTA system collected reactive N gases (NH , HNO) and particulate NH and NO , while passive samplers collected NO gas concentrations. These five reactive N forms were used to calculate individual and total N dry deposition fluxes.

1.2.1 DELTA System The DELTA (DEnuder for Long-Term Atmospheric sampling) system, developed by the Centre for Ecology and Hydrology (CEH), employs a diffusion tube-filter pack combination for long-term monitoring of NH , HNO , particulate NH , and NO concentrations. This system effectively separates gaseous NH and HNO from aerosols, substantially reducing errors associated with filter pack methods. Due to its relatively low cost, the DELTA system has been widely applied in environmental monitoring across the UK and Europe. A small pump in the system draws air at a flow rate of 0.3–0.4 $L \cdot min^{1}$, allowing reactive N components to be adsorbed by coatings on diffusion tubes while aerosol particles are collected on downstream filters. Specifically, the sampling train uses 1% KOH + 1% glycerol methanol solution for nitric acid collection, 5% citric acid methanol solution for ammonia collection, 5% KOH + 10% glycerol methanol solution for particulate NO collection, and 13% citric acid methanol solution for particulate NH collection. Further details regarding

the DELTA system are available in the literature [14]. The sampling height was 1.6 m above ground. Due to severe air pollution in China, this study employed 37 mm filters to collect NO and NH particles, larger than the 25 mm filters used in European monitoring.

Samples were collected over monthly intervals (from the beginning to the end of each month). After collection, samples were immediately stored at 4°C and analyzed within one month. HNO and NO particles were extracted with 10 mL of high-purity 0.05% H O , while NH and NH particles were extracted with 10 mL of deionized water. After dissolution, NO and NH concentrations were determined using an AA3 continuous flow analyzer (Bran+Luebbe GmbH, Norderstedt, Germany).

1.2.2 Passive Samplers for Atmospheric NO Collection Atmospheric NO was collected using passive samplers following the standard method employed by the UK Environmental Change Network (ECN). The passive samplers were impregnated with 20% triethanolamine as an adsorbent for NO gas. Samplers were deployed at 2 m height with monthly sampling durations of 10–14 days synchronized with other samplers. After two weeks of exposure, samplers were retrieved, stored at 4°C, and analyzed for collected NO concentration using the standard colorimetric method described on the ECN website (www.ecn.ac.uk).

1.2.3 Calculation of Atmospheric N Dry Deposition Atmospheric N dry deposition was calculated using the inferential model method, where dry deposition flux over a given period equals the product of atmospheric reactive N concentration and deposition velocity [15], expressed as:

$$F = C \times V_d$$

where F represents atmospheric N dry deposition flux [kg(N) · hm ^{2}], C represents atmospheric reactive N concentration [g(N) · m ^{3}], and V_d represents deposition velocity for different reactive N species [cm · s ^{1}].

Atmospheric N dry deposition velocities, which vary substantially among different N gases and between gases and particles, were typically obtained from atmospheric resistance models. For the period April 2011–March 2013, this study adopted deposition velocities for rural Yangqu County published by Xu et al. [16] (calculated through a collaborative model between China Agricultural University and Peking University). Since deposition velocities show minimal interannual variation but substantial monthly differences, average deposition velocities from the first three years were applied for the period April 2013–March 2015. Monthly deposition velocities for each reactive N species are presented in .

1.3 Wet Deposition Collection

Rainwater samples were collected after each rainfall event using a rain gauge (SDM6, Tianjin meteorological equipment). The rain gauge was placed adjacent to the DELTA system in an open, flat area. Collected rainwater was frozen and analyzed within one month. NH -N and NO -N concentrations were determined using the methods described above. Inorganic N wet deposition was calculated based on N concentrations and precipitation amounts [17] using the formula:

$$\mathrm{wdN} = P \times \mathrm{NC} \times 10$$

where wdN represents N wet deposition per rainfall event [g(N) · hm ^{2}], P represents precipitation amount per event (mm), NC represents NH -N or NO -N concentration [mg(N) · L ^{1}], and 10 is a unit conversion coefficient. Monthly or annual N wet deposition (m, yN) [kg(N) · hm ^{2}] equals the sum of N wet deposition from all rainfall events during that period.

1.4 Data Processing

All experimental data were processed and graphed using Microsoft Excel 2003 software.

