Atmospheric deposition of inorganic nitrogen in semi-arid grasslands of Inner Mongolia, China: Postprint

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Abstract

Due to increasing global demand for crop production and energy use, more and more reactive nitrogen (Nx) has been generated and emitted to the environment. As a result, global atmospheric nitrogen (N) deposition has tripled since the industrial revolution and the ecological environment and human health have been harmed. In this study we measured dry and wet/bulk N deposition from July 2013 to December 2015 in a semi-arid grassland of Duolun County Inner Mongolia, China. The samples of dry and wet/bulk N deposition were collected monthly with a DELTA (DEnuder for Long Term Atmospheric sampling system) and with Gradko passive samplers and a precipitation gauge. The measured results show that the annual mean

Full Text

Abstract

Atmospheric Deposition of Inorganic Nitrogen in a Semi-Arid Grassland of Inner Mongolia, China

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Abstract: Due to increasing global demand for crop production and energy use, more reactive nitrogen (Nr) has been generated and emitted to the environment, causing global atmospheric nitrogen (N) deposition to triple since the

Industrial Revolution with consequent harm to ecological environments and human health. This study measured dry and wet/bulk N deposition from July 2013 to December 2015 in a semi-arid grassland of Duolun County, Inner Mongolia, China. Samples were collected monthly using a DELTA (DEnuder for Long Term Atmospheric sampling) system, Gradko passive samplers, and a precipitation gauge. The annual mean atmospheric concentrations of NH_3, NO_2, HNO_3, particulate NH_4^+, and particulate NO_3^- were 2.33, 1.90, 0.18, 1.42, and 0.42 g N/m^3, respectively. The annual mean volume-weighted concentrations of NH_4^+-N and NO_3^--N in precipitation were 2.71 and 1.99 mg N/L, respectively. Concentrations of Nr components exhibited distinct seasonal variations: NO_2 and HNO_3 showed higher concentrations in autumn than summer, while other Nr components (NH_3, pNH_4^+, pNO_3^-, NH_4^+-N, and NO_3^--N) peaked in summer. Using measured concentrations and deposition velocities estimated from the GEOS-Chem global atmospheric chemical transport model, the calculated annual mean dry deposition fluxes were 3.17, 1.13, 0.63, 0.91, and 0.36 kg $N/(hm^2 \cdot a)$ for NH_3, NO_2, HNO_3, pNH_4^+, and pNO_3^-, respectively. The calculated annual mean wet/bulk deposition fluxes were 5.37 and 3.15 kg $N/(hm^2 \cdot a)$ for NH_4^+-N and NO_3^--N, respectively. The estimated total annual N deposition (dry + wet/bulk) reached 14.7 kg $N/(hm^2 \cdot a)$ in the Duolun County grassland, approaching the upper limit of the N critical load (10–15 kg $N/(hm^2 \cdot a)$). Dry and wet/bulk deposition fluxes of all Nr components (except HNO_3) showed similar seasonal patterns, with maximum fluxes in summer and minimum in winter. Reduced Nr components (gaseous NH_3 and pNH_4^+ in the atmosphere and NH_4^+-N in precipitation) dominated total N deposition at the sampling site (accounting for 64% of total N deposition), suggesting that deposited atmospheric Nr originated mainly from agricultural activities. Considering projected future increases in crop and livestock production in Inner Mongolia, ecological and human health risks from increased N deposition could rise without mitigation measures.

Keywords: atmospheric reactive nitrogen; dry deposition; wet/bulk deposition; reduced nitrogen; grassland ecosystem; Inner Mongolia

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1 Introduction

Nitrogen (N) is a primary controlling factor for net primary productivity in many ecosystems, particularly in regions with low N supply [**?**, **?**]. It plays a significant role in increasing food production and stimulating plant growth [**?**, **?**]. However, intensifying human activity has generated and emitted substantial reactive nitrogen (Nr) to the environment, altering ecosystem N balances. Large-scale fossil fuel consumption, heavy chemical N fertilizer application, population growth, and intensive livestock production have caused dramatic Nr emission increases in recent decades [**?**, **?**]. Consequently, atmospheric N emissions rose

from 34 Tg N in 1860 to 109 Tg N in 2010, with projections reaching 270 Tg N by 2050 [?, ?]. After release into the atmosphere, Nr compounds undergo various physical and chemical transformations before wet or dry deposition, adversely affecting ecological environments and human health.

