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Ammonia volatilization as the major nitrogen loss pathway in dryland agro-ecosystems☆

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ABSTRACT

The losses of excessive reactive nitrogen (N) from agricultural production pose detrimental impacts on water, air and land. However, N budgets of agroecosystems are still poorly quantified, presenting a barrier to understand the N turnover in agriculture. Agricultural ammonia (NH_3) volatilization has been recognized as a crucial contribution to the pollution of fine particulate matters over China through reacting with acid gases. Building on these challenges, the first national-scale model analysis was constructed on the N budgets to gain an overall insight into the current status of N flows in Chinese dryland systems towards sustainable N management. Total inputs of soil N in Chinese dryland soils were estimated at 121 kg N ha^{-1} in 2010, considering all pathways including N manure, fertilizer, atmospheric deposition and litter from crop residues. Atmospheric N deposition accounted for 25% of N fertilizer plus N manure in Chinese dryland soils, suggesting that N deposition could not be ignored when estimating total N inputs to Chinese dryland soils. The highest ratio of NH_3 volatilization to total N outputs was found at 43 kg N ha^{-1} (~21%) in Northern China, followed by 41 kg N ha^{-1} (~20%) in Sichuan Basin and 25 kg N ha^{-1} (~26%) in Northeastern China. The modeling results indicated that, if a 20% decrease in N fertilizer plus N manure was achieved, it would lead to a 24% (7–49%) reduction in NH_3 volatilization. Substantial reductions of NH_3 volatilization would also be achieved by making an improvement in changing management practices (controlled release fertilizer and full irrigation). The results would give an overall insight into N budgets in Chinese dryland soils. The constructed N budgets assisted with understanding agricultural N flows and NH_3 pollution, and evaluated the impacts of human activities on N cycle towards a precise way to regulate agricultural management.

1. Introduction

Applications of nitrogen (N) fertilizer from the Haber-Bosch N fixation and manure amendment can increase supply of soil N and hence substantially increase food production (Galloway et al., 2008; Gu et al., 2015; Liu et al., 2013; Zhang et al., 2017a). However, the increased N fertilizer enhances the risks of N losses through ammonia (NH_3) volatilization, N leaching and runoff (Huang et al., 2016; Pan et al., 2016a; Xu et al., 2012). NH_3 is an important precursor for fine particulate matters with an aerodynamic diameter smaller than 2.5 μm ($PM_{2.5}$), having adverse impacts on air visibility and human health, especially in Northern China, a region with severe haze pollution (Chan and Yao, 2008; Geng et al., 2015; Van et al., 2016). However, there are still very few studies reporting large-scale N budgets including all pathways of N inputs and outputs in Chinese croplands, which are critically important for understanding the soil N cycle.

Atmospheric deposition, soil mineralization (decomposed from organic matters in passive humus, humads, microbial biomasses and plant residues) and chemical fertilizers were acknowledged as

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major N nutrient resources into the soil for croplands (Li et al., 1992a; Liu et al., 2010). Atmospheric N deposition was the process through which N compounds (including NH_3, NH_4^+, NO_x, HNO_3, and NO_3^- and organic N) are deposited to the surface through dry and wet deposition (Pan et al., 2012; Xu et al., 2018; Xu et al., 2015). To date, China's N deposition had exceeded North America and West Europe, which were the three hotspots of global N deposition. Based on the Nationwide Nitrogen Deposition Monitoring Network (NNDMN), the highest N deposition could have been up to 83 kg N ha^{-1} in croplands during 2010–2014 (Xu et al., 2015) and could have been up to approximately 1/3 of total N inputs (He et al., 2010; Liu et al., 2011; Liu et al., 2017d). Therefore, the contribution of N deposition to total N inputs into cropping systems could not be ignored when making a quantitative research on N budgets in Chinese dryland soils.

Nitrogen losses from the soil included the output fluxes from NH_3 volatilization, emissions of nitric oxide (NO), nitrous oxide (N_2O), dinitrogen (N_2), crop uptake, leaching and runoff (Deng et al., 2015; Li, 2000b). Among all the pathways of N losses from the soil, crop N uptake was a way that was beneficial to increase crop yields (Li et al., 2012; Waldrip et al., 2013). NH_3 volatilization dominated the other pathways of N losses, especially in dryland soils (Bouwman et al., 2002; Pan et al., 2016a). NH_3 volatilization was highly variable both in time and space, and the most of the previous studies regarding NH_3 volatilization in Chinese dryland soils were focused on site observations (Chen et al., 2009; Liu et al., 2018; Ma et al., 2012). The responses of NH_3 volatilization to N application rates from cropland were exponential or quadratic, rather than linear, proven by a number of previous estimates (Liu et al., 2018; Mudi et al., 2013; Zhang et al., 2011). Besides N fertilizer application forms and methods (such as weeding, manure amendment, fertilization, tillage, liming as well as irrigation), NH_3 volatilization was also impacted by soil parameters (such as pH, soil organic matter, bulk density and clay) (Chantigny et al., 2004; Zheng et al., 2018) and environmental factors (such as precipitation and temperature). Thus, it is very challenging to quantify NH_3 volatilization under cultivated conditions in spatially large and diverse areas, which are subject to great agricultural management drivers, environmental factors and soil parameters.

A few studies modeled NH_3 volatilization based on statistical models (Bouwman et al., 2002; Feng et al., 2015; Søgaard et al., 2002) and process based models such as Volt'Air (Génermont and Cellier, 1997), AGRIN (Beuning et al., 2008) and DeNitrification DeComposition (DNDC) (Balasubramanian et al., 2017; Liu et al., 2018; Waldrip et al., 2013) to explore the nonlinear relationship of NH_3 volatilization with soil parameters, environmental factors and agricultural management drivers. The DNDC model accounted for complex ecological and physico-chemical processes in the soil and soil-atmosphere interactions, which had been applied to simulate NH_3 volatilization based on site-specific inputs describing climate, nutrient management practices and crop growth (Balasubramanian et al., 2015; Balasubramanian et al., 2017; Li, 2000a; Waldrip et al., 2013). Previous studies lacked evaluation of the impact of atmospheric N deposition into N budgets and NH_3 volatilization under the background that high N deposition contributed to Chinese dryland soils (especially in Northern China), and also lacked the evaluation of the contribution of NH_3 volatilization to total N outputs. An overall insight into systematic summary of N budget should be investigated including all pathways of N inputs (synthetic fertilizer application, atmospheric deposition, manure amendment, weeds incorporation and crop residues) and outputs (NH_3 volatilization, emissions of NO, N_2O, and N_2, crop uptake, weeds uptake, leaching and runoff). Hence, the detailed N budgets and the variations regarding the effects of N deposition, soil properties and field management practices on NH_3 volatilization in Chinese dryland soils need further researches.

The DNDC model with the regional modeling mode was used to perform a model analysis on N budgets in Chinese dryland soils, summarizing all N input and output pathways, assessing biogeochemical effects of N fertilizer plus manure and atmospheric N deposition on NH_3 volatilization, as well as systematically testing the sensitivities of soil parameters, meteorological variables, management practices, as well as N deposition on N budgets and NH_3 volatilization. It aimed to gain an overall insight into the current status of N budgets in Chinese dryland systems, as an important step towards sustainable N management.

