

## RESEARCH ARTICLE

10.1002/2017GH000098

## Key Points:

- Ammonia emissions from nitrogen fertilizer use and livestock excreta increased fourfold in southern Asia (SA) during 1961–2014
- China and India together contributed about two thirds of total ammonia emissions from agricultural activities in SA between 2000 and 2014
- Summer season accounted for ~60%, while winter season accounted for ~6% of the annual total ammonia emissions in SA during 2000–2014

## Supporting Information:

- Supporting Information S1

## Correspondence to:

H. Q. Tian,  
tianhan@auburn.edu

## Citation:

Xu, R. T., Pan, S. F., Chen, J., Chen, G. S., Yang, J., Dangal, S. R. S., ... Tian, H. Q. (2018). Half-century ammonia emissions from agricultural systems in southern Asia: Magnitude, spatiotemporal patterns and implications for human health. *GeoHealth*, 2, 40–53. <https://doi.org/10.1002/2017GH000098>

Received 24 JUL 2017

Accepted 9 DEC 2017

Accepted article online 21 DEC 2017





Published online 26 JAN 2018

This article was corrected on 15 JUL 2019. The online version of this article has been modified to include a Conflict of Interest statement.

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# Half-Century Ammonia Emissions From Agricultural Systems in Southern Asia: Magnitude, Spatiotemporal Patterns, and Implications for Human Health

R. T. Xu<sup>1</sup>, S. F. Pan<sup>1</sup> , J. Chen<sup>2,1</sup>, G. S. Chen<sup>1</sup>, J. Yang<sup>1</sup> , S. R. S. Dangal<sup>1</sup> , J. P. Shepard<sup>1</sup>, and H. Q. Tian<sup>1,3</sup> 

<sup>1</sup>International Center for Climate and Global Change Research and School of Forestry and Wildlife Sciences, Auburn University, Auburn, AL, USA, <sup>2</sup>Department of Computer Science and Software Engineering, Samuel Ginn College of Engineering, Auburn University, Auburn, AL, USA, <sup>3</sup>State Key Laboratory of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, China

**Abstract** Much concern has been raised about the increasing threat to air quality and human health due to ammonia (NH<sub>3</sub>) emissions from agricultural systems, which is associated with the enrichment of reactive nitrogen (N) in southern Asia (SA), home of more than 60% the world's population (i.e., the people of West, central, East, South, and Southeast Asia). Southern Asia consumed more than half of the global synthetic N fertilizer and was the dominant region for livestock waste production since 2004. Excessive N application could lead to a rapid increase of NH<sub>3</sub> in the atmosphere, resulting in severe air and water pollution in this region. However, there is still a lack of accurate estimates of NH<sub>3</sub> emissions from agricultural systems. In this study, we simulated the agricultural NH<sub>3</sub> fluxes in SA by coupling the Bidirectional NH<sub>3</sub> exchange module (Bi-NH<sub>3</sub>) from the Community Multi-scale Air Quality model with the Dynamic Land Ecosystem Model. Our results indicated that NH<sub>3</sub> emissions were  $21.3 \pm 3.9$  Tg N yr<sup>-1</sup> from SA agricultural systems with a rapidly increasing rate of  $\sim 0.3$  Tg N yr<sup>-2</sup> during 1961–2014. Among the emission sources, 10.8 Tg N yr<sup>-1</sup> was released from synthetic N fertilizer use, and  $10.4 \pm 3.9$  Tg N yr<sup>-1</sup> was released from manure production in 2014. Ammonia emissions from China and India together accounted for 64% of the total amount in SA during 2000–2014. Our results imply that the increased NH<sub>3</sub> emissions associated with high N inputs to croplands would likely be a significant threat to the environment and human health unless mitigation efforts are applied to reduce these emissions.

**Plain Language Summary** Farming practices in southern Asia (SA), including synthetic fertilizer and manure application, have resulted in the emission of tremendous amounts of ammonia, an atmospheric constituent that has been linked to human respiratory and cardiovascular ailments and also to atmospheric haze. Ammonia emission is expected to increase due to population growth and increased demand for animal products. Ammonia emission is a critically important environmental issue that needs to be addressed collaboratively by farmers and policymakers in the SA region.

## 1. Introduction

Anthropogenic perturbation of the global nitrogen (N) cycle has contributed approximately two thirds of the annual flux of reactive N (Nr) into the atmosphere in the early 21st century (Fowler et al., 2015; Galloway et al., 2004). This substantial enrichment of Nr is mainly caused by combustion, industrial ammonia (NH<sub>3</sub>) production (by the Haber-Bosch process), and leguminous crop cultivation (Erismann et al., 2008; Fowler et al., 2015). Ammonia, as one of the major forms of Nr loss from agricultural soils, has produced adverse impacts on the environment and human health. High N fertilizer use and production of livestock wastes have resulted in a substantial increment of NH<sub>3</sub> in the atmosphere (Sutton et al., 2014; Yan et al., 2003). Ammonia is one of the key precursors for aerosol formation in the atmosphere and has caused severe air pollution in East and South Asia, which could have adverse effects on human respiratory and cardiovascular systems and might contribute to visibility reduction and regional haze (Pinder et al., 2007; Seinfeld & Pandis, 1998). For example, the NH<sub>3</sub>-induced secondary inorganic aerosol was found as a main contributing factor to haze days in China (Fu et al., 2015). Due to the significant increase of NH<sub>3</sub> in the atmosphere, more ammonium (NH<sub>4</sub><sup>+</sup>) is returned to terrestrial and aquatic ecosystems through dry and wet deposition, introducing excessive amount of Nr into natural ecosystems, which has resulted in the perturbation of the global N and carbon cycles

(Lü & Tian, 2007; Tian et al., 2003). To better assess the potential impact of  $\text{NH}_3$  emissions on human health and the environment, it is critical to have robust estimates of the spatial and temporal patterns of  $\text{NH}_3$  emissions from agricultural activities in the southern Asia (SA), home of more than 60% the world's population (i.e., the people of West, central, East, South, and Southeast Asia) (Food and Agriculture Organization of the United Nations, 2016).

The southern Asia region has experienced rapid expansion of agricultural activities (Liu & Tian, 2010; Tian et al., 2014) and has become a hotspot for studies of N-related gas emissions (Tian et al., 2016). The Haber-Bosch process produced approximately 109 Tg N fertilizer per year globally in 2014, of which more than 50% was consumed in Asia since 2004 (FAOSTAT, 2016). Nitrogen fertilizer is applied to agricultural systems to increase crop yield to meet the rising demand for food. In addition, higher demand for livestock products has stimulated the rapid growth of livestock production in the past century (Dangal et al., 2017; L. Bouwman et al., 2013). Meat consumption in major countries of Asia has grown substantially since 1990 and this trend is expected to continue (Organisation for Economic Co-operation and Development/Food and Agriculture Organization, 2016). Consequently, annual livestock waste production has increased substantially and reached approximately  $44.3 \pm 0.7$  Tg N in the 2010s (Zhang et al., 2017). Increases in both N fertilizer use and livestock excreta have resulted in increased  $\text{NH}_3$  emissions, but the magnitude and spatiotemporal pattern of  $\text{NH}_3$  emissions from the agricultural sector in SA countries have not been thoroughly investigated.

