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Ammonia emissions may be substantially underestimated in China

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Abstract

China is a global hotspot of atmospheric ammonia (NH_3) emissions and, as a consequence, very high nitrogen (N) deposition levels are documented. However, previous estimates of total NH_3 emissions in China were much lower than inference from observed deposition values would suggest, highlighting the need for further investigation. Here, we reevaluated NH_3 emissions based on a mass balance approach, validated by N deposition monitoring and satellite observations, for China for the period of 2000 to 2015. Total NH_3 emissions in China increased from 12.1 ± 0.8 Tg N yr^{-1} in 2000 to 15.6 ± 0.9 Tg N yr^{-1} in 2015 at an annual rate of 1.9%, which is approximately 40% higher than existing studies suggested. This difference is mainly due to more emission sources now having been included and NH_3 emission rates from mineral fertilizer application and livestock having been underestimated previously. Our estimated NH_3 emission levels are consistent with the measured deposition of NH_x (including NH_4^+ and NH_3) on land (11-14 Tg N yr^{-1}) and the substantial increases in NH_3 concentrations observed by satellite measurements over China. These findings substantially improve our understanding on NH_3 emissions, implying that future air pollution control strategies have to consider the potentials of reducing NH_3 emission in China.

Keywords: fertilizer; nitrogen deposition; mass balance; satellite measurements; agriculture; temperature

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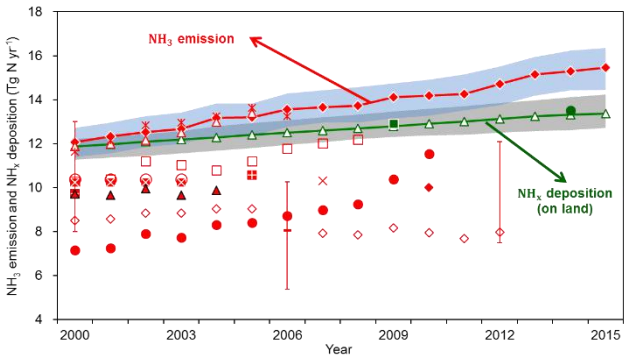

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INTRODUCTION

Nitrogen (N) plays an important role in all living systems and their environment.[1, 2] Reactive N (N_r) released to the environment is dispersed by atmospheric and hydrologic transport processes and can accumulate in air, soils, vegetation, and groundwater. [1] Almost all emitted N_r in the forms of nitrogen oxides (NO_x, defined as the sum of all species that contained oxidized nitrogen) and ammonia (NH_3) is transferred back to the Earth's surface within hours to days of its release. [2] NH_3 emitted to the atmosphere is either deposited directly or transformed into an ammonium aerosol (e.g., ammonium nitrates and ammonium sulfate) often transported over long distances. [3] Two types of NH_x (NH_3 (gas) + NH_4^+ (aerosol)) deposition usually occur: dry deposition of NH_3 close to the emission sources, [4, 5] and wet deposition of NH_4^+ that can occur at far distances downwind from the sources. [6] N biogeochemical cycles follow the principle of mass balance, and the N_r input to and output from the atmosphere system are normally balanced.[7] Therefore, NH_x deposition fluxes are closely related to NH_3 emissions. Galloway et al.[8] and Fowler et al.[2] elaborated the process of global N cycling and estimated that global NH_3 emissions and NH_x deposition are in balance (Table 1). Global NH_x deposition depends strongly on total NH_3 emissions, with the spatial distribution of emissions and atmospheric transport pathways affecting the downwind deposition fluxes to the oceans.[9]

Mainland China has long coastlines (total length about 18 000 km), bordering the Northwestern Pacific Ocean, which is primarily located downwind of emission sources on the Asian continent.[10] As a consequence, long-range transport will lead to a share of ammonium aerosols being deposited outside of China's land area, because prevailing wind directions and river catchment flows of China result in pollutants being carried from China to the North Pacific Ocean.[11-13] Figure S1 illustrates the sources and fates of NH_3 in China. It is noticeable that the Tibetan Plateau blocks most pollutant transfer into China from other countries in the west such as India though some evidence has emerged of transport of air pollutants across the Himalayas.[14, 15] China receives little NH_x from other countries through atmospheric circulation, but transports N_r to surrounding marine ecosystems, and is overall a net exporter of NH_x deposition.[14] Therefore, the flux of NH_x deposition on land could be used to constrain the spatial and temporal variations of NH_3 emission in China.[16]

Over the past two decades, China has witnessed a substantial increase in N_r pollution and has been a global "hotspot" for both NH_3 emissions and N deposition due to rapid increases in industrialization, urbanization and intensified agricultural production.[7, 17, 18] Rapid increases in atmospheric NH_3 concentrations and the subsequent N depositions have various effects on ecosystems, such as soil acidification, water eutrophication, biodiversity loss and air pollution.[19, 20] Although many studies on emission inventories in general and NH_3 emissions in particular have been conducted in China, large uncertainties and contradictory results were found in terms of spatial and temporal variations of NH_3

emissions (e.g., Dong et al.[21]; Huang et al.[22]; Kang et al.[23]). Total estimates of NH_3 emissions available for China range from 7-8 Tg N yr^{-1} [23, 24] to around 11-12 Tg N yr^{-1}. [25, 26] Some studies[24, 26] recorded an increasing trend, while other studies[23] argued a downward trend of total NH_3 emission in China for the recent decade. Previous studies rarely used other data apart from ground-based concentration measurements for emission verification, such as N deposition monitoring or satellite observation to calibrate estimates and validate NH_3 inventories.