2. Data and methods

2.1. Chinese dryland soils

The cultivated land refers to the land on which crops are grown including the paddy and dryland soils. Dryland here refers to dryland farming relating to arid and semiarid regions emphasizing the efficient use of irrigation and precipitation. Here we focused on the dryland soils, and used the percent dryland soils dataset at a resolution of 1 km (Fig. 1) from the Institute of Geographical Sciences and Natural Resources Research, Chinese Academy of Sciences (http://www.resdc.cn/data.aspx?dataid=99, last access: 21 September 2018). The dataset included the time span of every five years of 1990, 1995, 2000, 2005, and 2010 which were based on the Landsat TM/ETM remote sensing images as the main data sources and generated by supervised classification algorithms combining manual visual interpretation. In this study, we resampled the percent dryland soils in 2010 to a resolution of 0.5° for our simulations. These datasets were considered as the most accurate remote sensing monitoring data of land use in China, which had played an important role in national land resources investigation, hydrology and ecological research. It had successively supported important applications such as the scientific and technological plans for the development of the Western China, the study on the causes of sandstorms in North China in the spring of 2000, the second national soil erosion survey, the construction of the national ecological environment monitoring network, the post-disaster assessment of the Wenchuan earthquake in 2008, the post-disaster restoration and reconstruction of ice and snow disasters in the south of China in the spring of 2009.

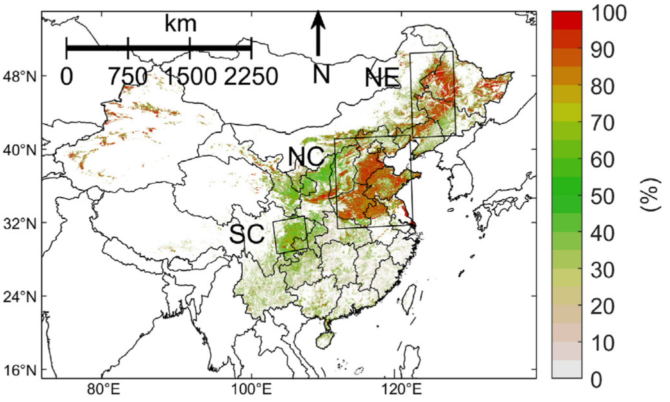

Fig. 1. Spatial distributions of percent dryland soils in China. The black rectangles represent Northern China (NC, 111–121°E and 31–41°N), Northeast China (NE,121–127°E and 41–50°N) and Sichuan Basin (SC, 103–107°E and 28–32°N).

2.2. Denitrification-decomposition (DNDC) model

The DNDC (Li et al., 1992a, b) was used in this study to model the soil N budgets and NH_3 volatilization in Chinese dryland soils. Its biogeochemical processes included NH_3 volatilization, denitrification, nitrification, decomposition, fermentation, allowing it to simulate the C and N transportation and transformations in agro-ecosystems (Babu et al., 2006; Butterbach-Bahl et al., 2001; Deng et al., 2011). DNDC had been extensively applied to modeling N budgets and evaluated against greenhouse gases (GHGs) and NH_3 volatilization fluxes (Congreves, 2016; Dutta et al., 2016; Li et al., 2012; Yang et al., 2012). DNDC was composed of two components. The first component (comprising of the decomposition sub-models, crop growth and soil climate) could be used to simulate substrate concentration profiles, soil redox potential (Eh), pH, moisture, temperature driven by ecological factors including anthropogenic management, soil, climate, and vegetation. The second component (comprising of the fermentation, denitrification and nitrification) could be used to simulate emissions of NH_3, N_2O, NO, N_2, CO_2 and CH_4 from the plant-soil systems.

Farming management practices, like fertilization, cultivation of cover crops, tillage, flooding and irrigation, had been parameterized in DNDC, to model soil environmental conditions, and hence could impact biogeochemical and biophysical reactions (Babu et al., 2006; Congreves et al., 2016; Deng et al., 2011). For example, N fertilization could influence soil N pools resulting from the types of fertilizers, methods of application and N application rates, and thereby could impact NH_3 volatilization, GHGs, crop growth as well as leaching and runoff. This enabled DNDC to model the impacts of fertilization methods on soil dynamics and NH_3 volatilization from denitrification and/or nitrification (Balasubramanian et al., 2017; Giltrap et al., 2017; Yan et al., 2003). More detailed information towards the DNDC algorithms, biogeochemical, chemical and physical processes could be found in the previous papers (Deng et al., 2015; Gilhespy et al., 2014; Li et al., 1992a; Li et al., 2012; Li, 2000a).

2.3. Fundamental theory of modeling soil N budgets and NH_3 volatilization

In DNDC, soil N basically existed in several forms: nitrate, NH_3, ammonium and organic N. Soil N dynamics in each pool were modeled at a daily time step through a number of biogeochemical processes including denitrification, nitrification, NH_3 volatilization, ammonium adsorption, leaching, decomposition, crop uptake and microbial assimilation. N cycled actively through soil, plants, air and water in agro-ecosystems. N obtained by the soil in DNDC included the input fluxes from biotic N fixation, synthetic fertilizer application, atmospheric deposition, mineralization N (from manure amendment, weeds incorporation and crop residue). N losses from the soil in DNDC included the output fluxes from NH_3 volatilization, emissions of nitric oxide (NO), nitrous oxide (N_2O) and dinitrogen (N_2), crop uptake, weeds uptake, leaching and runoff.

NH_3 volatilization to the atmosphere was linked with NH_3 concentration in the soil liquid phase as well as soil environmental factors including pH, moisture and temperature. Soil NH_3 concentration is controlled by a chemical reaction (Li, 2000b; Zhang et al., 2011):

$$[NH_4^+] + [OH^-] = [NH_{3\ (liquid)}] + H_2O (R1)$$

$$[NH_3(l)] = [NH_4^+]\,[OH^-] \,/\, K_a\ (R2)$$

where,

$$K_a = (1.416 + 0.01357 {}^{*} T) {}^{*} 10^{-5} \tag{1}$$

$$[OH^-] = K_w \,/[H^+] \tag{2}$$

$$[H^+] = 10^{-pH} \tag{3}$$

$$K_w = 1.945 \times e^{0.0645\times T} \times 10^{-15} \tag{4}$$

$[NH_4^+]$ is the ammonium concentration; [OH] was hydroxide ion concentration (mol/l); $[NH_3\ (l)]$ is the ammonia concentration in soil water; K_w is the water dissociation constant; pH is the soil pH.

Daily emitted fraction of the gas phase NH_3 was calculated by the following equation (Dubache et al., 2019; Li et al., 2019):

$$Flux(NH_3) = NH_3(l) \times ddf \tag{5}$$

where ddf is the volatilization coefficients, and Flux (NH_3) is the NH_3 fluxes.