Estimating  $\text{NH}_3$  volatilization from N fertilizer application and livestock excreta has been a research topic of continuous interest for atmospheric and environmental scientists. Previous studies have used numerous approaches to achieve this research goal. The most common methodology to estimate  $\text{NH}_3$  emissions was based on emission factors (EFs). Zhao and Wang (1994) generated an inventory of  $\text{NH}_3$  emissions using source-based EFs with considerations of various emission sources from livestock, energy consumption, and N fertilizer use in Asia. Using two constant EFs in upland and rice paddy soils, Zheng et al. (2002) found that the amount of  $\text{NH}_3$  volatilization was  $13.8 \text{ Tg N yr}^{-1}$  in 2000 and is expected to approach  $\sim 18.9 \text{ Tg N yr}^{-1}$  in 2030 in Asia, mainly from increased anthropogenic N. Streets et al. (2003) developed an emission inventory for Asia in 2000 using source-based EFs and provided emission estimates for each country in the region. In their study,  $\text{NH}_3$  emission was estimated to be  $27.5 \text{ Tg N yr}^{-1}$  from all major anthropogenic sources in Asia, among which 45% and 38% were from fertilizer application and animal productions, respectively. Later, Kurokawa et al. (2013) updated the regional emission inventory using country/subregional-source-based EFs. They estimated  $\text{NH}_3$  emissions to be  $32.8 \text{ Tg N yr}^{-1}$  in 2008, of which 57% was from fertilizer application, and 20% was from manure management.

A constant EF might not provide an accurate measure of  $\text{NH}_3$  volatilization and its variability in relation to crops growth or N fertilizer application practices because the conversion of N to  $\text{NH}_3$  involves several biological and chemical processes that are strongly sensitive to environmental conditions (Fowler et al., 2015; Sutton et al., 2014). For example, Gilliland et al. (2003) indicated that static emission factors are inadequate to reflect seasonal variability in  $\text{NH}_3$  emissions. Fu et al. (2015) pointed out that environmental factors could strongly affect the  $\text{NH}_3$  volatilization from N fertilizer application. Process-based modeling could therefore address this deficiency by considering climatic variables that are not explicitly considered in the EF approach (Fu et al., 2015; Sutton et al., 2014). The single-layer version of the bidirectional  $\text{NH}_3$  exchange model (Bi- $\text{NH}_3$ ) was developed in the late 1990s, and only simulated gas exchange processes occurring on leaf surface (Sutton et al., 1995). Later, a two-layer model was developed by also considering  $\text{NH}_3$  exchange between the soil surface and the atmosphere (Nemitz et al., 2001). Currently, the two-layer Bi- $\text{NH}_3$  model is capable of simulating  $\text{NH}_3$  emissions caused by N fertilizer application in agricultural systems under various environmental conditions (Sutton et al., 2014). Further, the Bi- $\text{NH}_3$  model has been fully coupled with various ecosystem/agricultural models to simulate  $\text{NH}_3$  emissions at the global, regional, and country scale (Bash et al., 2013; Fu et al., 2015; Riddick et al., 2016). For example, the Bi- $\text{NH}_3$  module in the Community Multi-scale Air Quality model (CMAQ, Foley et al., 2010) was coupled with the United States Department of Agriculture's Environmental Policy Integrated Climate (EPIC) agroecosystem model (Bash et al., 2013; Massad et al., 2010; Nemitz et al., 2000) to simulate  $\text{NH}_3$  emissions from N fertilizer application in the United States and China. The simulated results showed that the Bi- $\text{NH}_3$  module can be applied to other regions and even at the global scale. However, previous studies using the CMAQ-EPIC approach have provided only 1 year estimates of  $\text{NH}_3$  emissions.

No previous study has specifically assessed  $\text{NH}_3$  fluxes on a decadal scale and captured the influence of reactive N sources (synthetic N fertilizer and manure) on regional  $\text{NH}_3$  emissions. In this study, we have incorporated the Bi- $\text{NH}_3$  module into the process-based Dynamic Land Ecosystem Model (DLEM version 2.0) to estimate  $\text{NH}_3$  emission from agricultural systems in SA. Our research objectives are to (1) quantify the magnitude of  $\text{NH}_3$  emissions during the 1961–2014 period at the country level and across the SA subcontinent, (2) investigate spatial and temporal variations in  $\text{NH}_3$  emissions from agricultural ecosystems during that period, (3) quantify the relative contribution of synthetic N fertilizer use and manure application on  $\text{NH}_3$  emissions, and (4) discuss potential threats of increasing  $\text{NH}_3$  emissions to air quality and human health.

## 2. Material and Method

### 2.1. The DLEM Framework for Simulating $\text{NH}_3$ Fluxes

#### 2.1.1. Overall N Flow Processes in DLEM

The DLEM is a highly integrated process-based ecosystem model that makes daily, spatially-explicit estimates of carbon, nitrogen, and water fluxes within both natural and human-dominated ecosystems (Pan et al., 2015; Tian et al., 2011). There are five key components in the DLEM: biophysical characteristics, plant physiological processes, biogeochemical cycles, vegetation dynamics, and land use. A detailed description of these processes can be found in Tian et al. (2010). The DLEM is capable of simulating the impacts of environmental changes (e.g., land use, climate, N deposition, atmospheric  $\text{CO}_2$ , N fertilizer use, and manure application) on the full N cycling processes (e.g., volatilization, mineralization, immobilization, denitrification, nitrification, N leaching, and N export and uptake), and N fluxes (e.g.,  $\text{NH}_3$ , NO,  $\text{N}_2\text{O}$ , and  $\text{N}_2$ ) between terrestrial ecosystems and the atmosphere (Lu & Tian, 2013; Ren et al., 2011, 2012; Tian et al., 2010, 2011, 2012; Xu et al., 2012, 2017). The algorithms for  $\text{NH}_3$  emissions in the previous version of DLEM can only simulate the unidirectional release of  $\text{NH}_3$  from soils to the atmosphere but are unable to simulate canopy uptake and transport. Therefore, coupling the CMAQ Bi- $\text{NH}_3$  module with DLEM can greatly improve the simulation efficiency of  $\text{NH}_3$  processes.