2.1 Concentrations and Monthly Deposition Fluxes of Reactive N Species in Atmospheric Dry Deposition

The four-year average concentrations of atmospheric NH , HNO , NO , particulate NO (pNO), and particulate NH (pNH) in the study area were 4.17 g(N) · m ^{3}, 1.49 g(N) · m ^{3}, 6.10 g(N) · m ^{3}, 2.58 g(N) · m ^{3}, and 4.10 g(N) · m ^{3}, respectively, with NO showing the highest concentration among the five reactive N components. Due to substantial differences in monthly deposition velocities between gases and particles, the annual deposition fluxes of NH , HNO , NO , pNO , and pNH were 4.50 kg(N) · hm ^{2} · a ^{1}, 3.54 kg(N) · hm ^{2} · a ^{1}, 2.56 kg(N) · hm ^{2} · a ^{1}, 1.62 kg(N) · hm ^{2} · a ^{1}, and 2.75 kg(N) · hm ^{2} · a ^{1}, respectively. Oxidized N deposition (HNO , NO , and pNO) slightly exceeded reduced N deposition (NH and pNH), with a ratio of 1.06. Total atmospheric N dry deposition flux ranged from 12.38 to 18.95 kg(N) · hm ^{2} · a ^{1}, averaging 14.98 kg(N) · hm ^{2} · a ^{1} and accounting for 28–51% of total annual N deposition flux. Regarding individual reactive N contributions to dry deposition, the agricultural monitoring site showed the pattern: gaseous NH > NO , and particulate pNH > pNO .

Interannual comparison in [Figure 1: see original paper] reveals that monthly N dry deposition was highest in 2011 and lowest in 2014. [Figure 2: see original paper] shows that except for the three winter months, N dry deposition varied little across other months. Maximum dry deposition flux occurred in May and June (most frequently in May) across all years except 2011, coinciding with ammonia peaks in May and June. This indicates that N deposition increased during

the concentrated fertilizer application period, demonstrating that agricultural fertilization significantly influences N dry deposition in this region. Further analysis revealed significant positive correlations between monthly N dry deposition and ammonia deposition across all years from 2011 to 2015, with an overall correlation coefficient of 0.8112 [Figure 3: see original paper]. Although NO deposition ranked second in magnitude, its correlation with total reactive N deposition was not statistically significant. These findings confirm that total reactive N deposition in this region is strongly influenced by agricultural ammonia emissions.

2.2 Variation in Atmospheric N Wet Deposition and Relationships Between Inorganic N Species in Rainwater

Wet deposition has long been a research focus, with studies demonstrating close relationships between N wet deposition and rainfall amount, as well as substantial interannual variation. Rainfall in Hecun Village ranged from 285.25 to 523.87 mm during 2011–2015, with average NO -N and NH -N concentrations in rainwater of 3.20 mg(N) · L ^{1} and 2.43 mg(N) · L ^{1}, respectively. Atmospheric inorganic N wet deposition ranged from 11.67 to 41.31 kg(N) · hm ^{2} · a ^{1}, with NO -N accounting for 39–51% and NH -N comprising 49–61% of the total. Annual wet deposition was highest in 2012 and lowest in 2014 [Figure 4: see original paper], representing 49–72% of respective annual total N deposition. Despite being a dryland agricultural area, wet deposition exceeded 50% of total N deposition, suggesting that dryland characteristics were not pronounced.

[Figure 5: see original paper] illustrates significant positive linear relationships between monthly NO -N and NH -N deposition during 2011–2013, shifting to quadratic relationships in 2014. [Figure 6: see original paper] further demonstrates significant positive linear or quadratic relationships between rainfall and both NO -N and NH -N deposition across the four years, with average correlation coefficients of 0.7815 and 0.6783, respectively. This indicates that over two-thirds of NO -N and NH -N wet deposition depended on rainfall amount.

3 Discussion and Conclusion

Atmospheric N dry deposition constitutes an important component of total N deposition, accounting for 10–90% of N deposition in Europe. With methodological advances and increased research attention, dry deposition studies have gradually expanded across China in recent years. This study revealed that average atmospheric N dry deposition flux in Hecun Village, Lingjingdian Township, Yangqu County, Taiyuan City, reached 14.98 kg(N) · hm ^{2} · a ^{1} during April 2011–March 2015, representing 28–51% of total annual N deposition flux. Dry deposition showed a declining trend during the monitoring period, possibly related to reduced fertilizer inputs following lower maize prices in recent years. Significant positive correlations (r = 0.8112) between monthly N dry deposition and ammonia deposition were observed across all four years, with ammonia peaks occurring during the concentrated fertilization period of May–June. Atmospheric