In the original DNDC, the ddf was calculated by

$$ddf = f_{wind} \times f_{temp} \times f_{depth} \tag{6}$$

Dubache et al. (2019) proposed that the ddf was jointly affected by wind speed (f_{wind}), soil temperature (f_{temp}), soil depth (f_{depth}), vegetation canopy (f_{canopy}), clay content (f_{clay}), soil moisture (f_{water}) and rain induced canopy wetting (f_{rain}). We revised the DNDC95 code according to Dubache et al. (2019) by the following equations:

$$ddf = f_{wind} \times f_{temp} \times f_{depth} \times f_{canopy} \times f_{clay} \times f_{water} \times f_{rain} \tag{7}$$

$$\begin{cases} f_{wind} = 0.1 + 1.5S_{wind}/(1 + S_{wind}) \\ f_{canopy} = 0.4e^{-0.15lai} + 0.6 \\ f_{depth} = (q - j)\Big/q \\ f_{clay} = 0.4e^{-0.1clay} + 0.6 \\ f_{temp} = 0.1 + 2.0T_{soil}\Big/(45 + T_{soil}) \\ f_{water} = 0.45e^{-10WFPS} + 0.55 \\ f_{rain} = 0.65e^{-1.0\times lai\times P} + 0.35 \end{cases} \tag{8}$$

where, S_{wind} is the wind speed; lai is the leaf area index; q is the total soil layer number; clay is the soil clay content; T_{soil} is the soil temperature; WFPS is the air-filled porosity; P is the daily precipitation.

2.4. Database construction

The meteorological input data used to drive the DNDC included total precipitation, the maximum temperature and the minimum temperature at a daily time step. We used the daily precipitation from CPC Global Unified Precipitation data, and the maximum temperature and the minimum temperature reanalysis data from CPC Global Temperature data at a horizontal resolution of 0.5° latitude × 0.5° longitude provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, which could be downloaded freely on the Website (https://www.esrl.noaa.gov/psd/, last access: 21 September 2018). Spatial distribution of annual mean temperature and total precipitation in Chinese dryland soils in 2010 could be found in Fig. S1. The annual mean temperature and precipitation in 2010 in Chinese dryland soils were 12.13 °C and 865.50 mm, respectively.

We obtained the N fertilizer data produced from McGill

University in Canada (Potter et al., 2010), by spatializing the country-level N fertilizer use amount (including chemical fertilizer and manure). The maps of N input through fertilizer were developed by merging the harvested area from the M3-crops database (Monfreda et al., 2008) with the national-level fertilizer-use data for the same crops. The country-level N fertilizer use amount was gained from the Food and Agricultural Organization (FAO) and the International Fertilizer Industry Association (IFA). FAO total fertilizer consumption estimates were distributed using the spatial patterns of the cropland map (Ramankutty and Foley, 1999). The country-level crop N fertilizer use rates were calculated and allocated by harvested area of each crop group into a resolution of 0.5° × 0.5° for the year 2002. We followed a previous study (Zhao et al., 2017) using scaling factors to estimate the fertilizer data in 2010 from the 2002 values based on the statistical fertilizer data by provinces from the China Statistical Year Book (Gu et al., 2015). The total harvested area calculated from M3-crops was not always fully consistent with the harvested areas reported in IFA because of the various assumptions involved in developing M3-crops. In cases where no appropriate crop maps could be found, the M3-cropland map was used to distribute the fertilizer rates. These datasets could be downloaded freely on the Website: http://sedac.ciesin.columbia.edu/(last access: 21 September 2018). We used the N deposition datasets including both dry and wet processes, which had been described in detail in previous studies (Liu et al., 2017a; Liu et al., 2017b; Liu et al., 2017c; Zhang et al., 2017b). Total N deposition could be calculated by sum of wet N deposition, dry gaseous and particulate N deposition. Dry N deposition was calculated by ground N concentrations and dry deposition velocity using the inferential method (Adon et al., 2013; Cheng et al., 2013; Flechard et al., 2011). The ground N concentrations were derived using the satellite columns combining the vertical profiles (Liu et al., 2017b; Liu et al., 2017c; Zhang et al., 2017b). The mixed effects models (Liu et al., 2017a; Zhang et al., 2018) were used to estimate wet N deposition based on NO_2 columns and NH_3 columns and meteorological factors. The correlation of estimated N deposition with the national measurements from the Chinese Nationwide Nitrogen Deposition Monitoring Network (NNDMN) was 0.89 for all sites at a yearly scale.

The basic inputs of soil parameters used for driving DNDC included soil physical and chemical attributes: pH values, soil organic matter fractions (SOMs), clay fractions and bulk densities. We used the datasets at a resolution of 0.0083° from Soil Database of China for Land Surface modeling, which could be freely downloaded at the Website: http://globalchange.bnu.edu.cn/research/soil2d.jsp (last access: 21 September 2018). The spatial maps of gridded soil properties were consistent with common knowledge of Chinese soil scientists, which could be incorporated into land models to better study the role of soils in biogeochemical and hydrological cycles in China. More details on this comprehensive datasets could be found in a previous paper (Shangguan et al., 2013).

2.5. *Model application*

We used the DNDC95 model as one of the latest DNDC versions, which could be freely downloaded at the Website: http://www.dndc.sr.unh.edu/(last access: 21 September 2018). The basic inputs to drive DNDC included (1) meteorological data of daily precipitation, the maximum and the minimum temperature; (2) soil properties of pH, SOC, bulk density and content clay fraction; (3) crop parameters of yield potential, thermal degree days (TDD) for maturity and the water demand; (4) management practice parameters of harvest, tillage, irrigation, fertilization and flooding. For the inputs of meteorological data and soil properties, we have described them in the section of database construction. For the crop parameters in Chinese dryland soils, we used the EarthStat Dataset of crop categories and crop harvest area at 0.083333333° grids. Here we resampled them to a resolution of 0.5°. For the management practice parameters, the planting dates, fertilizer dates and harvested dates were gained from previous estimates by assuming average parameters for each region (Sacks et al., 2010; Zhang et al., 2017a; Zhang and Zhang, 2011).

We used DNDC to model NH_3 volatilization in 2010 based on the inputs of meteorological data of daily precipitation, the maximum, the minimum temperature, soil properties of pH, SOC, bulk density, content clay fraction, crop parameters of yield potential, thermal degree days (TDD) for maturity, the water demand, management practice parameters of harvest, tillage, irrigation, fertilization and flooding. We recognized that the uncertainties may come from the various inputs of DNDC for our simulations of N budgets and NH_3 volatilization, and thereby conducted one base simulation and several scenarios with regard to meteorological data, soil parameters, crop parameters and management practices. For the base simulation, we used the DNDC to simulate the daily growth of corn, wheat and vegetable (cabbage, beans and melons) in 2010 in dryland soils in China. Corn and wheat constituted higher 90% of cereal crop production and total staple food production (not considering rice which was widely distributed in paddy soils rather than dryland soils). We used the 2010 conditions for irrigation, soil, crop parameters, cropping systems (rotations) and fertilization. The crop rotation systems involving corn and wheat in China were as follows: corn, spring wheat, winter wheat, corn-winter wheat, vegetable-winter wheat and vegetable-corn. The distribution of these crop rotation systems in the DNDC was prescribed according to geographical information and county-level archives (Yu et al., 2019). Management practices in dryland soils included fertilization, planting, tillage and harvest. We set the fertilizer, planting, and harvest time according to a previous work (Zhang et al., 2017a), while the conventional tillage was used for the tillage method. In our base simulation, all the inputs were the actual values that were described above including meteorological data, soil parameters, N deposition, N fertilizer (urea) and manure without nitrification inhibitor conventional tillage and full irrigation. For simulations of different scenarios, only one parameter was set varied, such as the meteorological factors (by ±20% in temperature and precipitation) and soil factors (by ±1 in pH and ±20% in clay, SOC and bulk density); set N deposition as zero to test the effects of N deposition; changed the fertilizer types one by one including urea plus nitrification inhibitor, no irrigation, controlled release fertilization, a ±20% change in N fertilizer plus N manure.

