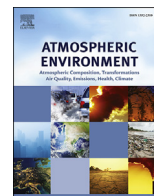




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Modeling dry deposition of reactive nitrogen in China with RAMS-CMAQ



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HIGHLIGHTS

- The air quality modeling system was applied to simulate dry deposition of Nr over China in 2010–2014.
- Major inorganic nitrogen components (10 species) and selected organic nitrogen components (5 species).
- Model results indicated a total dry N deposition budget of 9.31 Tg N yr⁻¹ in China.

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ABSTRACT

China has the world highest production of reactive nitrogen (Nr), and the Nr consumption increased sharply during the last decade. However, the potential environmental influence of dry nitrogen (N) deposition in China remains uncertain due to that the long-term measurement or remote sensing of various N species are difficult. This requires a better understanding of dry N deposition over China in its various forms and including magnitude and distribution features. Thus, the air quality modeling system RAMS-CMAQ was applied to simulate dry deposition of Nr over China from 2010 to 2014. The model results were then analyzed to investigate the long-term spatial and temporal distributions of major inorganic nitrogen (N) components (10 species) and selected organic N components (5 species). Comparisons between modeled and observed deposition rates and surface mass concentrations showed generally good agreement. Model results indicated a total dry N deposition budget of 9.31 Tg N yr⁻¹ in China, including 4.29 Tg N yr⁻¹ as NO_y and 4.43 Tg N yr⁻¹ as NH₃. NO_y was the main component of dry N deposition in the Beijing-Tianjin-Hebei area (0.31 Tg N yr⁻¹), the Yangtze River Delta (0.29 Tg N yr⁻¹), and the Pearl River Delta (0.09 Tg N yr⁻¹), where the major megacity clusters of China are located. NH₃ was the main component of dry N deposition in Shandong province (0.24 Tg N yr⁻¹), Northeast China (0.46 Tg N yr⁻¹), the Sichuan Basin (0.48 Tg N yr⁻¹), and central China (0.95 Tg N yr⁻¹), where the major agricultural regions are located. The highest values of the deposition flux for NH₃ occurred in Shandong province (19.40 kg N ha⁻¹ yr⁻¹) and Beijing-Tianjin-Hebei (17.20 kg N ha⁻¹ yr⁻¹). The seasonal variation of total dry N deposition was obvious in the east part of China, and was higher in July and lower in January. The spatio-temporal variations and major sources of dry N deposition were strongly heterogeneous, implying that the comprehensive pollution control strategies should be implemented to reduce the risk associated with N deposition.

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1. Introduction

Nitrogen (N) is an indispensable element for natural ecosystems. Unlike nitrogen gas (N₂), which is largely nonreactive, reactive nitrogen (Nr) is a crucial nutrient for living organisms that circulates naturally within and between the atmosphere,

pedosphere, hydrosphere, and biosphere. During the early twentieth century, the global cycle of N was significantly disturbed by human activities, particularly fertilizer use and energy production. Key developments that led to this disturbance included the invention of Haber-Bosch process (which allowed N_2 convert to NH_3) and widespread use of fossil fuels (which involves the conversion of atmospheric N_2 to NO_x under high temperature combustion). Although crop yields and economic development increased as a result of these developments, the concomitant increase in anthropogenic Nr has led to environmental consequences such as global climate change, the eutrophication of fresh water, soil acidification, and tropospheric ozone pollution.

The rate of anthropogenic Nr fixation has increased approximately 20-fold in the last century (Galloway et al., 2008), and emissions of global Nr reached approximately 109 Tg N yr^{-1} in 2010 (Fowler et al., 2013). Most of the emitted Nr is deposited back to ground and water bodies via dry and wet depositional processes. Dry deposition accounts for more than 60% of the total flux (Vet et al., 2014) and is the major contributor to N deposition. Previous studies have indicated that Europe, North America, India, and East Asia experience the highest levels of atmospheric N deposition in the world (Endo et al., 2010; Lelieveld and Dentener, 2000; Zhang et al., 2012a,b,c). Due to rapid population expansion and economic growth in recent decades, emissions of NO_x and NH_3 have increased significantly, thereby enhancing N deposition in China. Furthermore, as the nation with the largest agricultural production, China consumed about 35 Tg Nr yr^{-1} for agricultural and industrial uses in 2012, which corresponds to an increase of approximately 52% compared with that in 2000 (Ge et al., 2015). Thus, an enhanced understanding of the magnitude and spatial distribution of Nr deposition in China, as well as its corresponding influence on environment is desirable.