In China, rapid economic development through agricultural and industrial expansion has sharply increased N deposition [?]. An integrated analysis of historical and monitoring data revealed that annual bulk N deposition increased by approximately 8 kg N/hm^2 in the 2000s compared to the 1980s, with this trend persisting [?]. Jia et al. [?] evaluated current dry N deposition rates in China using satellite observations, finding rates up to 7.5 Tg N/a, consistent with recent model findings of 6.4 Tg N/a [?]. Excessive N deposition causes numerous environmental problems, including soil acidification, lake eutrophication, biodiversity loss, and increased N_2O emissions [?, ?, ?]. Furthermore, NH_3 and NO are important precursors for secondary inorganic aerosol formation and haze development [?, ?].

Grassland is China' s most widely distributed vegetation type, covering nearly 40% of the country' s surface area [?]. The Inner Mongolia grassland, an important component of this system, significantly contributes to the global carbon cycle and serves as a crucial ecological barrier in North China, vital for biodiversity conservation. However, over-fertilization and unsustainable land use have caused ongoing land degradation and desertification [?]. Numerous field experiments in China [?, ?] and abroad [?] have documented negative impacts of N deposition on grassland biodiversity, including altered community structure and reduced net primary productivity [?, ?]. However, some studies have questioned these negative impacts, suggesting they may be artifacts of experimental manipulations [?, ?].

Chinese scientists have conducted studies quantifying N deposition impacts on grasslands [?, ?, ?], but long-term continuous observations of wet/bulk and dry deposition in Inner Mongolia grasslands remain scarce. Most existing observations come from the North China Plain [?, ?, ?, ?], Yangtze River Delta [?], and Pearl River Delta [?]. To fill these geographic research gaps and provide data for national N deposition evaluation, we conducted a three-year study in Duolun County, southeastern Inner Mongolia grassland. The objectives were: (1) to estimate Nr component concentrations and deposition fluxes, and (2) to characterize seasonal variations in dry and wet/bulk N deposition.

2.1 Study Site

The study was conducted in a semi-arid grassland in Duolun County, Inner Mongolia Autonomous Region, China. The sampling site is located at a meteorological observation station of the Chinese Academy of Sciences (42°22 N, 116°49 E; 1,324 m a.s.l.). The region features a typical semi-arid continental monsoon climate with average annual precipitation of 380 mm, peaking in July. The growing season generally runs from May through September (approximately

150 days), accounting for 88% of annual precipitation. The annual average temperature is 2.2°C, with monthly averages ranging from -16.9°C in January to 19.4°C in July. The dominant soil type is chestnut soil. Livestock grazing and crop cultivation are the two major land-use practices in this area.

2.2 Sampling and N Deposition Analysis

2.2.1 Dry N Deposition

Reactive nitrogen (Nr) components, including NH_3, NO_2, HNO_3, particulate NH_4^+, and particulate NO_3^-, were sampled monthly from July 2013 to December 2015. NH_3, HNO_3, particulate NH_4^+, and particulate NO_3^- were measured using a DELTA (DEnuder for Long Term Atmospheric sampling) system. The DELTA system consisted of three parts: a circular sampling chain with diffusers and a particle collection device, a small low-volume pump (0.2–0.4 L/min), and a high-precision gas flow meter. System details have been described previously [**?**, **?**]. Briefly, when ambient air passes through the DELTA system, HNO_3 and other acidic gases are captured by two long diffusion tubes coated on the inner wall with a mixed solution of 10 g/L K_2CO_3 and 10 g/L glycerol in methanol, while NH_3 is captured by two short diffusion tubes coated with citric acid (5% m/v in methanol). Subsequently, acidic particulate aerosols are collected by a filter membrane with basic adsorbent in the DELTA train membrane system, and alkaline particulate aerosols are collected by a membrane with acidic adsorbent.

