Acta Ecologica Sinica 36 (2016) 424–432

Contents lists available at ScienceDirect

Acta Ecologica Sinica

journal homepage: www.elsevier.com/locate/chnaes

Quantification of nitrogen dry and wet deposition in Fujian tobacco planting area

Wen Xu [a], Wenqing Li [b,*], Shunhui Chen [b], Daowei Yang [a], Yangyang Zhang [a], Jing Xue [a], Xuejun Liu [a,*]

[a] China Agricultural University, College of Resources and Environmental Sciences, Beijing 100193, China
[b] Fujian Institute of Tobacco Agricultural Sciences, Fuzhou 350003, China

ARTICLE INFO

Article history:
Received 13 July 2015
Received in revised form 18 September 2015
Accepted 16 November 2015

Keywords:
Reactive N
Dry deposition
Wet deposition
Anthropogenic impact
Tobacco field

ABSTRACT

In this study, the wet/bulk and dry atmospheric deposition of different inorganic nitrogen (N) species was investigated in the Fujian tobacco planting area from 2010 to 2014. Wet/bulk N deposition flux of inorganic N was monitored at five sites (Fuzhou, Shanghang, Changting, Taining, and Wuyishan) of Fujian by using precipitation gauges. While dry deposition fluxes of gaseous NH_3, NO_2, and HNO_3 ($_gNH_3$, $_gNO_2$, and $_gHNO_3$), and particulate NH_4^+ and NO_3^- ($_pNH_4^+$ and $_pNO_3^-$) were estimated only at the Fuzhou site through multiplying monthly mean N_r concentrations obtained from DELTA (DEnuder for Long-Term Atmospheric sampling) system ($_gNH_3$, $_gHNO_3$, $_pNH_4^+$, $_pNO_3^-$) and passive samplers ($_gNO_2$) and the modeled monthly mean dry deposition velocities provided by the GEOS-Chem global chemical transport model. Annual mean NH_4^+-N, NO_3^--N, and total inorganic N (TIN) concentrations in precipitation (rainwater and snow) were 0.21–1.05, 0.33–0.74, and 0.59–1.68 mg N L^{-1}, respectively, and the averages over the monitoring period were 0.55, 0.49, and 1.04 mg N L^{-1}, respectively. Across the five monitoring sites, the average annual wet/bulk N deposition was 17.6 kg N ha^{-1} a^{-1}, while that of NH_4^+-N and NO_3^--N amounted to 9.2 and 8.4 kg N ha^{-1} a^{-1}, respectively. Wet/bulk deposition was lowest at Changting, but no significant differences were found between the other four sites. Annual mean atmospheric concentrations of $_gNH_3$, $_gNO_2$, $_gHNO_3$, $_pNH_4^+$, and $_pNO_3^-$ were 1.87, 2.95, 0.45, 2.36, and 1.23 μg N m^{-3}, respectively. Gaseous NH_3 concentrations were highest in summer and lowest in winter. In contrast, seasonal fluctuations in other measured N_r species (e.g., $_gNO_2$) were not as large as those of $_gNH_3$.

The annual mean total dry deposition was estimated to be 9.6 kg N ha^{-1} a^{-1}, and those of the individual species, $_gNH_3$, $_gNO_2$, $_gHNO_3$, $_pNH_4^+$, and $_pNO_3^-$ were estimated to be 4.1, 2.2, 2.0, 0.8, and 0.4 kg N ha^{-1} a^{-1}, respectively. Based on the similarities in wet deposition, if we assume a similar level of dry deposition at all the sites, the total (dry and wet) N deposition was 27.2 kg N ha^{-1} a^{-1} on average, which twice the critical load (10–15 kg N ha^{-1} a^{-1}) for forest ecosystems, suggesting a risk of "N saturation" in the local natural environment.

1. Introduction

Nitrogen is an essential element for all organisms in the biosphere, and commonly a limiting factor for the net primary productivity of aquatic and terrestrial ecosystems [1]. Triple-bonded nitrogen gas (N_2) makes up approximately 80% of the total mass of the Earth's atmosphere. This huge reservoir of N, however, exists as stable atmospheric N_2 and thus, is not biologically available to most organisms, unless it is converted into reactive N (N_r, which includes reduced inorganic N forms (e.g., NH_3, NH_4^+), oxidized inorganic N forms (e.g., NO, NO_2, HNO_3, NO_3^-), and organic compounds (e.g., urea, amines, proteins, nucleic acids). Over the past century, global emission of N_r has been increased dramatically due to accelerated anthropogenic activities, such as over-fertilization, intensive animal husbandry, and increasing combustion of fossil fuels). Most of this emitted N_r are deposited to the earth's surface by wet deposition and/or dry deposition [2]. Although N is the very stuff of life, excessive N deposition induces a considerable burden on forest, grassland, and aquatic ecosystems, resulting in the eutrophication of aquatic and terrestrial systems, reduction of biodiversity and soil acidification [3].

