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Original article

Atmospheric nitrogen deposition in the Loess area of China

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ABSTRACT

We present the quantification information on atmospheric nitrogen (N) deposition in the Loess area in Yangling District of Shaanxi Province, China using combined methods of a long-term field measurement (2006–2012), a potted experiment (2009–2011) and a long-term site-specific experiment (1991–2008). Our results showed that the annual deposition fluxes of dissolved inorganic N ($NH_4^+ - N$ and $NO_3^- - N$) via both precipitation (wet-deposition) and dustfall (dry-deposition) ranged from 12.73 to 37.87 kg N ha^{-1} yr^{-1} and averaged 21.76 kg N ha^{-1} yr^{-1}, with large contributions from wet deposition. The annual mean ratios of deposition amounts of ammonium N ($NH_4^+ - N$) to nitrate N ($NO_3^- - N$) were less than 1 in all the study years with an exception of the year 2007. Based on crop N uptake (approximately 52 kg N ha^{-1} yr^{-1}) in the pot experiment, it was estimated that the dry N deposition fluxes (including gas and particles) was about 23.81 kg N ha^{-1} yr^{-1}, accounting for 45.8% of the total N deposition. According the long-term site-specific experiment, the contribution of N input to farmland from the atmosphere was estimated to account for 52.8% of the environmental N input. Our results suggested that atmospheric N deposition was an important N input that must be taken into consideration when calculating nutrient budgets in agricultural ecosystems.

1. Introduction

The nitrogen (N) compounds are divided into nonreactive N and reactive N. The reactive nitrogen (N_r) refers to all biologically active, photochemically reactive, and radiativelly active N compounds in the atmosphere and biosphere of Earth. Thus, N_r includes inorganic reduced forms of N (e.g., NH_3 and NH_4^+), inorganic oxidized forms (e.g., NO_x, HNO_3, N_2O and NO_3^-), and organic compounds (e.g., urea, amines, and proteins), by contrast to unreactive N_2 gas (Galloway et al., 2008). The N compounds in the atmosphere mainly come from industries (NO_x), the combustion of fossil fuels (NO_x), fertilizer N applications and intensive livestock production (NH_3) (Vitousek et al., 1997). Atmospheric nitrogen (N) deposition refers to the process that air-borne nitrogenous compounds deposited to the earth's surface wet and/or dry deposition (Goulding, 1990). In general, wet N deposition is the result of precipitation events (rain, snow, etc.) that remove atmospheric Nr to the earth's surface, mainly including dissolved inorganic nitrogen (DIN, $NH_4^+ - N$ and $NO_3^- - N$) and a little of dissolved organic nitrogen (DON). In contrast, dry deposition is the transfer to the landscape of particles and gases through a number of atmospheric process in the absence of precipitation, including gaseous compounds (NO, N_2O, NH_3, HNO_3), particles ($(NH_4)_2SO_4$, NH_4NO_3) and organic N (Li and Tang, 2010).

Atmospheric N deposition is an important source and contributor to acidic deposition, and thus an important approach N input to biosphere (Larssen et al., 1999). Previous research has shown that anthropogenic N_r increased almost 12.5-fold from 15 Tg N yr^{-1} in 1860 to 187 Tg N yr^{-1} in 2005, which is about 1.9 times that of the global N critical load, and 70% reached the earth's surface due to

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wet and/or dry deposition (Galloway et al., 2003, 2004, 2008; Kaiser, 2001). The amount of N_r deposited to variety of terrestrial and aquatic ecosystems was estimated to be 105 Tg N yr^{-1} at a global scale (Galloway et al., 2008). Nitrogen is an essential nutrient for plant growth, and its demand is relatively large. Crops mainly receive N through fertilization, irrigation water, biological N fixation and atmospheric N deposition. Obviously, atmospheric N deposition is an important source of N for both terrestrial and aquatic ecosystems (Gao and Zhang, 2002; Li, 2002). However, excessive N deposition can pose adverse ecologic risks, resulting in soil acidification (Chen et al., 2008; Fang et al., 2011), eutrophication of surface water (Erisman et al., 1994; Boumans et al., 2004) and loss of biodiversity (Stevens et al., 2004; Song et al., 2005; Phoenix et al., 2006; Clark and Tilman, 2008).

Over the past two decades, China has witnessed serious N pollution and has been a global "hotspot" for N deposition due to rapid economic development (van Egmond et al., 2002; Dentener et al., 2006; Xie et al., 2010; Wang et al., 2011). Several studies have shown that the atmospheric N deposition in the North China are as high as 54–117 kg N ha^{-1} yr^{-1} (Shen et al., 2009; Liu et al., 2006; Zhang et al., 2006; He et al., 2010; Luo et al., 2013). The average value of wet N deposition in Shanghai from 1998 to 2003 was 58.1 kg N ha^{-1} yr^{-1} (Zhang, 2006), and the highest fluxes of wet and dry deposition in the Mid-South Region of China was 63.53 kg N ha^{-1} yr^{-1} (Lü and Tian, 2007). In general, the average annual bulk N deposition in China increased by approximately 8 kg N ha^{-1} between the 1980s (13.2 kg N ha^{-1}) and the 2000s (21.1 kg N ha^{-1}) (Liu et al., 2013).