NO_2 was monitored with Gradko passive tubes (Gradko International Limited, Winchester, UK), composed of an acrylic acid tube (71.0 mm length, 11.0 mm internal diameter), two polyethylene caps (gray and white) at both ends, and two layers of stainless steel wire. NO_2 was absorbed into a 20% triethanolamine solution coated onto two stainless steel wire meshes in the gray cap. Sampling heights for the DELTA system and NO_2 diffusion tubes were maintained at 2 m above ground. Three NO_2 sampler replicates were deployed for monthly sampling. Exposed DELTA sample trains and NO_2 diffusion tubes were replaced monthly, preserved at 4°C, and analyzed immediately after collection. Gaseous HNO_3 collected in glass denuders and particulate NO_3^- on filter membranes were extracted using 10 mL of 0.05% H_2O_2 solution, while NH_3 and particulate NH_4^+ in denuders and membranes were extracted using 6 mL and 10 mL of high-purity water, respectively. An AA3 continuous flow analyzer (Bran+Luebbe GmbH, Norderstedt, Germany) measured NH_4^+ concentrations in extracted solutions. NO_2 in Gradko passive samplers was extracted with a mixed solution of sulfonamide, H_3PO_4, and N-1-naphthylethylene-diamine, with concentration determined by absorbance at 542 nm. For quality assurance, monthly blank samples were established both in the laboratory and field and analyzed using identical procedures.

2.2.2 Wet/Bulk N Deposition

Wet/bulk N deposition was collected using a precipitation gauge (SDM6, Tianjin Weather Equipment Inc., China) from May 2013 to November 2015. The precipitation gauge was a passive sampler without power input, consisting mainly of a stainless steel funnel and glass container. Precipitation was automatically collected and measured with a graduated cylinder. Before laboratory analysis at China Agricultural University, collected precipitation was recorded, mixed, and stored in clean polyethylene bottles (50 mL) at -18°C. The precipitation gauge was rinsed with high-purity water after each collection to prevent contamination. Precipitation samples were filtered through 0.45 m filter membranes; 15 mL of filtered fluid was frozen and stored in polyethylene bottles. NH_4^+-N and NO_3^--N concentrations were determined by continuous flow analyzer (Bran+Luebbe GmbH, Norderstedt, Germany) within one month.

2.3 Calculations of N Deposition Flux

Wet/bulk N deposition flux was calculated based on NH_4^+-N and NO_3^--N concentrations and precipitation amount per event. The volume-weighted mean concentration of Nr components (C_w; mg N/L) was obtained using Equation 1:

$$C_w = \frac{\sum_{i=1}^{n} C_i P_i}{\sum_{i=1}^{n} P_i}$$

where C_i is the inorganic Nr concentration in the ith precipitation event (mg N/L), P_i is precipitation amount in the ith event (mm), and n is the number of precipitation events.

The wet/bulk deposition flux of Nr components (D_w; kg N/hm^2) was calculated using Equation 2:

$$D_w = \frac{\sum_{i=1}^{n} C_i P_i}{100}$$

Dry N deposition is affected by various chemical and environmental factors including meteorological conditions, Nr component physicochemical properties, underlying surface roughness, and surface capture/absorption capacity [**?**]. Additional complexity arises from bidirectional N flux characteristics, making quantification challenging. Nr emission dominates when surface concentrations exceed atmospheric concentrations, while deposition occurs when the gradient reverses. Inferential models have been extensively used operationally to determine dry N deposition in monitoring networks worldwide (e.g., NADP, EMEP). Using this method, we obtained monthly dry deposition fluxes by multiplying measured Nr component concentrations with deposition velocities from a global

atmospheric chemical transport model simulation. Dry deposition flux was expressed by Equation 3:

$$F_d = C_z \times V_d$$

where F_d is dry deposition flux of an Nr component (kg N/hm^2), C_z is measured atmospheric concentration (g N/m^3), and V_d is dry deposition velocity (cm/s). V_d values were obtained from a nested-grid version of the Goddard Earth Observing System (GEOS)-Chem chemical transport model for Asia, with 0.50° latitude × 0.67° longitude spatial resolution and 6-hour temporal resolution. V_d estimation follows a standard big-leaf resistance-in-series model determined by meteorological conditions and land-use types (grassland in this study). Hourly V_d data were averaged to obtain monthly values for all Nr components. Model description and dry N deposition flux simulation details are available in Zhang et al. [**?**]. Note that some wet/bulk and dry N deposition results (e.g., July 2013–September 2014 data) were previously published by Xu et al. [**?**].