#### 2.1.2. $\text{NH}_3$ Fluxes in the DLEM-Bi- $\text{NH}_3$

Ammonia production is from the transformation of organic compounds and various sources of N inputs. The ammonium ( $\text{NH}_4^+$ ,  $\text{g N m}^{-2}$ ) dynamics in soils:

$$\text{NH}_4^+ = \text{NH}_4^+_{\text{soil}} + \text{NH}_4^+_{\text{input}} \quad (1)$$

where  $\text{NH}_4^+_{\text{soil}}$  ( $\text{g N m}^{-2}$ ) is the soil  $\text{NH}_4^+$  pool involved in biogeochemical processes,  $\text{NH}_4^+_{\text{input}}$  ( $\text{g N m}^{-2}$ ) is the input of  $\text{NH}_4^+$  from N deposition from the atmosphere, N fertilizer application by human, and crop N fixation (Figure 1).

In the DLEM,  $\text{NH}_4^+$  inputs into soils experience the following:  $\text{NH}_3$  volatilization, plant uptake, nitrification, or N leaching, resulting in variable amounts of  $\text{NH}_4^+$  over time. Soil  $\text{NH}_4^+$  decreases because of soil immobilization, nitrification, organic, and inorganic N leaching and runoff, while  $\text{NH}_4^+$  increases due to soil mineralization, crop N fixation, N deposition, and fertilizer/manure N application (Figure 1). These processes are regulated by environmental factors (e.g., soil temperature, moisture, and pH) and vegetation (e.g., crop type and  $\text{NH}_4^+$  uptake).

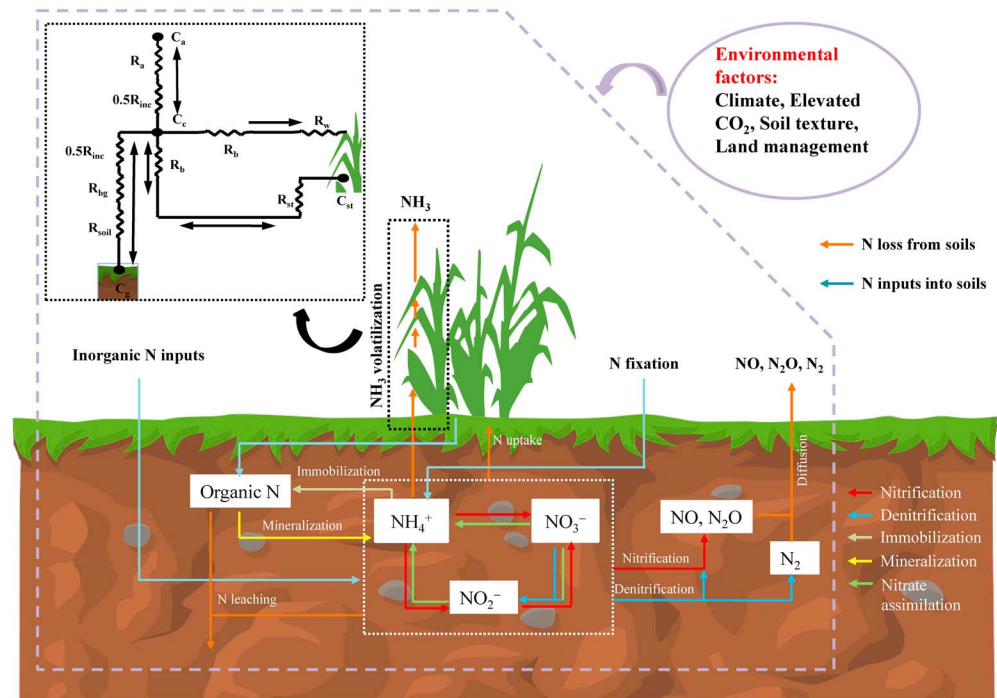
The emission of  $\text{NH}_3$  is calculated as follows:

$$F_{\text{emis}} = C_c / (R_a + 0.5 R_{\text{inc}}) \quad (2)$$

where  $C_c$  is the canopy  $\text{NH}_3$  compensation point, calculated in equation (3),  $R_a$  is the aerodynamic resistance, and  $R_{\text{inc}}$  is the aerodynamic resistance within the canopy. The unit of  $\text{NH}_3$  fluxes is  $\mu\text{g m}^{-3} \text{s}^{-1}$ ; the unit of all compensation points is  $\mu\text{g m}^{-3}$ ; the unit of all the above resistances is  $\text{s m}^{-1}$ .

$$C_c = \frac{\frac{C_{\text{st}}}{R_b + R_{\text{st}}} + \frac{C_g}{0.5R_{\text{inc}} + R_{\text{bg}} + R_{\text{soil}}}}{(R_a + 0.5R_{\text{inc}})^{-1} + (R_b + R_{\text{st}})^{-1} + (R_b + R_w)^{-1} + (0.5R_{\text{inc}} + R_{\text{bg}} + R_{\text{soil}})^{-1}} \quad (3)$$

where  $R_b$  is the quasi-laminar boundary layer resistance at the leaf surface,  $R_{\text{st}}$  is the stomatal resistance,  $R_{\text{bg}}$  is the quasi-laminar boundary layer resistance at the ground surface,  $R_w$  is the cuticular resistance,  $R_{\text{soil}}$  is the resistance to diffusion through the soil layer (Cooter et al., 2012; Sakaguchi & Zeng, 2009).



**Figure 1.** The framework of N biogeochemical processes and fluxes in the DLEM-Bi-NH<sub>3</sub>.  $R_a$ ,  $R_{soil}$ , and  $R_w$ : aerodynamic, soil, and cuticular resistance;  $R_b$ : the quasi-laminar boundary layer resistance;  $R_{bg}$ : the soil boundary layer resistance;  $R_{st}$ : stomatal resistance;  $R_{inc}$ : the aerodynamic resistance within the canopy;  $C_a$ ,  $C_g$ , and  $C_{st}$ : canopy, ground, and stomatal layer compensation point.

The soil emission potential of NH<sub>3</sub> is regulated by the ratio of NH<sub>4</sub><sup>+</sup> concentration ([NH<sub>4</sub><sup>+</sup>]) to H<sup>+</sup> concentration ([H<sup>+</sup>]), which is defined as

$$\Gamma_g = \frac{N_{fer}/(\theta_s M_N d_l)}{10^{-pH}} \quad (4)$$

where  $N_{fer}$  is the fertilizer application rate (g N m<sup>-2</sup>),  $d_l$  is the depth of soil layer (m), pH is the pH of the soil water solution,  $M_N$  is the molar mass of N (14 g mol<sup>-1</sup>), and  $\theta_s$  is the soil volumetric water content in m<sup>3</sup> m<sup>-3</sup>.

The ground layer ( $C_g$ ) compensation points are defined as follows:

$$C_g = M_n/V_m \frac{161500}{T_s} e^{(-\frac{10380}{T_s})} \Gamma_g \quad (5)$$

where  $M_n = 1.7 \times 10^7$  μg mol<sup>-1</sup>,  $V_m = 1 \times 10^{-3}$  (convert L to m<sup>3</sup>), and  $T_s$  is soil temperature in K.