NH -N is generally derived from volatilization of soil, fertilizer, and livestock manure NH , as well as combustion of nitrogenous organic matter, while NO -N originates primarily from industrial activities, vehicle emissions, and natural oxidation of nitrogen (e.g., lightning). Liu et al. [22] also noted that other emission sources such as ammonia from livestock facilities and traffic generally show higher concentrations during the hot season (July–August). These findings indicate that N dry deposition in this region is strongly influenced by agricultural ammonia emissions, consistent with conclusions from nationwide atmospheric Nr monitoring by Xu et al. [16] showing highly significant positive correlations between annual emissions and deposition of NH and NO . Additionally, the four-year average molar concentration ratio of NH /NO was 1.9 (>1), suggesting substantial contributions from agricultural sources [23]. Considering the absence of industrial pollution sources near the sampling area and the singular nature of Nr emission sources, agricultural fertilizer application appears to significantly influence N dry deposition in this region.

Although a dryland area, wet deposition exceeded dry deposition in some years, accounting for 49–72% of respective annual total N deposition. By contrast, a 2010 study in Gucheng Town, Shanxi Province, reported dry deposition three times greater than wet deposition [24], while research across different ecological zones in Shaanxi Province indicated wet deposition exceeding 62% [25–26]. Compared with other domestic studies, the average concentrations of inorganic N in rainfall (NO -N: 3.20 mg(N) · L ^{1}; NH -N: 2.43 mg(N) · L ^{1}) were higher than results from eastern and southern China [27]. Except for 2012, atmospheric wet deposition in this study was lower than levels reported for forest sites in Zhaoqing, Guangdong, farmland areas in Zhejiang, and urban sites in Shanghai and Guangzhou [28–30]. However, compared with N inputs from precipitation in northern China (North China, Northeast, and Northwest regions) ranging from 5.1 to 25.4 kg(N) · hm ^{2} · a ^{1} [31–34], some years in this study exceeded the upper limit of 25.4 kg(N) · hm ^{2} · a ^{1}. Consistent with other researchers' findings, rainfall showed significant positive linear or quadratic relationships with NO -N and NH -N deposition, with average correlation coefficients of 0.7815 and 0.6783, respectively, confirming that rainfall significantly influences N wet deposition [26,35–36]. The linear and quadratic positive correlations between NO -N and NH -N in rainwater suggest these N species exist in relatively stable forms in precipitation.

Regular monitoring of atmospheric N inputs in Hecun Village, Yangqu County, Taiyuan suburbs over four years revealed that wet deposition of NH -N and NO -N and dry deposition of NH , HNO , NO , pNH , and pNO contributed 26.7%, 33.4%, 12.0%, 9.4%, 6.8%, 7.3%, and 4.3% of total deposition, respectively, with dry deposition averaging 39.9% of total deposition. Atmospheric N dry deposition ranged from 12.38 to 18.95 kg(N) · hm ^{2} · a ^{1}, while wet deposition ranged from 11.67 to 41.31 kg(N) · hm ^{2} · a ^{1}. The average total deposition was slightly lower than the national average of 39.9 kg(N) · hm ^{2} · a ^{1} from 43 monitoring sites [16] and below some southern urban sites such as typical farmland in the Leizhou Peninsula (42.9 kg(N) · hm ^{2} · a ^{1}) [37], but slightly higher than

atmospheric N deposition (including dry and wet) in subtropical forest and farmland areas of Hunan (22–55 kg(N) · hm ^{2} · a ^{1}) [15]. Although some regions show wet deposition exceeding 90.4% of total deposition [25], increasing research on atmospheric Nr dry deposition demonstrates its substantial contribution to total deposition [26], a conclusion confirmed by this study. Moreover, this study' s dry deposition velocities, calculated through a collaborative model between China Agricultural University and Peking University, provide higher accuracy than previous estimates. In summary, the four-year average total N deposition of 38.9 kg(N) · hm ^{2} · a ^{1} far exceeds the critical load of 10 kg(N) · hm ^{2} · a ^{1} at which N deposition significantly impacts terrestrial ecosystems, indicating that current N deposition levels will substantially influence nutrient cycling in this farmland ecosystem [38–39]. This monitoring effort provides important data for rationally estimating atmospheric N inputs when applying N fertilizers or monitoring N cycling in regional farmland.

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Note: Figure translations are in progress. See original paper for figures.