3. Results

Total soil N inputs included synthetic N fertilizer application, atmospheric N deposition, manure amendment and crop residues (Deng et al., 2015; Li, 2000a). Fig. 3 shows the spatial distributions of N fertilizer, N deposition and N manure amendment in Chinese dryland soils in 2010. Considering all pathways (including N fertilization, N manure, N deposition and crop residues) (Fig. 2), total N inputs in Chinese dryland soils in 2010 were estimated at 121 kg N ha^{-1}. Atmospheric N deposition (21 kg N ha^{-1}) accounted for approximately 25% of N fertilizer plus N manure (84 kg N ha^{-1}) in Chinese dryland soils, and was highly linked with the N fertilizer plus N manure ($R = 0.77$ and $p = 0.00$). Also, weeds incorporation and crop residues (roots and stub) contributed 13% of total N inputs, in addition to chemical N fertilizer, N manure and N deposition. The mineralization N from crop residues was associated with soil organic matter decomposition consumed by the soil microbes (Giltrap et al., 2010; Li et al., 1992b). The decomposed organic N can

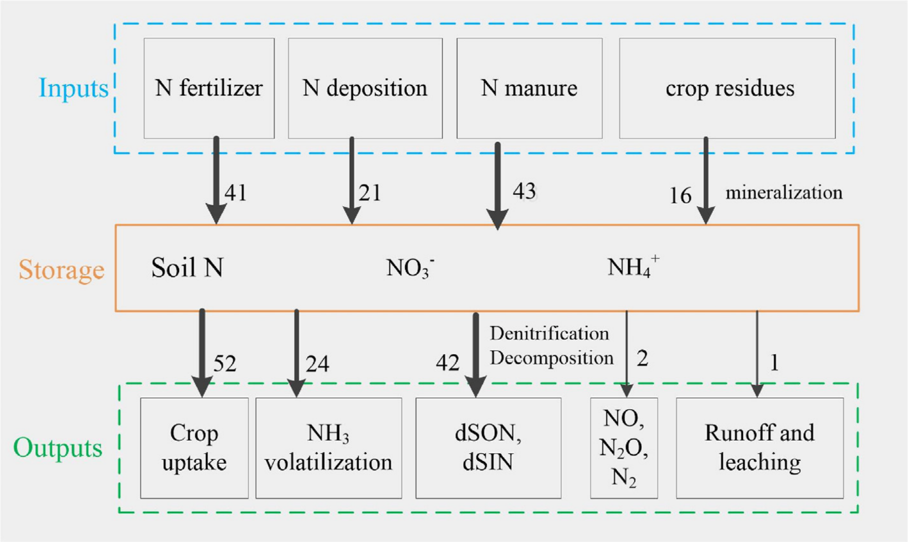

Fig. 2. Soil N inputs and outputs in dryland soils in China. The soil N inputs include N fertilizer, deposition, manure and crop residues, while the soil N outputs include crop uptake, NH_3 volatilization, dissolved soil organic and inorganic N (dSON and dSIN), gaseous NO, N_2O and N_2, runoff and leaching.

also partially be mineralized to ammonium (NH_4^+) (Deng et al., 2015; Li, 2000b). The mineralization N was highly linked with the activity of the soil microbes, which were impacted highly by soil temperature (Congreves et al., 2016; Li et al., 2017a). The correlation of mineralization N and temperature was 0.98 (Fig. S2).

In terms of different regions, the highest N fertilizer, N manure and N deposition occurred in Northern China (NC), followed by Sichuan Basin (SC) and Northeast China (NE) (Fig. 1). Total N inputs in NC were 208 kg N ha^{-1}, in which N fertilizer plus N manure together accounted for 75%, N deposition and crop residues accounted for 15% and 10%, respectively (Fig. 3). The estimate of total N inputs in NC in this study (208 kg N ha^{-1}) was close to a previous estimate (221 kg N ha^{-1}) (Liu et al., 2020), nearly double than that in Western Europe (113 kg N ha^{-1}) and Eastern United States (91 kg N ha^{-1}). The SC also had high N inputs (198 kg N ha^{-1}), which were dominated by N fertilizer and N manure (78%), followed by N deposition (12%) and crop residues (10%). The NE had relatively less N inputs (98 kg N ha^{-1}), in which N fertilizer plus N manure, N deposition and crop residues accounted for 67%, 18% and 15%, respectively.

The possible soil N outputs included crop uptake, NH_3 volatilization, denitrification, nitrification, runoff and leaching (Congreves et al., 2016; Deng et al., 2011). Crop N uptakes (43%), dissolved inorganic or organic N (dSIN, dSON) and remains (35%), NH_3 volatilization (20%) were identified as the major pathways of soil N outputs, while the other N losses (1%) were through N leaching, N runoff, and gases (NO, N_2O, N_2) pathways. Spatial patterns of NH_3 volatilization in dryland soils were shown in Fig. 4. The highest NH_3 volatilization was found at 43 kg N ha^{-1} (~21% to total soil N outputs on average) in NC, followed at 41 kg N ha^{-1} (~20% on average) in SC, and 25 kg N ha^{-1} (~26% on average) in NE.

The estimates of NH_3 volatilization in this study was compared with the REAS (Regional Emission inventory in Asia) NH_3 volatilization, MIX NH_3 volatilization (Li et al., 2017b) and NH_3 volatilization by Zhang's estimates (Zhang et al., 2017a). In general, our estimates of NH_3 volatilization were similar to the REAS NH_3 volatilization ($R = 0.80$, 23.67 vs. 23.67 kg N ha^{-1}), MIX NH_3 volatilization ($R = 0.77$, 23.67 vs. 25.56 kg N ha^{-1}), and were lower than Zhang's estimates ($R = 0.83$, 23.67 vs. 29.96 kg N ha^{-1}) (Fig. 5). Our estimates of NH_3 volatilization were compared with the satellite NH_3 retrievals from IASI, which could reveal the aggregated impacts of the NH_3 emissions from N fertilizer and manure management in arable soils (Van Damme et al., 2014a; Van Damme et al., 2014b). NH_3 volatilization estimates in this study also showed a high correlation with the IASI NH_3 columns ($R = 0.74$ and $p = 0.00$ in Fig. S3), reflecting the same spatial gradients of NH_3 abundance in Chinese dryland soils.