3.1 Concentrations of Nr Components in Atmosphere

Monthly atmospheric concentrations of NH_3, NO_2, HNO_3, pNH_4^+, and pNO_3^- ranged from 0.34–6.79, 0.24–6.92, 0.01–0.53, 0.84–5.63, and 0.33–1.48 g N/m^3, respectively, from July 2013 to December 2015 (Fig. 1 [Figure 1: see original paper]). NH_3 showed remarkable seasonal variation with an annual mean concentration of 2.33 g N/m^3, peaking in summer (June–August) and reaching minimum in winter (December–February). The annual mean NO_2 concentration was 1.90 g N/m^3, with highest values in April and lowest in January. The annual mean HNO_3 concentration was 0.18 g N/m^3. Compared to NH_3 and NO_2, HNO_3 monthly concentrations were lower and less variable, except notably higher values in September. The annual mean pNH_4^+ concentration was 1.42 g N/m^3, with maximum monthly concentration in July and minimum in January. Similar to pNH_4^+, pNO_3^- also showed highest values in July, with an annual mean concentration of 0.62 g N/m^3.

3.2 Concentrations of Nr Components in Precipitation

During the monitoring period (May 2013–December 2015), monthly volume-weighted concentrations of NH_4^+-N and NO_3^--N in precipitation varied from 0.27–12.19 and 0.10–11.68 mg N/L, respectively (Fig. 2 [Figure 2: see original paper]). Average annual concentrations were 2.71 mg N/L for NH_4^+-N and 1.99 mg N/L for NO_3^--N. NH_4^+-N concentrations generally exceeded NO_3^--N except in September and October 2013. Higher NO_3^--N concentrations occurred in months with lower precipitation (February–May), while lower concentrations accompanied higher monthly precipitation (June–September). Precipitation amount substantially affected NH_4^+-N and NO_3^--N concentrations.

3.3 N Deposition Flux

Monthly dry deposition velocities for each Nr component are listed in Table 1 . Based on these values, annual dry deposition fluxes were calculated for NH_3, NO_2, HNO_3, pNH_4^+, and pNO_3^- (Fig. 3 [Figure 3: see original paper]). Annual dry deposition fluxes were 3.17, 1.13, 0.63, 0.91, and 0.36 kg N/(hm^2 · a), respectively, during the monitoring period. Note that 2013 monitoring data were excluded from annual dry deposition flux calculations due to incompleteness. During the monitoring period (excluding 2013 data), the summed annual dry deposition flux for the five atmospheric Nr components averaged 6.21 kg N/(hm^2 · a). Due to high NH_3 concentration, its dry deposition flux was the largest among atmospheric Nr components, followed by NO_2, HNO_3, pNH_4^+, and pNO_3^-. NH_3 contributed 51% to total dry N deposition, exceeding the combined contribution of the other four Nr components (49%).

From 2013–2015, annual wet/bulk deposition fluxes ranged from 3.22–7.90 kg N/(hm^2 · a) for NH_4^+-N and 2.72–3.38 kg N/(hm^2 · a) for NO_3^--N. The average annual wet/bulk deposition flux was 5.37 kg N/(hm^2 · a) for NH_4^+-N and 3.16 kg N/(hm^2 · a) for NO_3^--N. The annual wet/bulk deposition flux averaged 8.53 kg N/(hm^2 · a). The total N deposition (dry + wet/bulk) flux averaged 14.73 kg N/(hm^2 · a), with NH_4^+-N deposition 1.7 times that of NO_3^--N.

4.1 Concentrations of Nr Components in Atmosphere and Precipitation

Intensive fossil fuel consumption to support China' s population and economic growth has generated large Nr emissions and subsequent N deposition to ecosystems. Simultaneously, lack of effective atmospheric Nr emission control measures has aggravated increasing N deposition trends [**?**]. Consequently, China has become one of the world' s N deposition hotspots [**?**] and faces continued serious Nr pollution threats.