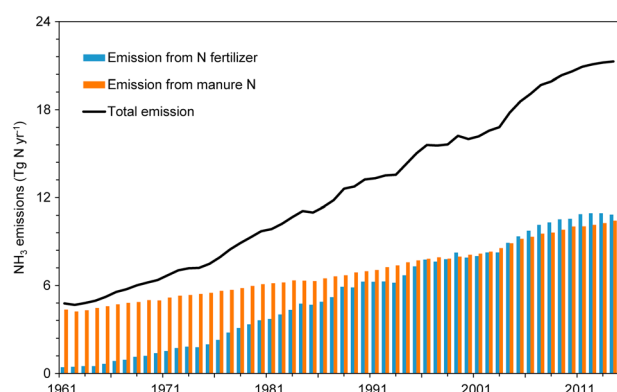
The stomatal compensation points are defined as follows:

$$C_{st} = M_n/V_m \frac{161500}{T_{can}} e^{(-\frac{10380}{T_{can}})} \Gamma_s \quad (6)$$

where  $T_{can}$  is the canopy temperature in K,  $\Gamma_s$  is the ratio of [NH<sub>4</sub><sup>+</sup>] to [H<sup>+</sup>] in the apoplast. To execute this module on a large scale, all resistances ( $R_a$ ,  $R_b$ ,  $R_{inc}$ ,  $R_{bg}$ ,  $R_{st}$ ,  $R_{soil}$ , and  $R_w$ ) and  $\Gamma_s$  were parameterized according to Massad et al. (2010).

## 2.2. NH<sub>3</sub> Emission From Livestock Excreta

Emission factors for estimating NH<sub>3</sub> emissions from livestock excreta were adopted from previous studies. A. Bouwman et al. (2002) provided a median value of EF (23%) to estimate the global NH<sub>3</sub> loss from animal manure. The range of EF in A. Bouwman et al. (2002) was 19–29%. Based on the work by Beusen et al. (2008), Riddick et al. (2016) applied a revised EF (17%) to estimate NH<sub>3</sub> emissions from animal manure. In this study, we adopted the minimum EF (17%) from Riddick et al. (2016) and the maximum EF (29%) from A. Bouwman



**Figure 2.** Annual  $\text{NH}_3$  emissions from N fertilizer application and manure production in southern Asia during the period 1961–2014.

Environmental Prediction climate forcing data (Viovy, 2014). The monthly  $\text{CO}_2$  concentration data set was obtained from NOAA GLOBALVIEW- $\text{CO}_2$  data set derived from atmospheric measurements. Spatially-explicit time series data set of agricultural N fertilizer use was developed through spatializing International Fertilizer Industry Association (IFA)-based country-level N fertilizer consumption data according to crop-specific N fertilizer application rates, distribution of crop types, and historical cropland distribution during the 1961–2013 period (Lu & Tian, 2017). All mentioned data sets were applied to drive model simulations during the 1961–2014 period and estimate  $\text{NH}_3$  emissions from N fertilizer application in SA agricultural systems.

The manure production data set was derived from Food and Agriculture Organization of the United Nations statistics website (FAO, <http://faostat.fao.org>) and defaulted for N excretion rate as described in the Intergovernmental Panel on Climate Change (IPCC) 2006 Guidelines Tier1 (Zhang et al., 2017). We used the gridded manure production data set to estimate  $\text{NH}_3$  emissions from livestock excreta during 1961–2014.

### 3. Results

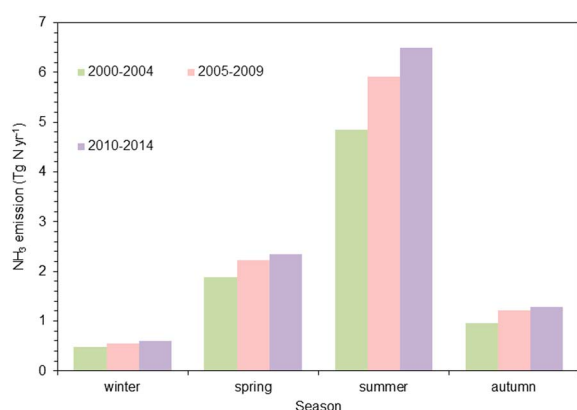
#### 3.1. Contemporary and Historical Patterns of $\text{NH}_3$ Emissions

In this study, we estimated  $\text{NH}_3$  emissions from N fertilizer application and livestock excreta in SA between 2000 and 2014. Our results indicated that the mean  $\text{NH}_3$  emission was  $19.1 \pm 3.5 \text{ Tg N yr}^{-1}$ . Ammonia emissions from N fertilizer application and livestock excreta were 9.7 and  $9.4 \pm 3.5 \text{ Tg N yr}^{-1}$ , respectively, during that period.

In SA, our results indicated that  $\text{NH}_3$  emissions were  $21.3 \pm 3.9 \text{ Tg N yr}^{-1}$  in 2014 with a rapidly increasing rate of  $\sim 0.3 \text{ Tg N yr}^{-2}$  during 1961–2014 (Figure 2). Nitrogen fertilizer application increased from 2.6 to