Between the 1980s and the 2000s, rapid economic growth in China has increased the consumptions of chemical fertilizer and fossil fuel by 2.0- and 3.2-fold, respectively [4]. As a consequence, the estimated total emission of NH_3 reached 9.8 Tg in 2006, which was responsible for nearly 15% and 35% of the global and Asian NH_3 emissions. Likewise, emission of NO_x (NO + NO_2) in China increased from 1.1 Tg in 1980 to 6.0 Tg in 2010 [5]. Such emissions have resulted in China becoming a global hotspot for N deposition.

Deposition measurement plays an important role in identifying trends in N emissions, understanding how to control eutrophication of surface water, making recommendations for the sustainable use of N fertilizer,

* Corresponding authors.
E-mail addresses: li-wqfjyc@163.com (W. Li), liu310@cau.edu.cn (X. Liu).

http://dx.doi.org/10.1016/j.chnaes.2016.09.007
1872-2032/

and developing deposition models [6]. In mid-eastern China, including the North China Plain and coastal developed areas, we should focus on the contribution of environmental N (particularly input from the atmosphere) to agroecosystems and water eutrophication, so as to make full use of this part of nutrient from the environment and reduce excessive input of nitrogenous fertilizer for the efficient utilization of nutrient resources [7].

Fujian Province, located in southeast China, is a high forested region. A previous study has reported that the flux of wet N deposition in Jiulong River watershed in Fujian province was about 10 kg N ha^{-1} a^{-1} [8]. In Fujian province, the tobacco fields are mainly distributed in Longyan, Sanming and Nanping cities. The preliminary results regarding wet/bulk N deposition in those areas have been reported by our past work for the year 2009 [9]. However, due to lack of long-term in situ observation it is difficult to obtain accurate information on the amount of wet N deposition as well as its annual variation. In addition, the data on the dry deposition fluxes of N_r species in this targeted area are very limited. Therefore, in this case study we set up five long-term in situ monitoring sites to make accurate estimates of atmospheric dry and wet N deposition in tobacco cropping system, an importation data set in support of N management and evaluation of N ecological risks in tobacco planting field as well as its surrounding areas.

2. Materials and methods

2.1. Site description

The five monitoring sites (Fuzhou, Shanghang, Changting, Taining and Wuyishan) are homogeneously distributed in four tobacco planting areas in Fujian province, China (Fig. 1). The selection of monitoring sites is based on comprehensive consideration of geographical distribution and land use types of sampling sites, as well as convenience of sampling. All the sites are located in vegetated, flat areas without obstacles. A detailed description of the monitoring sites has been previously reported [9]. In brief, this area is characterized by a typical oceanic monsoon climate. Annual average temperature is from 16.5 to 20.3 °C. The accumulated temperature (≥10 °C) is 6005–6703 °C, the frost-free period 250–336 days. Annual precipitation is around 1500–1900 m, of which 50–60% occurs during the period from March to June. Typical application rates of N fertilizer are 97.5–127.5 kg N ha^{-1} for tobacco planting. In this paper, the sampling sites for Fuzhou, Shanghang, Changting, Taining and Wuyishan are abbreviated as FZ, SH, CT, TN and WYS.

2.2. Sample collection and analytical methods

During 4 consecutive years (2010–2014), precipitation samples (wet/bulk deposition) at 5 monitoring sites were collected using stainless steel gauges (SDM6, Tianjin Weather Equipment Inc., China) installed at the height of approximately 1.5 m above the ground. Rainwater samples were collected, recorded, thoroughly mixed and stored in clean plastic bottles (50 mL) immediately after each rain event. In the analytical laboratory, each sample was filtered by a 0.45 μm syringe filter to remove insoluble particulates, and then stored at −18 °C until chemical analysis of NH_4^+-N and NO_3^--N by an AA3 continuous-flow analyzer (Bran + Luebbe GmbH, Norderstedt, Germany) within 1 month. Total inorganic nitrogen (TIN) was defined as the sum of NH_4^+-N and NO_3^--N.