NH_3 volatilization could happen when all ammonium and ammonia-based fertilizers were applied in dryland soils in China (Balasubramanian et al., 2017; He et al., 2010). The modeling results could be applied to comprehensively evaluate the impacts of driven factors and management alternatives on potential NH_3 volatilization mitigation for agroecosystems. The base simulation of NH_3 volatilization in dryland soils considered all the inputs including meteorological data, soil parameters, N deposition, N fertilizer (urea) and N manure without nitrification inhibitor. To explore the potential sensitivities of each parameter for estimating NH_3 volatilization, several scenarios were tested by setting only one parameter changing and the others unchanged (Fig. 6).

A 20% decrease in N fertilizer and N manure could cause a 24% decrease in NH_3 volatilization, and the estimates of NH_3 volatilization could be approximately underestimated by 4% in case the N deposition was ignored in the simulations. Subsequently, NH_3 volatilization increased with a higher temperature but decreased with more precipitation. High temperatures caused higher rates of volatilization because warm soil water could not hold as much NH_3 gas, while more precipitation in a dry cropland could leach urea deep into the soil by protecting it from volatilization (Huang et al., 2016; Levine et al., 1980). A 20% increase in precipitation and one degree increase in temperature could cause a ±2% increase in NH_3 volatilization. Soil pH had a positive impact on NH_3 volatilization, and a 20% increase in pH could lead to a 13% increase in NH_3 volatilization. A high soil pH could lead to high NH_3 volatilization mainly due to the fact that a high pH could increase soil NH_3 dissolved in water (increases the rate of ammonium conversion to dissolved NH_3) (Bussink and Oenema, 1998; Génermont and Cellier, 1997). A 20% increase in bulk density, SOC and clay would make NH_3 volatilization vary within ±5%. Different fertilizer types also had important impacts on NH_3 volatilization. In the base simulations, the urea (without nitrification inhibitor) was the fertilizer type used in dryland soils in China. The common fertilizer types

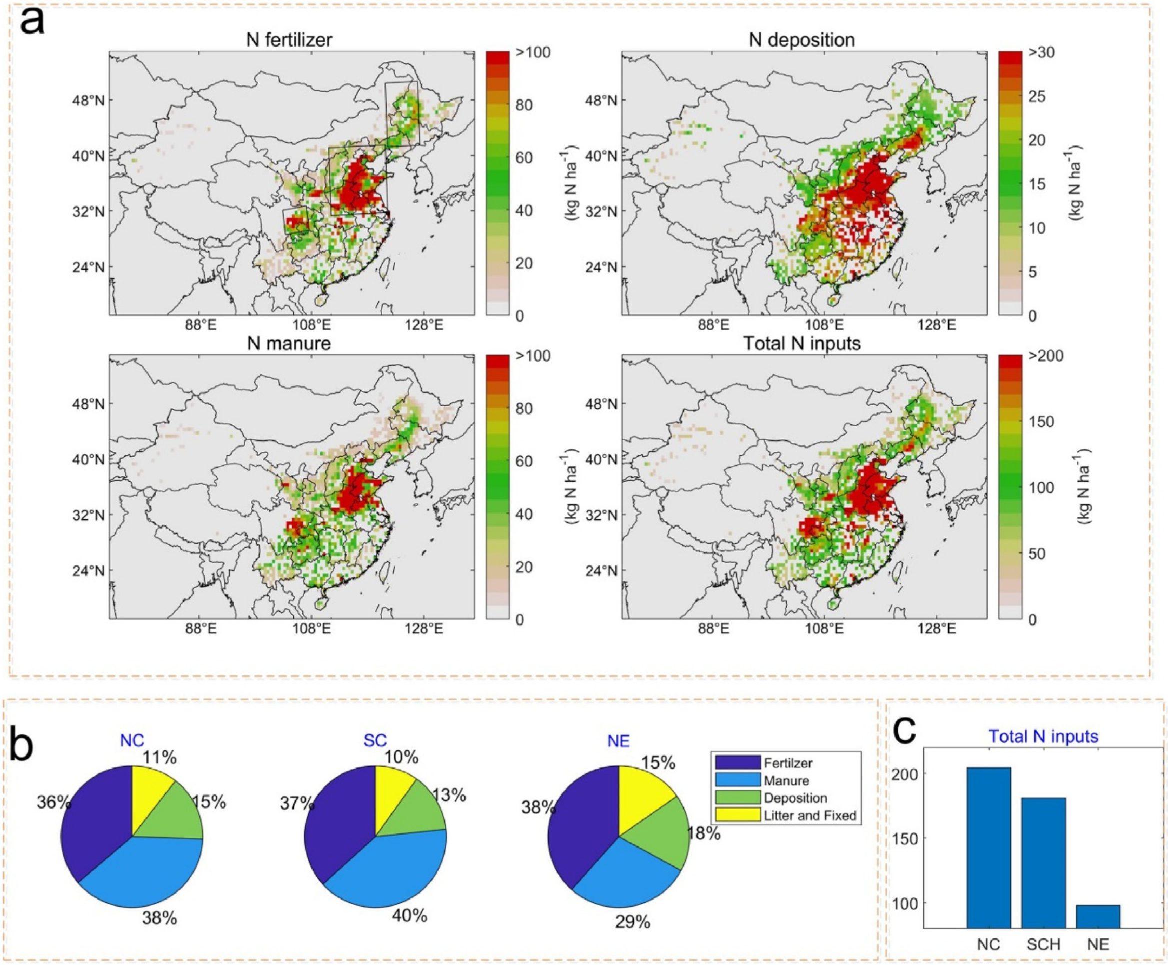

Fig. 3. Spatial patterns of soil N inputs in Chinese dryland soils in 2010. (a) Chemical N fertilizer, atmospheric N deposition, N from manure amendment and total N inputs; (b) contributions of each factor to total N inputs; (c) summary of total N inputs in NC (111–121°E and 31–41°N), SC (103–107°E and 28–32°N) and NE (121–127°E and 41–50°N).

were tested using urea plus a nitrification inhibitor (Fan et al., 2018; Sun et al., 2016). Nitrification inhibitors could delay or slow down the nitrification process (ammonium is oxidized to nitrate by bacteria), thereby avoiding large losses of nitrate before the fertilizer N was taken up by plants (Bouwman et al., 2002; Congreves, 2016). However, this process unexpectedly could increase NH_3 volatilization by 10% since nitrification inhibitors could slow down the conversion of NH_4^+ to NO_3^- and maintain higher soil NH_4^+ potentially for volatilization. The controlled release fertilizer (such as using polymer sulphur-coated urea) was identified as the most effective way, which could cause a 48% decrease in NH_3 volatilization (Fig. 6b) by delaying urea hydrolysis (Pan et al., 2016a). In addition, compared with no irrigation, full irrigation would make a 21% reduction in NH_3 volatilization.

4. Discussions

Reactive N compounds emitted to the atmosphere had a great influence on the environment (Levine et al., 1980; Søgaard et al., 2002), but estimating N budgets and NH_3 volatilization from croplands on a large spatial scale still had large uncertainties (Bouwman et al., 2002; Feng et al., 2015). It was mainly due to the nonlinear relationships between N budgets and relevant driven factors (such as soil parameters, meteorological data as well as atmospheric N deposition) (Bouwman et al., 2002; Feng et al., 2015). To our knowledge, this was the first study to investigate the soil N budgets in dryland soils in China, and to perform a model analysis on soil N budgets, summarizing all kinds of N input and output pathways.