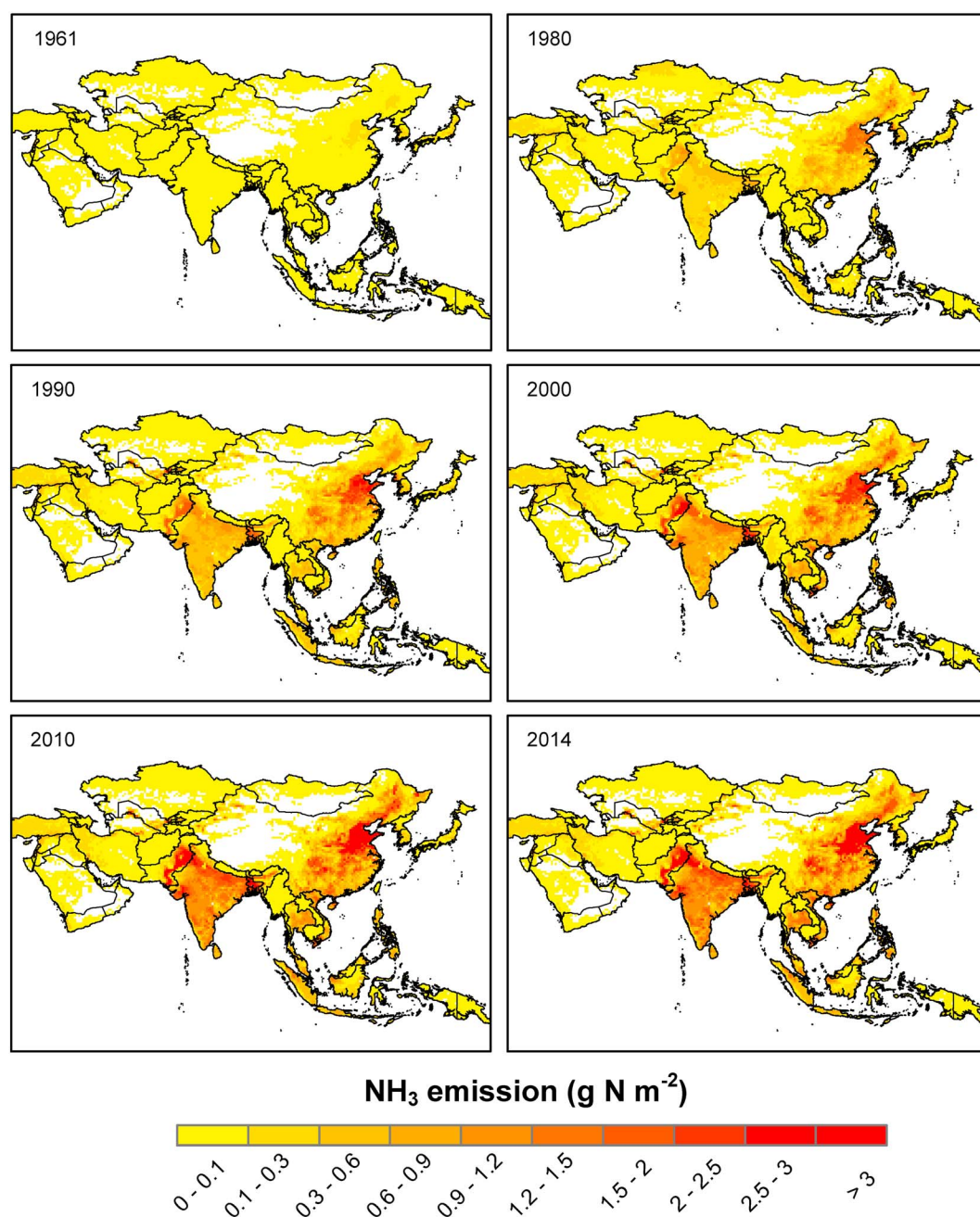
71  $\text{Tg N yr}^{-1}$  at a rate of  $\sim 1.4 \text{ Tg N yr}^{-2}$  during 1961–2013. Consequently,  $\text{NH}_3$  emissions from synthetic N fertilizer increased from 0.4 to  $10.8 \text{ Tg N yr}^{-1}$  in SA. Livestock excreta increased from 19 to  $45 \text{ Tg N yr}^{-1}$  at a rate of  $\sim 0.5 \text{ Tg N yr}^{-2}$  during 1961–2014. The  $\text{NH}_3$  emission from manure increased from  $4.3 \pm 1.6$  to  $10.4 \pm 3.9 \text{ Tg N yr}^{-1}$  in SA.

Ammonia emissions from N fertilizer application during 2000–2014 showed a substantial seasonal variation (Figure 3), which was highly correlated with monthly emission variations. On average, the summer season accounted for 59.8%, while the winter season accounted for 5.6% of annual total emissions. The highest amount of  $\text{NH}_3$  emission was in July, followed by June and August (Figure 3). The lowest amount of  $\text{NH}_3$  emission was found in January. Spring and autumn accounted for 22.5% and 12% of annual total emissions, respectively. Both seasons had an increase of  $\text{NH}_3$  emissions during 2010–2014



**Figure 3.** Seasonal variation in  $\text{NH}_3$  emissions from N fertilizer application in southern Asia during the periods 2000–2004, 2005–2009, and 2010–2014.



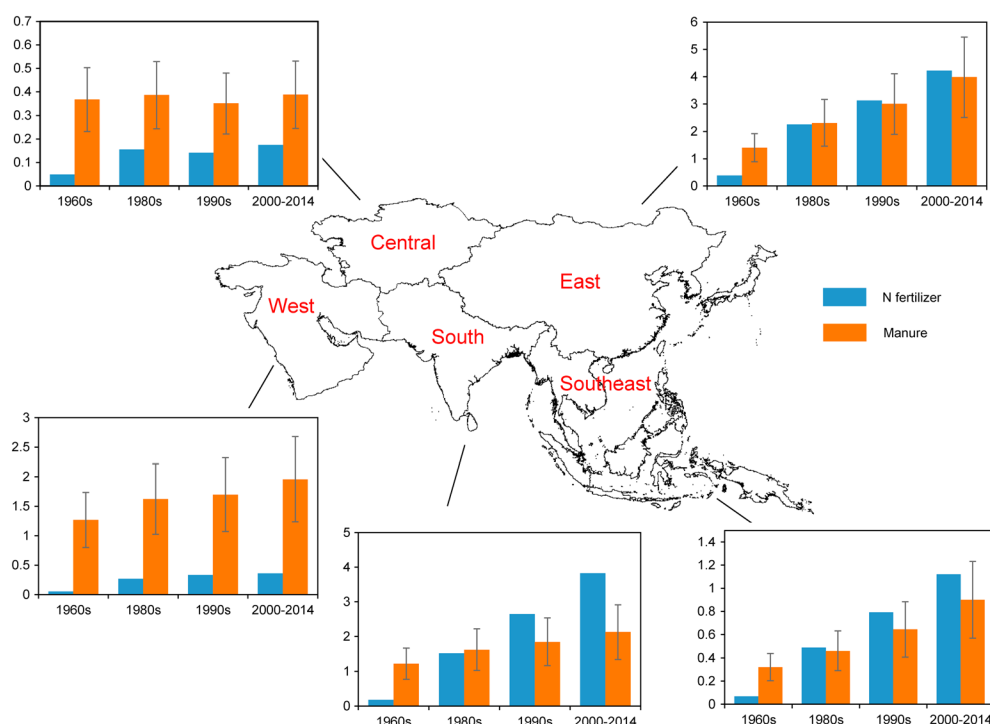


**Figure 4.** The spatial distribution of annual NH<sub>3</sub> emissions from N fertilizer application in southern Asia.

compared with emissions during 2000–2004, but with different magnitudes. In comparison to the 2000–2004 period, summer emission grew substantially during 2010–2014. The increase in emission between these two periods was of a lesser magnitude when one considers the spring and autumn months (Figure 3).

### 3.2. Spatial Pattern of NH<sub>3</sub> Emissions

Ammonia emissions from croplands varied significantly across the SA region during the past half century (Figure 4). In this study, SA was divided into five regions: Southeast, East, South, West, and central Asia. As shown in Figure 5, East and South Asia were two regions exhibiting the largest emissions and experiencing the most rapid increase in NH<sub>3</sub> emissions.