This study found that monthly average Nr component concentrations in atmosphere and precipitation in Inner Mongolia grassland were considerably lower than those in developed regions such as the North China Plain [**?**, **?**] and Southwest China [**?**]. Due to fossil fuel burning and intensified agriculture and livestock production, the North China Plain has become a Chinese NH_3 hotspot [**?**, **?**], with the largest annual mean NH_3 concentration of 16.9 ± 5.9 g N/m^3 in Quzhou [**?**], far exceeding this study' s value of 2.33 g N/m^3. Our atmospheric NH_3 concentrations were higher than some European observations [**?**]. Although the sampling site' s annual NH_3 concentration fell within China' s regional background range [**?**], it exceeded or approached the critical level of 3 ($\pm1)\mu gN/m\hat{}\{3\}$ established for protecting fragile regions [**?**], posing potential environmental threats to Inner Mongolia grassland.

During the monitoring period, monthly NH_3 concentrations peaked in summer and reached minimum in winter. These seasonal variations resulted from human

activities and meteorological factors. Summer' s high temperatures enhanced microbial activity and ammonification, favored ammonia volatilization, and promoted NH_3 emissions from fertilizer application during nearby potato cultivation. During the growing season, herders applied N fertilizer to pastures, further elevating summer NH_3 concentrations. In winter, low temperatures severely restricted biological activity and fertilizer application was minimal, reducing ammonia volatilization. High winter wind speeds also promoted pollutant dispersion, further lowering NH_3 concentrations.

NO is primarily produced through various combustion processes including transportation, industry, coal use, and power plants [**?**, **?**]. Due to low industrialization at the sampling site, NO_2 concentrations were markedly lower than most urban Chinese sites [**?**]. However, the annual mean NO_2 concentration was more than double the previous value measured by Shen et al. [**?**]. Recent increases in vehicle numbers partially explain this rise—private cars in Inner Mongolia grew from 1.88$\times$10^{6}$ in 2011 to 3.35$\times$10^{6}$ in 2015 (National Bureau of Statistics of China, 2011–2015). Higher NO_2 concentrations observed in spring and autumn differed from typical seasonal patterns [**?**]. Elevated spring NO_2 concentrations likely resulted from coal combustion for domestic heating. HNO_3 showed high concentrations in autumn and low concentrations in summer, consistent with NO_2 seasonal variations.

The annual mean pNH_4^+ concentration was much lower than many Chinese [**?**] and Western European observations [**?**], but comparable to rural U.S. sites [**?**] and higher than UK reports [**?**]. Summer pNH_4^+ concentrations exceeded those in spring, autumn, and winter, likely caused by high NH_3 concentrations and relative humidity favoring gas-to-particle conversion, although increased temperatures did not favor ammonium nitrate formation. Due to low HNO_3 concentrations, pNO_3^- concentrations were lower than most Chinese measurements [**?**, **?**]. The seasonal pNO_3^- pattern matched pNH_4^+, with higher summer values and lower values in other seasons.

Inorganic N in precipitation primarily derives from water-soluble atmospheric Nr components scavenged by rain or snow. The sampling site's annual NH_4^+-N and NO_3^--N concentrations in wet/bulk deposition were lower than many Chinese urban and rural sites [**?**, **?**], but higher than a semi-arid region in Urumqi [**?**]. In this study, the highest monthly average NH_4^+-N concentrations occurred in summer and the lowest in winter, showing significant negative correlation with precipitation, suggesting a dilution effect [**?**, **?**]. Compared with previous data [**?**], annual NH_4^+-N concentration in precipitation increased by 19% while NO_3^--N decreased by 6% in Duolun County grassland.