Shaanxi is located in the heartland of the Loess Plateau, and it has 4.8 million ha of cultivated land. Nitrogen is applied as fertilizer at rates ranging from 480 to 520 kg N ha^{-1} yr^{-1} (Zhang et al., 2011) in order to achieve high crop yields. However, crops take up less than 20%–30% of the applied fertilizer, and a large amount (20–30%) is lost as NH_3 emissions (Cai et al., 2002; Zhang et al., 2008a; Ju et al., 2009). We suspected that the amount of N input to farmland from the environment has greatly increased via wet and dry deposition. However, very few studies have focused on that section in this region with extensive farmland area (the coverage percent is 46.4%). Therefore, a long-term monitoring experiment, a pot experiment without N fertilizer, and a long-term site-specific experiment were conducted in Yangling, Shaanxi Province. The objectives of this study were (1) to preliminarily quantify the amount of atmospheric N deposition, and (2) to provide essential information for nutrient balance calculations in agriculture.

2. Materials and methods

2.1. Study sites

A long-term monitoring experiment (2006–2012), pot experiment (2009–2011), and long-term specific-site experiment (1991–2008) were conducted in Wuquan Agricultural Experimental Base (34°17′ N, 108°0′ E, 520 m a.s.l.), Northwest Agricultural & Forestry University in Yangling, Shaanxi province (Fig. 1). The annual average temperature is about 12.9 °C and the annual mean precipitation is 632 mm, with most rain events are concentrated during the period from June to September. The monitoring site is about 10 km northwest of the downtown area, and may be polluted by emissions from motor vehicles, agricultural fields and burning of coal and straw for domestic heating in winter. The local cropping system is a winter wheat–summer maize rotation. Winter wheat is usually sown in October and summer maize is sown in June of the following year. N-fertilizers are applied to the cropping system in April (top dressing for winter wheat), August (top dressing for summer maize) and October (base fertilizer for winter wheat) of each year.

2.2. Sample collection and analytical methods

2.2.1. Monitoring experiment

An auto precipitation and dust sampler (APS-2A, Wuhan Tianhong Inc., China) controlled by sensors was installed at the study site. The system collected rainwater samplers only while the rainfall was occurring based on detection by rain sensors, whereas dustfall samples were also collected when there was no rain event. Rainwater was collected, recorded, thoroughly mixed and stored in clean 50 ml plastic bottles. Dustfall was collected in a bottle (300 mm in height and 150 mm in diameter) containing distilled water and 70 ml glycol (keep 5 cm liquid level) to ensure moisture in the bottle and avoid re-dusting or bacterial reproduction (Wei et al., 2010a). All samples was filtered using a syringe filter (0.45 mm, Tengda Inc., Tianjin, China), then frozen in a refrigerator at −17 °C until an analysis of NH_4^+ – N and NO_3^- – N by continuous flow analyzer (TRACCS 2000, Germany) within two months (Tartarti et al., 1995; Balestrin et al., 2000; Xie et al., 2010).

2.2.2. Pot experiment

There were two different solution application treatments in the pot experiment: (1) nutrient solution without N (as nutrient) and (2) no-nutrient solution (distilled water) (as no-nutrient). Each treatment was made with replication. Wheat (growth periods: October 30, 2009–June 15, 2010; October 15, 2010–June 12, 2011) and maize (growth periods: April 15, 2009–October 30, 2009; June 15, 2010–October 5, 2010; June 12, 2011–October 7, 2011) were sown in the treatment pots during the planting season. Each pot was filled with quartz, which was washed with diluted hydrochloric acid and distilled water. The crops were fertilized with an N-free nutrient solution and irrigated with distilled water. There was a plate under each pot to collect the leakage water, which could be reused for pot irrigation, if necessary. We assumed that the entire N plant uptake came from atmospheric dry and wet deposition. After being harvested, the plant samples were dried to constant weight at 80 °C before being ground up for analyzing the N concentration (microKjeldahl).

2.2.3. Long-term specific-site experiment

The long-term specific-site experiment included two treatments: (1) wheat–maize rotation with irrigation and without fertilization and (2) only wheat without irrigation or fertilization. The two treatments had three replications and were randomly distributed in six plots of 5×6 m^2. When the soil C and N contents reached a dynamic balance, the atmospheric N deposition could be estimated using Formula (1). Under the conditions of long-term depletion, uptake from sources such as soil N could be ignored. Therefore, the atmospheric N deposition was determined by calculating the difference between the N absorption by plants and the N input from irrigation water (Formula (2)).

$$\begin{aligned} \textit{Plant uptake N} = {} & \textit{Soil N} + \textit{Fertilizer N} + \textit{Deposition N} \\ & + \textit{Irrigation N} + \textit{Biological N fixation} - \textit{N loss} \end{aligned} \quad (1)$$

$$\textit{Deposition N} = \textit{Plant uptake N} - \textit{Irrigation N} \quad (2)$$

The measurements of the wheat and maize samples were the same as those described for the pot experiment. Irrigation water samples were collected from 34 different sites in Yangling to obtain the N content of the irrigation water. The concentrations of NH_4^+ –