**Figure 5.** Decadal  $\text{NH}_3$  emissions in different regions of southern Asia. Emission (y axis of each graph) is reported in  $\text{Tg N yr}^{-1}$ .

In 1961, as a small amount of N fertilizer was applied to croplands in East Asia (e.g., China, South Korea, and Japan),  $\text{NH}_3$  emissions were concentrated in the north and northeast regions of China, but uniform across Japan and South Korea. A substantial amount of N fertilizer was applied in East and South Asia since the 1980s. There was a clear trend of increased  $\text{NH}_3$  emissions in most grids of Pakistan and India especially in the northern regions, and China, especially in the North China Plain during 1980–2000. After 2000, more intense  $\text{NH}_3$  emissions entirely covered India and a majority of arable land in China. A significant increase of  $\text{NH}_3$  emissions was detected in West Asia especially in Turkey and western Iran, and Southeast Asia especially in Thailand during 1961–2014.

In the 1960s, East Asia emitted more than 50% of the SA's total emissions, while South Asia accounted approximately for 25%. However, a faster rate of  $\text{NH}_3$  emission increases was found in South Asia during the past half century. During 2010–2014, South Asia accounted for more than 40% of the total emission. In Southeast Asia,  $\text{NH}_3$  emissions increased from  $35 \text{ Gg N yr}^{-1}$  to  $1.3 \text{ Tg N yr}^{-1}$  during 1961–2014 ( $1 \text{ Gg} = 10^{-3} \text{ Tg}$ ). Compared to South and East Asia, there was a slight increase in  $\text{NH}_3$  emission in Central and West Asia during the observation period (1961–2014).  $\text{NH}_3$  emissions increased sharply from  $25 \text{ Gg N yr}^{-1}$  to  $0.4 \text{ Tg N yr}^{-1}$  in West Asia during the 1961–2000 period, but only moderately during the last decade (2001–2014). In central Asia,  $\text{NH}_3$  emission was estimated to be  $0.2 \text{ Tg N yr}^{-1}$  in 2014, which was approximately sevenfold higher than the emission amount in 1961 (Figure 5).

Similar to N fertilizer-induced  $\text{NH}_3$  emissions, South and East Asia were two dominant regions that accounted for more than 50% of the total  $\text{NH}_3$  emission from manure during 1961–2014. Ammonia emission from South Asia grew from  $1.2$  to  $2.12 \text{ Tg N yr}^{-1}$ , while emission from East Asia increased from  $1.4$  to  $4.0 \text{ Tg N yr}^{-1}$  during that period (Figure 5). West Asia was the third largest source of manure-induced  $\text{NH}_3$  emission and showed an increasing trend (Figure 5). A moderate trend of increased  $\text{NH}_3$  emission was found in Southeast Asia. There was no significant increase in manure-related  $\text{NH}_3$  emission in central Asia, a sub-region that contributed to less than 10% of the total emission from manure in SA.

### 3.3. $\text{NH}_3$ Emissions From Major Countries

Eleven countries from SA (Table 1) contributed to ~65% of global total N application. China and India together contributed ~77% of total N application among these 11 countries, as shown in Table 1. In

**Table 1**  
*NH<sub>3</sub> Emissions in Major Countries of Southern Asia During 2000–2014 (1 Tg = 10<sup>3</sup> Gg): The Left Side Represents NH<sub>3</sub> Emissions From Synthetic N Fertilizer; The Right Side Represents NH<sub>3</sub> Emissions From Livestock Excreta*

Countries	N fertilizer application (Tg N yr <sup>-1</sup> )	NH <sub>3</sub> emissions (Gg N yr <sup>-1</sup> )	Countries	Livestock excreta (Tg N yr <sup>-1</sup> )	NH <sub>3</sub> emissions <sup>a</sup> (Gg N yr <sup>-1</sup> )
China	30.8	4,148	China	16.2	3,736
India	14.5	2,840	India	6.2	1,435
Pakistan	3.1	716	Pakistan	2.3	517
Indonesia	2.9	365	Turkey	1.8	406
Thailand	1.5	264	Iran	1.8	406
Vietnam	1.4	253	Jordan	1.3	296
Bangladesh	1.1	246	Syria	1.2	280
Turkey	1.4	143	Indonesia	1.0	228
Iran	1.2	105	Vietnam	0.8	188
Philippines	0.6	105	Myanmar	0.7	153
Uzbekistan	0.8	97	Afghanistan	0.5	116

<sup>a</sup>The uncertainty of NH<sub>3</sub> emissions from livestock excreta was ±26%.

consequence, ~75% of total NH<sub>3</sub> emissions were from both countries during 2000–2014. Ammonia emissions from Chinese croplands increased from 0.2 in 1961 to 4.5 Tg N yr<sup>-1</sup> in 2014. Similarly, in India, NH<sub>3</sub> emission rose from 60 Gg N yr<sup>-1</sup> in 1961 to 3.3 Tg N yr<sup>-1</sup> in 2014. Compared to China and India, the remaining countries contributed less than 25% of total N application, among which Pakistan had the highest N application and NH<sub>3</sub> emission. Turkey, Vietnam, Bangladesh, Thailand, and Iran showed similar total amounts of N fertilizer application; however, Turkey and Iran showed much less NH<sub>3</sub> emissions compared to the South and Southeast Asian countries noted above.

We identified 11 countries that showed large manure production amounts during 1961–2014 (Table 1). China and India were the two top producing countries and accounted for approximately 67% of the total manure production during 2000–2014. Ammonia emissions from Chinese livestock wastes increased from 1.0 ± 0.4 in 1961 to 4.1 ± 1.5 Tg N yr<sup>-1</sup> in 2014. In contrast, NH<sub>3</sub> emissions did not show a rapid increase in India. In the remaining countries, Pakistan produced the highest amount of manure, followed by Turkey and Iran. West Asian countries (Turkey, Iran, Syria, and Jordan) and Southeast Asian countries (Indonesia, Vietnam, and Myanmar) were identified as hotspots for NH<sub>3</sub> emissions from manure.

## 4. Discussion

### 4.1. The Importance of Asia's Contribution to Global NH<sub>3</sub> Emissions

Previous studies have presented global estimates of NH<sub>3</sub> emissions from agricultural systems (Beusen et al., 2008; Bouwman et al., 1997; Dentener & Crutzen, 1994; Riddick et al., 2016; Sutton et al., 2013; Van Aardenne et al., 2001). In 2000, the mean estimate of global NH<sub>3</sub> emission was 29.9 ± 5.2 Tg N yr<sup>-1</sup>. In this study, the estimated NH<sub>3</sub> emission from southern Asian agricultural systems was 16.0 ± 3.0 Tg N yr<sup>-1</sup>, which accounted for ~55% of the total global emissions. Although gaseous NH<sub>3</sub> has a short lifetime and low emission height (Clarisse et al., 2009), the transport of deposited NH<sub>4</sub><sup>+</sup> from one ecosystem to another could lead to widespread environmental pollution (Liu et al., 2013).