4.2 Seasonal Variations of Wet/Bulk and Dry N Deposition

A distinct seasonal N deposition pattern was observed at the sampling site (Fig. 4 [Figure 4: see original paper]). Nr component deposition fluxes peaked in summer and reached minimum in winter, except for HNO_3 which showed maxi-

mum deposition flux in autumn. Summer NH_3 deposition fluxes were markedly higher than in spring, autumn, and winter, accounting for 57% of annual NH_3 deposition flux, consistent with NH_3 concentration seasonal variation. NO_2 and pNH_4^+ deposition fluxes in spring, autumn, and winter contributed equally to annual deposition. pNH_4^+ deposition dominated summer annual deposition, with pNO_3^- seasonal variations similar to pNH_4^+. HNO_3 dry deposition flux showed different seasonal patterns with highest autumn values, contributing 56% of estimated annual HNO_3 deposition, partially explained by higher autumn concentrations and deposition velocities.

Due to varying emission sources and meteorological conditions, individual Nr component contributions to total dry N deposition varied geographically. In this study, NH_3 contributed most to total dry N deposition (51%), significantly higher than NO_2 (18%), HNO_3 (10%), pNH_4^+ (15%), and pNO_3^- (6%). Gaseous Nr components accounted for 79% of total dry N deposition, highly consistent with North China Plain findings [**?**].

Similar to dry N deposition, wet/bulk deposition fluxes of NH_4^+-N, NO_3^--N, and total N deposition were higher in summer than other seasons, agreeing well with precipitation seasonal distribution. Summer precipitation accounted for 62% of the annual total, while winter precipitation was relatively small. Positive relationships between precipitation amounts and wet N deposition fluxes have been widely reported [**?**, **?**]. Summer deposition fluxes of NH_4^+-N and NO_3^--N (averaging 2.64 and 1.41 kg N/($hm^2 \cdot a$), respectively) contributed 49% and 44% to annual wet/bulk deposition. Both higher precipitation amounts and high atmospheric Nr component concentrations contributed to elevated summer N deposition fluxes. The NH_4^+-N to NO_3^--N concentration ratio is widely used as an indicator of atmospheric Nr sources [**?**]. Ratios >1 suggest agricultural sources, while ratios <1 indicate industrial sources [**?**]. In this study, summer and spring NH_4^+-N to NO_3^--N concentration ratios indicated that wet/bulk deposition N originated mainly from agricultural activities. In autumn and winter, agricultural and industrial contributions to wet/bulk N deposition were equal.

4.3 Comparison of Oxidized and Reduced N in Dry and Wet/Bulk Deposition

During the monitoring period, annual mean fluxes of oxidized (e.g., HNO_3 and NO_3^-) and reduced (e.g., NH_3 and NH_4^+) N in dry deposition were 2.12 and 4.09 kg N/($hm^2 \cdot a$), respectively. Dry N deposition in the current study was obviously lower than observations in northern China [**?**, **?**, **?**] but higher than rural Canadian sites [**?**]. Reduced N accounted for 66% of dry N deposition, consistent with previous Chinese studies [**?**]. NH_3 deposition accounted for 78% of dry reduced N deposition. The major atmospheric NH_3 sources are universally recognized as livestock wastes and agricultural N fertilizers [**?**]. According to Kuang et al. [**?**], NH_3 emissions from chemical fertilizer use and animal husbandry in China increased from 4.9 Tg N in 1980 to 8.0 Tg N in 2012. Agri-

cultural fertilizer application in Inner Mongolia increased by 13% from 2011 to 2015 (National Bureau of Statistics of China, 2011–2015). NO_2 was the largest oxidized N contributor to dry deposition, followed by HNO_3 and pNO_3^-. Mean deposition fluxes of NO_2, HNO_3, and pNO_3^- were 1.13, 0.63, and 0.36 kg N/($hm^2 \cdot a$), respectively, corresponding to 53%, 30%, and 17% of oxidized N in dry deposition. NH_3 and NO_2 were the two major dry N deposition contributors at the sampling site, together accounting for 69% of dry N deposition. NO_2 is a precursor to HNO_3. NH_3 can react with acid gases such as H_2SO_4 and HNO_3 in air, promoting PM2.5 formation. PM2.5 reduces atmospheric visibility, and long-term exposure can cause respiratory and cardiovascular diseases [**?**].