Atmospheric N deposition was an important pathway of reactive N inputs into Chinese croplands (Liu et al., 2013). However, the current monitoring networks in general do not measure N deposition including both dry and wet pathways except the Nationwide Nitrogen Deposition Monitoring Network (NNDMN) (including 43 sites), and the most of previous works examining N deposition in croplands often had very few sites, which leave much unknown information on the magnitude, variability, and the biogeochemical effects of N deposition (Liu et al., 2010; Pan et al., 2012; Xu et al., 2015). Our previous works (Liu et al., 2017a; Liu et al., 2017b; Liu et al., 2017c; Zhang et al., 2018; Zhang et al., 2017b) used the satellite-based approaches to estimate dry and wet N deposition in China, constrained with the measurements from the NNDMN. This work addressed this knowledge gap of atmospheric N deposition on N budgets, and presented that N deposition in farmland areas was greatly variable, elevated, linked with N manure and fertilization in agricultural ecosystems. Atmospheric N deposition in Chinese dryland soils was 21 kg N ha^{-1} on average based on our estimates, which accounted for approximately 25% of N fertilizer plus N manure, suggesting that the N deposition could not be ignored in estimating total N inputs to croplands. The ratio of N

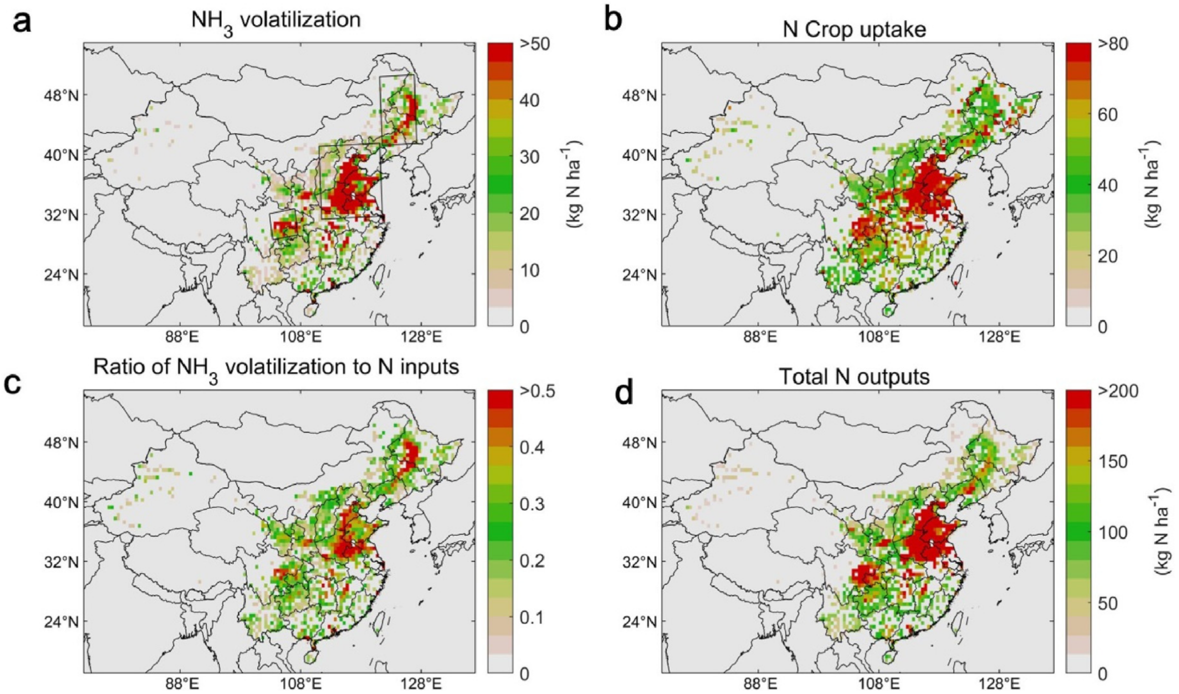

Fig. 4. Spatial patterns of soil N outputs in dryland soils. (a) NH_3 volatilization, (b) N uptake by crops, (c) ratio of NH_3 volatilization to total N inputs, (d) total N outputs.

deposition to N fertilizer in Chinese dryland soils was 51%. In fact, the importance of N deposition in cropping systems had been highlighted by a previous study (Liu et al., 2010), in which their estimates of the ratio of N deposition to N fertilizer was approximately 50% in China during 2001–2007 (Liu et al., 2010), which was close to our estimate. Notably, Liu et al. (2010) presented a rough estimate on the estimate of N deposition to N fertilizer based on national statistical data, and they did not consider all the total N inputs and outputs all over China. To our knowledge, the first national-scale model analysis was constructed on the N budgets investigating the current status of N flows in Chinese dryland systems assisting with understanding N budgets and NH_3 pollution. A high N deposition in Chinese croplands could be also tracked by a number of previous studies based on N deposition measurements (Liang et al., 2016; Liu et al., 2006; Liu et al., 2013; Pan et al., 2012; Shen et al., 2009). Due to the high percentage of N deposition to total N inputs in Chinese dryland soils systems, the importance of including atmospheric N deposition for our simulations was recognized in order to better model the biogeochemical effects of soil N budgets and NH_3 volatilization in agricultural systems. The finding of this study added to increasing works of understanding N deposition and its biogeochemical effects on soil N budgets and NH_3 volatilization in agricultural systems.

On the other hand, crop N uptakes, soil N remains, NH_3 volatilization (20%) were the major pathways of soil N outputs, while the other N losses (1%) was through N leaching, N runoff and gaseous NO, N_2 and N_2O in Chinese dryland soils. High NH_3 volatilization were found in NC (43 kg N ha^{-1} on average), followed by SC (40 kg N ha^{-1}) and NE (25 kg N ha^{-1}), contributing to the reduction of fertilizer use efficiency (Clarisse et al., 2009; Kang et al., 2016). Notably, the percent dryland soils were the highest in NC (61%), followed by NE (41%) and SC (32%), which was not consistent with the ranking of absolute values of NH_3 volatilization. This inconsistency was mainly because the percent dryland soils represented the proportional dryland in per hectare, while the NH_3 volatilization was highly linked with the amounts of N fertilizer related with cropping systems, meteorological and soil parameters. The core factor affecting the NH_3 volatilization was the N fertilizer (chemical N and manure N), for which the N application rate in SC (155 kg N ha^{-1}) was much higher than that in NE (65 kg N ha^{-1}). A meta-analysis regarding global NH_3 volatilization showed that the average percentage of NH_3 volatilization to total N inputs was 18% (Pan et al., 2016b), which was close to this value (20%) in Chinese dryland soils with low N use efficiency, high N application rate as well as smallholder farms (mainly depending on the family labors) in China (Ju et al., 2016). In particular, Northern China was experiencing serious and persistent particulate matter pollution in recent years (Wei et al., 2020; Wu et al., 2016). NH_3 was an important precursor for $PM_{2.5}$, and high NH_3 volatilization contributes to $PM_{2.5}$ pollution (Wei et al., 2019a; Wei et al., 2019b), particularly under the background that SO_2 and NO_x emissions had decreased in recent decade. Thus, it was critically important to increase fertilizer use efficiency in Northern China to control the NH_3 volatilization, such as increasing farm size and improving technological innovation in the future (Gu et al., 2012; Ju et al., 2016; Wu et al., 2018). The analysis of N budgets and NH_3 volatilization in this study was based on the national-scale model analysis by DNDC. In the future, monitoring measurements should be conducted to verify the modeling results covering six sub regions including northeastern, northern, western, southwestern and southeastern China.