### 4.2. Comparison With Other Studies in SA

There are few regional-scale reports on NH<sub>3</sub> emissions in SA, and large uncertainties exist in the regional estimates of NH<sub>3</sub> emissions (Table 2). The research on NH<sub>3</sub> emissions from Asia conducted by Zhao and Wang (1994) and Streets et al. (2003) provided comprehensive estimates in different countries in Asia. Their estimates of total NH<sub>3</sub> emission from N fertilizer in 1990 and 2000 were 8.3 and 12.4 Tg N yr<sup>-1</sup>, which were 32% and 57%, respectively, higher than our estimates (Figure 2). Yan et al. (2003) provided a total estimate of NH<sub>3</sub> emissions in South, Southeast, and East Asia in 1995 from N fertilizer use, which was 8% lower than our estimate (6.3 Tg N yr<sup>-1</sup>). There are two primary reasons why our estimates differ from previous estimates. First, we applied a process-based model to simulate NH<sub>3</sub> emissions that considered environmental factors,



**Table 2**  
Comparison of  $\text{NH}_3$  Emissions From N Fertilizer Application and Livestock Excreta Between This Study and Previous Estimates

Country	Year	Method	NH <sub>3</sub> emissions			References
			Fertilizer	Manure	Total	
China	2010	DLEM-Bi-NH <sub>3</sub> for N fertilizer; EF for manure	4.3	4.1 ± 1.5	8.4 ± 1.5	This study
	2000		3.3	3.2 ± 1.2	6.5 ± 1.2	
	1990		2.8	2.5 ± 0.9	5.3 ± 0.9	
	2011	EPIC-CMAQ	3.0	-	-	Fu et al. (2015)
	2010	Correction EFs	4.5	5.1	9.6	Xu et al. (2015)
	2008		3.3	-	-	
	2006		3.2	5.3	8.5	
	2005	Region-specific EFs	3.6	-	-	Zhang et al. (2011)
	2005–2008		3.0	4.8	7.8	
	2005		3.5	2.8	6.3	
	2000	EPA	6.8	5.2	12	Streets et al. (2003)
	1995	EEA	3.6	2.0	5.6	Yan et al. (2003)
	1994	IPCC	6.3	1.8	8.1	Zhao and Wang (1994)
	1990		3.7	3.2	6.9	
	1990		2.0	3.8	5.8	
India	2010	DLEM-Bi-NH <sub>3</sub> for N fertilizer; EF for manure	3.3	1.5 ± 0.6	4.8 ± 0.6	This study
	2000		2.2	1.3 ± 0.5	3.5 ± 0.5	
	1990		1.5	1.3 ± 0.5	2.8 ± 0.5	
	2003	Region-specific EFs	2.1	1.7	3.8	Aneja et al. (2012)
	2000		3.3	2.8	6.1	
	1995		1.5	1.6	3.1	
	1990	IPCC	2.0	3.8	5.8	Olivier et al. (1998)

while the previous studies mentioned above applied an empirical EF. Second, the N fertilizer application data used in this study and previous studies were quite different. For example, the data set of N fertilizer application rates used in our estimate was developed by Lu and Tian (2017), while the data set used in Streets et al. (2003) was adopted from International Fertilizer Industry Association (IFA, 1998). The comparisons of  $\text{NH}_3$  emission from N fertilizer with other studies in China and India are listed in Table 2.

For  $\text{NH}_3$  emission from livestock excreta, the estimate by Yan et al. (2003) for 1995 was 4.7 Tg N yr<sup>-1</sup>, which was 17% lower than our estimate (5.6 Tg N yr<sup>-1</sup>, Figure 2). However, the estimate (10.5 Tg N yr<sup>-1</sup>) from Streets et al. (2003) was 29% higher than our estimate (8.1 Tg N yr<sup>-1</sup>) for 2000. The differences could be associated with the application of different EFs and different livestock excreta amount. In this study, we adopted a mean EF of 23% (17~29%) to estimate  $\text{NH}_3$  emissions from livestock excreta, while Yan et al. (2003) adopted the European Environment Agency (EEA) EFs based on the animal types with an average EF less than 20%. Streets et al. (2003) conceded that the use of European-based EFs in their study might have introduced uncertainty in their estimates of  $\text{NH}_3$  emission from Asian countries. Thus, future work is needed to better constrain the uncertainties associated with  $\text{NH}_3$  emission from livestock excreta. The comparisons of  $\text{NH}_3$  emission from manure with other studies in China and India are listed in Table 2.

### 4.3. Comparison of Monthly $\text{NH}_3$ Emissions With Other Studies in China

The uncertainty on monthly  $\text{NH}_3$  emissions was associated with the input climate data sets, the cropland area, and N fertilizer application dates and ratios. Large uncertainties appeared in the estimate of monthly  $\text{NH}_3$  emissions from N fertilizer application (Figure S1). Similar to the result of Xu et al. (2016),  $\text{NH}_3$  emission peaked in July and was followed by secondary peaks in June and August. Our estimates of  $\text{NH}_3$  emission in September and October were different from Xu et al. (2016). Ammonia emissions in summer contributed nearly 60% of the annual total, which was consistent with the results in Zhang et al. (2011). As shown in Fu et al. (2015),  $\text{NH}_3$  emission in September was low because of the lower amount of N fertilizer application during that time of the year.

### 4.4. Uncertainties

Uncertainties in modeled  $\text{NH}_3$  emission are due to parameters and input data sets. Fertilizer application rates are a major factor that may have affected the accuracy of  $\text{NH}_3$  emission estimates. The input data sets of soil

properties and climate are two other important factors that might affect the spatial pattern and total amount of  $\text{NH}_3$  emissions in SA. In the DLEM, we assumed that the applied N fertilizer could be used, stored, or lost within 30 days, and these assumptions could introduce uncertainties in the simulations. In addition, DLEM used empirical methods to calculate model parameters  $\Gamma_s$  and  $\Gamma_g$ , which might result in some uncertainties due to a lack of observational data to evaluate the validity of this approach (Bash et al., 2013; Fu et al., 2015). Different N fertilizer types have different EFs and thus could result in different rates of  $\text{NH}_3$  emissions. For example, urea, a major form of N fertilizer applied to croplands in developing countries, contributed more than 50% of the total  $\text{NH}_3$  emitted from N fertilizer (Zhang et al., 2011). The EF of urea was estimated as 0.2–0.25 (Bouwman et al., 1997; Matthews, 1994). In this study, we did not differentiate between N fertilizer types. Instead, all types of N fertilizer were considered as total N inputs for model simulation and that may have reduced the accuracy of the  $\text{NH}_3$  estimates. A constant EF for  $\text{NH}_3$  emissions from manure was used in this study, which might introduce more uncertainty in our estimate compared to other studies that have used region-based or animal species-based EFs. In addition, although  $\text{NH}_3$  emissions from livestock excreta are affected by climate, soil properties, and plant phenology (Hansen et al., 2017; Massad et al., 2010), the timing and rates of animal manure application were not considered in this study. This may also have added to the uncertainty of our estimates.