Several long-term monitoring networks have been established to capture the deposition of reactive nitrogen at a national scale. Examples of these monitoring networks include EANET (Acid Deposition Monitoring Network in East Asia; www.eanet.cc) and NNDMN (Nationwide Nitrogen Deposition Monitoring Network; Xu et al., 2015). The EANET surface monitoring network was built and operated in April 1998. This network consists of more than 40 observation stations in 12 East Asia countries that monitor aerosol mass, wet deposition, dry deposition, and the effects of these processes on soil and vegetation. NNDMN was established for obtaining comprehensive observations of dry and wet N deposition in China, including both gaseous and particle phases, and has been operational since 2010. This network includes 43 stations in different ecosystem types, including urban, rural, and background area located throughout China. Such ground-based networks provided valuable and highly reliable data for quantifying the intensity of N deposition in China, as well as the long-term trend and spatial distribution of this deposition.

Remote sensing from space is also an effective method for estimating the mass concentration of NO_x . Unlike ground-based in situ measurements, satellite monitoring can capture spatial heterogeneities and provides continuous temporal coverage of pollutants (Zhang et al., 2012a,b,c). Numerous studies have relied upon this approach to estimate the spatial distribution of NO_x in China (Jiang et al., 2016; Lee et al., 2014; Zhang et al., 2015). Furthermore, deposition velocities derived from ground observations (Cheng et al., 2013), empirical models (Jia et al., 2016), and chemical transport modeling systems (Pan et al., 2012) have been used to calculate flux of dry N deposition.

However, the magnitude and potential environmental influence of N deposition in China remain uncertain. Ground-based in situ observations are point measurements; thus, the regional features may not be revealed because of the spatial limitations of this

method. In addition, past work mainly focused on short periods and was limited to urban areas. Moreover, atmospheric N consists of more than one chemical species. Many of the previous studies significantly underestimated the deposition fluxes because they treated only gaseous NO_x , but not NH_3 , HNO_3 , or particulate components such as nitrate and ammonium (Luo et al., 2013; Nowlan et al., 2014; Shen et al., 2009). Therefore, the long-term N deposition flux, as well as the total load of deposited N, remain unknown for vast regions of China.

In this study, a regional air quality modeling system RAMS-CMAQ (Regional Atmospheric Modeling System-Models-3 Community Multiscale Air Quality) was used to investigate the long-term dry deposition of N over China (The distribution features about wet deposition of N over China has been discussed by applying the simulation results from this modeling system in Ge et al., 2011). This approach can simulate the long-term spatial and temporal distributions of almost all major inorganic N components and some organic N components. The geographic distributions of N mass concentration and dry deposition over China and the surrounding regions from 2010 to 2014 were simulated and analyzed. The modeling system is described in Section 2. Modeled dry N deposition is evaluated in Section 3. The simulation results are described and analyzed in Section 4. Section 5 presents our conclusions.

2. Methodology

RAMS provided the meteorological fields for the modeling system. This highly versatile numerical atmospheric model simulates and forecasts meteorological phenomena. The background meteorological fields and sea surface temperatures were obtained from the European Centre for Medium-Range Weather Forecasts reanalysis datasets ($1^\circ \times 1^\circ$ spatial resolution) and were based on weekly mean values and observed monthly snow cover information, respectively. A general description of RAMS and its capabilities has been provided by Cotton et al. (2003).

The mass burdens and deposition rates of pollutants were simulated using CMAQ (Version 4.7.1), which was developed by the US Environmental Protection Agency (US EPA) to model multiple air quality issues over a variety of scales. Gas-phase reactions were treated using the updated and expanded SAPRC mechanisms SAPRC-99 (Carter, 1999), which simulates in detail the reaction mechanisms for NO_x and volatile organic compounds (VOCs) over urban and regional scales. Compared with a closely similar mechanism (CB05) in CMAQ, SAPRC-99 is computationally expensive. However, this mechanism was developed specifically to describe the chemical conversions of atmospheric VOCs, contains approximately 400 types of VOC reactions and estimates approximately 550 VOC reactivity categories. This level of sophistication is needed for reasonable simulation of the behavior of NO_x and O_3 in the environment. The ISORROPIA (Nenes et al., 1999) model was applied to simulate thermodynamic equilibrium between inorganic aerosol species and gas-phase concentrations. The Regional Particulate Model (RPM) (Binkowski and Shankar, 1995) was used to describe aerosol dynamics in CMAQ.

The dry deposition velocities of sixteen gas phase chemical species were calculated by M3DDEP, Models-3/CMAQ dry deposition Module. The core method for gas phase pollutants can be expressed as:

$$V_d = (R_a + R_b + R_c)^{-1} \quad (1)$$

where R_a is the aerodynamic resistance, R_b is the quasi-laminar boundary layer resistance, and R_c is the canopy resistance. M3DDEP is coupled with the land surface model (Tremback and

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