For dry deposition sampling, ambient N_r concentrations of gaseous NH_3 ($_gNH_3$), HNO_3 ($_gHNO_3$), and particulate NH_4^+ and NO_3^- ($_pNH_4^+$ and $_pNO_3^-$) were measured monthly at FZ site using DELTA systems (DEnuder for Long-Term Atmospheric Sampling). For the other 4 sites, the measurements were not performed due to power limitations. The DELTA system, described in detailed by Xu et al. [10] comprised of a denuder filter sampling train, an air pump to provide low sampling flow rates of 0.2–0.4 L min^{-1} and a high sensitivity dry gas meter to record sampled volume. Briefly, the air passed through a denuder filter train, where $_gHNO_3$, $_gNH_3$, $_pNO_3^-$ and $_pNH_4^+$ were absorbed by coated chemical solutions in sequence. Two denuders coated with a solution of 1% (m/v) K_2CO_3 and 1% (m/v) glycerol in methanol were used for capturing $_gHNO_3$, whereas the two denuders for capturing $_gNH_3$ were coated with 5% (m/v) citric acid in methanol. For collection of aerosol components, a 2-stage filter pack at the end of the sampling train was adopted, with a first filter impregnated with 5% (m/v) K_2CO_3 + 10% (m/v) glycerol in methanol for absorbing $_pNO_3^-$ and a second filter impregnated with 13% (m/v) citric acid in methanol for absorbing $_pNH_4^+$. Gaseous NO_2 ($_gNO_2$) was sampled using Gradko diffusion tubes (Gradko International Limited, UK) which consists of an acrylic tube, two colored and white thermoplastic rubber caps in two ends of the tube and two stainless steel mesh disks. A 20% triethanolamine/deionized water solution was added onto two meshes within the colored cap for collection of $_gNO_2$.

All the samplers were deployed ~10 m above ground level on the rooftop of the building housing of the Institute of Tobacco Agricultural Sciences, Fuzhou city. There was no anthropogenic disturbance in the surrounding of the sampling site. The air intakes of the DELTA system and the NO_2 tubes were set at a height of 1.8 m above the roof, and these samplers were exposed to ambient air for 1 month. After sampling, the samples were returned to the laboratory and stored in a refrigerator at 4 °C and analyzed in less than a month. For each sampling and sample analysis, we made strict quality control measures using three laboratory and field (travel) blanks. The exposed HNO_3 denuders and alkaline-coated filters were extracted with 0.05% H_2O_2 in aqueous solution, whereas exposed NH_3 denuders and acid-coated filters were extracted with high purity water. The NH_4^+-N and NO_3-N in the extraction were determined with an AA3 continuous-flow analyzer as described before. The NO_2 diffusion tubes were extracted with a solution containing sulfanilamide, H_3PO_4, and N-1-naphthylethylenediamine, and the NO_2 content in the extract determined using a colorimetric method by absorption at a wavelength of 542 nm.

2.3. Deposition calculation

2.3.1. Wet/bulk deposition

Wet/bulk deposition flux was calculated using the following formulas (1) and (2)

$$C_w = \sum_{i=1}^{n}(C_i \times P_i)/\sum_{i=1}^{n} P_i \quad (1)$$

where C_w is the volume-weighted mean concentration (VWM, mg N L^{-1}) calculated from the n valid samples within a month or a year, and the individual sample concentration C_i is weighted by the precipitation amount Pi for each sample.

$$F_w = \sum_{i=1}^{n} C_i \times P_i/100 \quad (2)$$

where F_w is the wet/bulk deposition flux (kg N ha^{-1}), P_i is the total precipitation amount (mm), and 100 is a conversion factor.

2.3.2. Dry deposition

Dry deposition flux was calculated using the inferential method which combines measured concentrations and modeled dry deposition velocities. The formula is expressed as

$$F_d = C_n \times V_d \quad (3)$$

where F_d is the dry deposition flux (kg N ha^{-1}), C_n is the measured concentration (μg N m^{-3}), V_d is the deposition velocity (cm s^{-1}).

In this study, the hourly V_d of gaseous and particulate N_r for the whole of 2012 at FZ site was calculated by the GEOS (Goddard Earth Observing System)-Chem chemical transport model (CTM) (http://geos-chem.org). Then, the monthly V_d was averaged based on the hourly dataset for further estimation of dry deposition flux of each N_r species during the observation. This model is driven by GEOS-5 assimilated meteorological data from the NASA Global Modeling and Assimilation