Modeling impacts of the driving factors and management alternatives on N budgets, NH_3 volatilization as well as greenhouse emissions (Goglio et al., 2014; Herrero et al., 2013) were crucially important towards environmental safety. Through the modeling estimates in N budgets and NH_3 volatilization in this work, potential opportunities were demonstrated to mitigate NH_3 volatilization emissions in dryland agriculture. A 20% decrease in N fertilizer and N manure could cause a 24% decrease in NH_3 volatilization. The controlled release fertilizer (for example polymer sulphur-coated urea) could cause a 48% decrease in NH_3 volatilization (Fig. 6b) by delaying the N availability for crop uptake and urea hydrolysis (Pan et al., 2016a). However, nitrification inhibitors

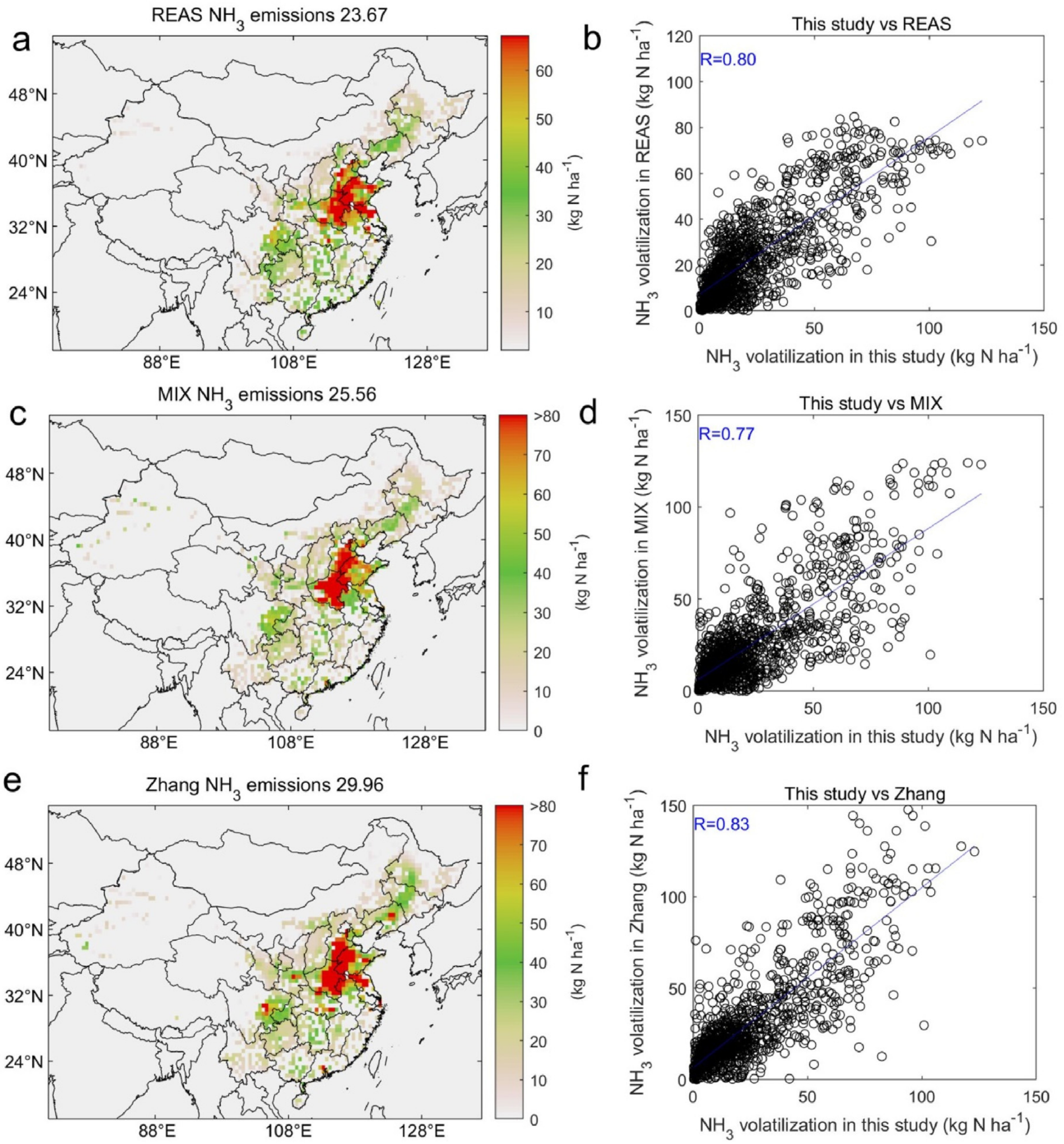

Fig. 5. Comparison of NH_3 volatilization estimates in this study with NH_3 volatilization from Regional Emission inventory in ASia (REAS) (a and b), MIX (http://www.meicmodel.org/dataset-mix.html) (c and d), and Zhang's estimates (http://www.phy.pku.edu.cn/~acaq/data/nh3_agr_emis.html) (e and f) (Zhang et al., 2017a). Panel a, b and c represent the spatial maps of REAS, MIX and Zhang's estimate, while c, d and e indicates the comparison between the estimate in this study with theirs.

(designed for decreasing N_2O emissions) unexpectedly increased NH_3 volatilization by 10% through prolonging the time of NH_4^+ in soil and stimulating the conversion of NH_4^+ to NH_3 (Pan et al., 2016a). In other words, nitrification inhibitors could reduce the speed of the conversion of NH_4^+ to NO_3^-, leading to higher soil NH_4^+ for volatilization. Regarding the spatial patterns (Fig. 7), the controlled release fertilizer could decease NH_3 volatilization by 45%, 63 and 49% in NC, SC and NE, respectively; the reduced N fertilizer by 20% could decrease NH_3 volatilization by 33%, 17 and 15% in NC, SC and NE, respectively; compared with no irrigation, full irrigation would make a 33%, 15% and 17% reduction in NH_3 volatilization in NC, SC and NE, respectively. Moreover, substantial decrease of NH_3 volatilization (~13%) would be achieved if a 20% decrease was set in pH. This indicated that applying management alternatives at dryland soils might be effective towards mitigating NH_3 volatilization, and suggested that the future regulated policies required taking account of the soil conditions with a more precise way to regulate agricultural management.