Due to strict implementation standards and technical improvements limiting atmospheric pollution, NO emissions in the USA dropped by 41% from 1990–2010 [**?**]. Butler et al. [**?**] quantified that reducing NO emissions by 50% would decrease total NO_3^- deposition by 37% and total N deposition by 25%, demonstrating the importance of controlling NO emissions, especially upwind of sensitive ecosystems. China has also implemented NO emission controls, proposing a 10% reduction in the 12th Five-Year Plan (2011–2015) and releasing stricter NO emission standards in late 2012 (Ministry of Environmental Protection of the People' s Republic of China). However, substantial work remains to control N deposition.

NH_4^+-N and NO_3^--N deposition fluxes in precipitation at the sampling site were lower than seriously N-polluted northern China regions [**?**], but higher than European estimates [**?**] and Urumqi, Xinjiang values [**?**]. Zhu et al. [**?**] found that wet N deposition, precipitation, N fertilizer application, and energy consumption are closely related, suggesting that wet N deposition could be reduced to acceptable levels by decreasing energy consumption and N fertilizer application. Reduced N dominated wet/bulk deposition with 63% contribution, similar to U.S. results [**?**]. Li et al. [**?**] noted that U.S. wet N deposition shifted from nitrate-dominated to ammonium-dominated due to decreased nitrate wet deposition fluxes (consistent with NO emission reductions) and increased ammonium wet deposition. In China, reduced N also dominates total N deposition, but oxidized N' s percentage is increasing rapidly [**?**], demonstrating the importance of continuous NO emission control measures.

Total inorganic N deposition (dry + wet/bulk) was 14.7 kg N/($hm^2 \cdot a$) in this study, approaching the upper limit of the N critical load (10–15 kg N/($hm^2 \cdot a$)) [**?**]. Approximately 58% of total N deposition came from wet/bulk deposition and 42% from dry deposition. Annual mean N deposition was slightly higher than Irish grassland estimates [**?**] but lower than most Chinese site studies [**?**, **?**]. Dry N deposition' s fractional contribution (42%) was lower than most Chinese studies [**?**, **?**, **?**, **?**]. Reduced N constituted 64% of total N deposition, 1.5 times higher than oxidized N' s contribution. Experimental studies show reduced N is more effective than oxidized N in driving biodiversity decline in acidic and mesotrophic grasslands [**?**]. The recognized N saturation critical load in terrestrial ecosystems is 10–15 kg N/($hm^2 \cdot a$), derived from field experiments [**?**].

Total N deposition in this study exceeded the lowest critical load value and may pose ecosystem threats. Field N addition experiments in Inner Mongolia temperate grassland indicated that increased N deposition decreased plant species richness [**?**] and altered plant community composition [**?**], threatening ecosystem stability. Additionally, substantial species richness loss is expected in low N deposition regions when deposition increases [**?**].

5 Conclusions

Average atmospheric and precipitation N deposition was estimated at 14.7 kg N/($hm^2 \cdot a$) from 2013–2015 in Duolun County' s semi-arid grassland, approaching the upper limit of the N critical load (10–15 kg N/($hm^2 \cdot a$)) for most grassland ecosystems. Specifically, 58% of total N deposition originated from wet/bulk deposition and 42% from dry deposition. Gaseous N deposition pathways dominated dry deposition (79% contribution). Among gaseous Nr components, NH_3 and NO_2 were most important, accounting for 69% of dry deposition. Affected by emission sources and meteorological conditions, most Nr components (except HNO_3) showed similar seasonal dry deposition patterns: maximum flux in summer and minimum in winter. The highest NH_4^+-N and NO_3^--N precipitation concentrations occurred in summer. A widely reported negative correlation between inorganic N concentrations and precipitation amounts was observed. Since summer N deposition fluxes contributed most to annual totals and high fluxes were partially attributable to agricultural and industrial activities, attention should focus on restraining these summer activities to reduce N deposition.

This work provides valuable background N deposition flux information for Inner Mongolia grassland and can serve as a reference for manipulative field experiments in this semi-arid region. Although total N deposition indicates Duolun County is not as heavily polluted as many other developed Chinese regions, the observed N deposition exceeded the lowest established critical value and could threaten grassland ecosystems.

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