#### 4.5. Implications

More than half of the N added to croplands can be lost in various ways (Lassaletta et al., 2014; Tian et al., 2012). This study indicated that  $\text{NH}_3$  volatilization was one of the major outlets for Nr losses (Figure 1). As a significant source of  $\text{NH}_3$  and other precursor gases that lead to the generation of  $\text{PM}_{2.5}$ , agriculture not only affects the Earth's radiation budget but causes adverse health effects (Aneja et al., 2008; Cao et al., 2009; de Leeuw et al., 2003; Fowler et al., 1998; Seinfeld & Pandis, 1998). First,  $\text{NH}_3$  can react with other air pollutants and form aerosols (e.g.,  $\text{PM}_{2.5}$ ) in the atmosphere, thus lead to the reduction of visibility (Cheng et al., 2014). Second, the increasing concentration of  $\text{PM}_{2.5}$  is positively related to the rates of premature mortality, which increased by 21% and 85%, respectively, in East Asia and South Asia from 1990 to 2010 (Wang et al., 2017). Last, the dry and wet deposition of  $\text{NH}_4^+$  from  $\text{NH}_3$  emissions may alter soil and water chemistry (e.g., eutrophication) and reduce biological diversity (Clark & Tilman, 2008; Vitousek & Farrington, 1997).

Using a global atmospheric chemistry model (Lelieveld et al., 2015), a recent study reported that outdoor air pollution, mainly by  $\text{PM}_{2.5}$ , could lead to 3.3 (1.61–4.81) million premature deaths per year worldwide, predominantly in Asia. That study considered agriculture as the largest contributor to global premature deaths due to the releases of  $\text{NH}_3$  from livestock wastes and N fertilizer. Although Jerrett (2015) argued that Lelieveld et al. (2015) may have overestimated the toxicity of  $\text{PM}_{2.5}$  from agricultural sources, he suggested that scientists and policymakers should pay much more attention to  $\text{PM}_{2.5}$  formed by agricultural sources and its adverse effects on human health. Recently, Wang et al. (2017) indicated that reduction of  $\text{NH}_3$  emissions could maximize the effectiveness of nitrogen oxides emission controls and potentially provided some health benefits. Thus, effective farming strategies should be adopted by farmers in order to minimize  $\text{NH}_3$  loss from N fertilizer or manure application and mitigate associated threats to human health.

How can we reduce N losses from Nr? One effective strategy is to reduce the application of N fertilizer and to rotate legume crops (e.g., soybean and alfalfa) with other cereal crops (e.g., wheat and maize). Nitrogen use efficiency (NUE) was found to be generally higher in agricultural systems with a higher proportion of N inputs from symbiotic N fixation (Lassaletta et al., 2014). The second strategy is to identify the optimal crop-specific N needs to avoid excessive N application to soils (Chen et al., 2011). Mueller et al. (2017) applied N input-yield response functions to investigate regional- or country-scale NUE based on the historical N budget data (1961–2009). Their studies provided an overview of the optimal N input rates in major agricultural countries and showed that East, South, and Southeast Asia received excessive N inputs up to  $10 \text{ g N m}^{-2}$  greater than needed for optimal crop yield. The third strategy is to minimize the loss of N through improved management techniques (Bouwman et al., 1997, 2002; Chen et al., 2011, 2014; Fowler et al., 2015). In addition, some strategies have been approved to be effective for reducing N losses from animal manure in SA. For example, modifying feeding (diet with optimized N content) is an effective strategy to reduce gaseous N losses from animal manure (Chadwick et al., 2011; Hou et al., 2015). In comparison with composting, solid manure storages via compaction and covering could substantially decline  $\text{NH}_3$  emissions (Chadwick, 2005; Hou

et al., 2015). Compared to surface application, manure application through band spreading, incorporation, and injection could significantly reduce  $\text{NH}_3$  emissions (Hou et al., 2015).

Asia, whose population could increase by 0.9 billion by 2050, has been projected to be the second largest contributor to future population growth (United Nations, Department of Economic and Social Affairs, World Population Prospects, 2015). Valin et al. (2014) reported that food demands would increase by 59–98% between 2005 and 2050 in the reference scenario (the GDP and population pathways of the “middle of the road” Shared Socio-economic Pathway (SSP2) developed by the climate change impacts research community). In the projection of FAO and agriculture models, South and East Asia are expected to experience a rapid increase in crop foods and livestock products during the first half of the 21st century (Alexandratos & Bruinsma, 2012; Valin et al., 2014). Under this scenario, N fertilizer application to global croplands would double by 2100 (Stocker et al., 2013), while manure production may increase to 50% by 2050 compared to 2000 (Bouwman et al., 2013). China and India are the top two countries with the largest N fertilizer consumption and are expected to remain so in the foreseeable future. Unfortunately, NUE has rapidly decreased in these countries (Tian et al., 2012; Zhang et al., 2015). The decrease in NUE and increase in N fertilizer use would translate into greater emission of fertilized N as  $\text{NH}_3$  and other N gases (Zhang et al., 2015). Thus, farmers as well as policymakers in Asian countries ought to consider using N fertilizer more efficiently for the benefits of environmental quality and human health (Tian et al., 2016; Zhang, 2017). Moreover, livestock excreta in SA increased from 19 to 45 Tg N  $\text{yr}^{-1}$  during 1961–2014 (Zhang et al., 2017). In 2000, Southeast, East, and South Asia contribute ~30% of global GHG emissions from ruminants (Herrero et al., 2013). In order to reduce both GHG and  $\text{NH}_3$  emissions from livestock systems, it is important to implement an effective strategy for managing animal manure (Bouwman et al., 2013; Fowler et al., 2015).

#### Acknowledgments

This study has been supported by National Key Research and Development Program of China (2017YFA0604702), National Science Foundation (1210360 and 1243232), the Chinese Academy of Sciences STS Program (KFJ-STS-ZDTP-0), SKLURE grant (SKLURE2017-1-6), NASA Land Cover/Land Use Change Program (NNX08AL73G S01), and Auburn University Peak of Excellence Program. We would thank two anonymous reviewers who have provided thoughtful comments and suggestions, which led to a major improvement in the paper. The availability of climate data,  $\text{CO}_2$  concentration data, global cropland distribution data, and manure/fertilizer N inputs is described in section 7 and the supporting information. The model output data in this study are archived in International Center for Climate and Global Change Research at Auburn University (<http://wp.auburn.edu/cgc/>).

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