5. Conclusion

Considering all pathways including N fertilization, N deposition and mineralization N (from N manure amendment, weeds incorporation and crop residues), total N inputs in Chinese dryland soils in 2010 were estimated at 121 kg N ha^{-1}. Atmospheric N deposition accounted for 25% of chemical N fertilizer plus N manure in Chinese dryland soils. Crop N uptakes (43%), soil N remains (35%), NH_3 volatilization (20%) were identified as the major pathways of soil N

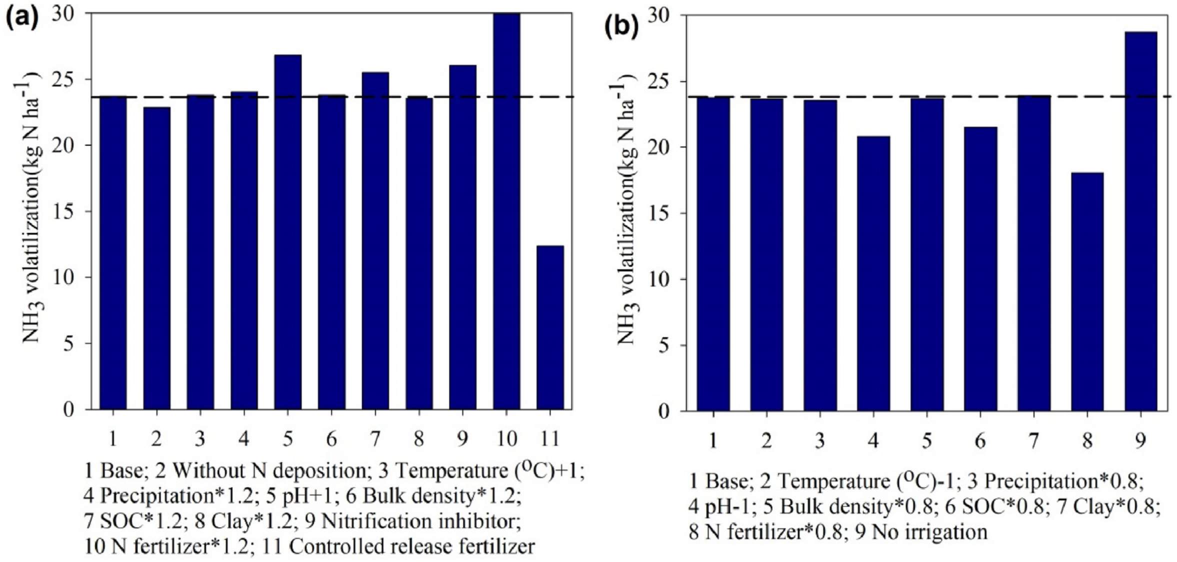

Fig. 6. Sensitivities of different variables for estimating NH_3 volatilization. The base simulation used the actual meteorological data, N deposition, soil parameters with urea and no nitrification inhibitor. Panel a and b indicate the estimation of NH_3 volatilization using the enhanced and reduced driven data including pH, bulk densities, SOC, clay and N fertilizer by the DNDC.

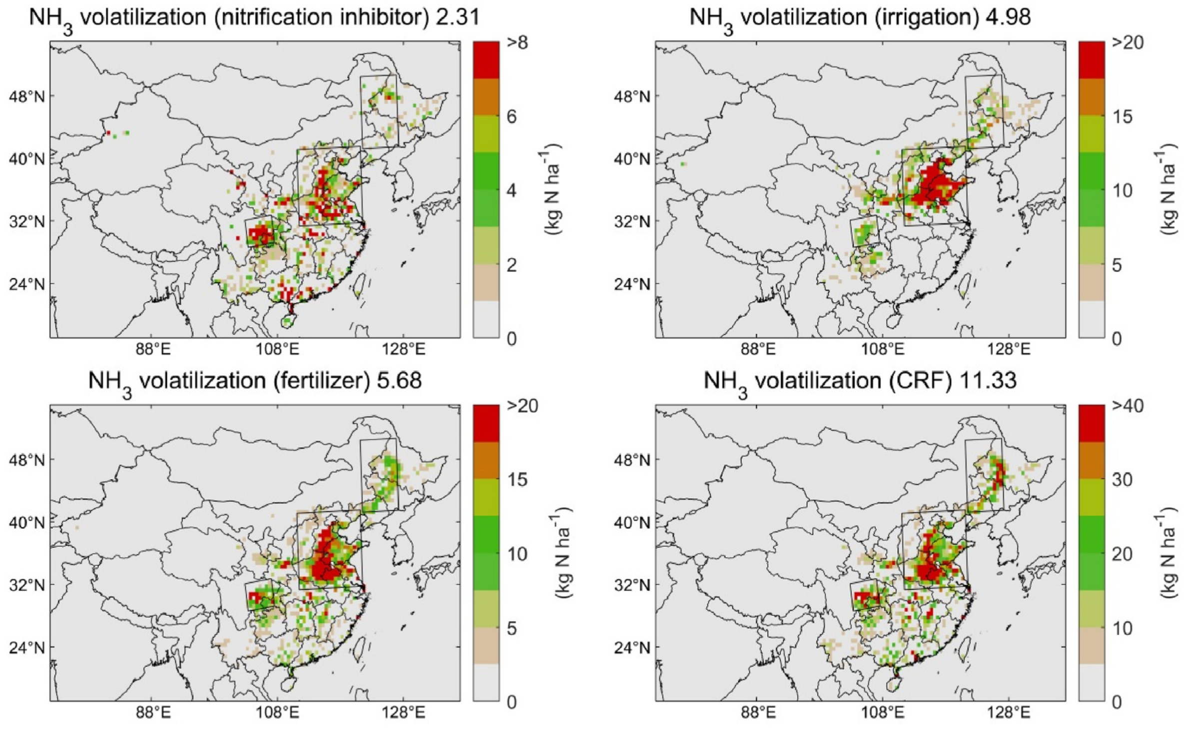

Fig. 7. Differences of NH_3 volatilization caused by different factors including nitrification inhibitor, irrigation, reduced N fertilizer amounts by 20% and controlled released fertilizer (CRF).

outputs, while the other N losses (1%) were through N leaching, N runoff, and gases (NO, N_2O, N_2) pathways. The highest NH_3 volatilization was found in NC (43 kg N ha^{-1}), followed by SC (41 kg N ha^{-1}), and NE (25 kg N ha^{-1}). The modeling results showed a 20% decrease in N fertilizer plus N manure can lead to a 25% decrease in NH_3 volatilization, and substantial decrease of NH_3 volatilization would be caused by improving management practices. This study provided improved insights into the N budgets and NH_3 volatilization towards sustainable N management in agricultural ecosystems. Our scenario analysis showed that NH_3 volatilization could be substantially impacted by soil, meteorology, N inputs as well as managements, and considering all the complex process was still a challenge for modeling NH_3 and N budgets using the revised DNDC. Nevertheless, the modified DNDC was a valuable tool for quantifying management options towards reducing NH_3 volatilization all over the agricultural systems. In the future, more verification should be done towards better parameterizing the complex processes of N budgets using ground-based measurements.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Lei Liu: Conceptualization, Methodology. **Xiuying Zhang:** Formal analysis, Writing - review & editing. **Wen Xu:** Formal analysis, Writing - review & editing. **Xuejun Liu:** Formal analysis, Writing - review & editing. **Yi Li:** Formal analysis, Writing - review & editing. **Jing Wei:** Formal analysis, Writing - review & editing. **Zhen Wang:** Formal analysis, Writing - review & editing. **Xuehe Lu:** Formal analysis, Writing - review & editing.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.114862.

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