Recent studies have addressed the important role of NH_3 in the formation of fine particles ($PM_{2.5}$) in China,[27, 28] putting more of an emphasis on the need to refine and better quantify NH_3 emissions and their contribution to air pollution, including for urban areas, in China. Thus, in this paper we aimed to (i) advance our understanding by designing a systematic framework for the analysis of NH_3 sources, emissions, and environmental fates in China; (ii) review and revise the NH_3 emission inventory from 2000 to 2015 in China with a mass balance approach; and (iii) evaluate the uncertainties attributed to NH_3 emissions by validation of NH_x deposition and satellite measurements.

METHODS

Datasets. This study covers the entire land area of mainland China; Taiwan, Hong Kong, and Macao were excluded owing to data limitations. Data used in this study can be divided into two categories: (i) summary information for China such as population, GDP, land use, fertilizer use, crop/livestock production, and energy consumption in different sectors, all taken from the national data center[29] and FAO statistics[30]; (ii) parameters and coefficients used for the calculation of NH_3-N fluxes, both obtained from synthesis of peer-reviewed literature and field measurements. We established multiple datasets for the calculation of NH_x emissions from 2000 to 2015 in China on a provincial scale. Note that all the units of NH_x fluxes have been converted to Tg N yr^{-1}. Details about the selection criteria applied to and parameters and coefficients can be found in SI methods.

Model Description. We used the Coupled Human And Natural Systems (CHANS) model to quantify NH_3 fluxes within China. The CHANS model incorporates and integrates all N_r fluxes and their interactions that can be identified, together with the linkages among subsystems (Figure S2). The basic principle of the CHANS model is mass balance for the whole system and each subsystem. A detailed description of CHANS can be found in Gu et al.[31] In this study, we focus on the atmosphere subsystem (AT) which receives NH_3 input from 13 subsystems and deposits NH_x to land subsystems. In addition, it can also transfer NH_x to or receive NH_x from other countries/oceans through atmospheric circulation. The input of NH_3 is either larger or equal to the output of NH_x. A summary of the main source categories comprised in the NH_3 emission inventory is listed in Table S1.

Meteorological conditions strongly influence the rate of NH_3 emissions.[32, 33] We

quantitatively estimate the impacts of climate change on NH_3 emission based on the climate-dependent paradigm developed by Sutton et al.[32] This climate-dependent paradigm is generally universal among regions, and can be used in China. In principle, according to solubility and dissociation thermodynamics, NH_3 volatilization potential nearly doubles for an increase of temperature by 5°C, equivalent to a Q_{10} (the relative increase over a range of 10°C) of 1–4.[32] Note that in this study, we only consider the temperature-dependence effect on agricultural sources given that to date only few studies have emerged in literature which thoroughly quantify the effect of temperature on non-agricultural sectors.[34] Base on Sutton et al.,[32] an average Q_{10} of 2 was used for NH_3 EFs from fertilizer application across China; an average Q_{10} of 1.25 was used for NH_3 EFs from pigs, sows, poultry, rabbits, sheep and goats while an average Q_{10} of 2.5 for cattle, horses, donkeys, mules. Prior to the calibration of temperature-dependence effects, we have summarized the average NH_3 EFs using correction coefficients for various factors. Details on the approach can be found in SI text.

The annual average temperature for fifteen years (2000-2015) at 9.7°C across China has been selected as a reference temperature and warmer or colder annual averages as a proxy for calibration of the NH_3 emission. The calculation principles are as follow:

$$AT_{IN} = \sum_{i=1}^{13} E_{Item,i} + ATIN_{Import} \quad (1)$$

$$E_{Item,i,j} = \sum_p EF_{i,j,p} \times \mathrm{f}(T_{j,p}) \times AL_{i,j,p} \quad (2)$$

$$\mathrm{f}(T_{j,p}) = \frac{(Q_{10,j,p}-1)}{10} \times (T_j - T_0) + 1 \quad (3)$$

$$AT_{OUT} = ATOUT_{Dep} + ATOUT_{Export} \quad (4)$$

$$AT_{IN} \geq AT_{OUT} \quad (5)$$

where AT_{IN} and AT_{OUT} are the total NH_x input to and output from atmosphere subsystem; $E_{Item,i,j}$ is the NH_x emission from other 13 subsystems to atmosphere; i, j and p represent the subsystem, the year and the source type, respectively; $EF_{i,j,p}$ is the corresponding emission factor (EF); $\mathrm{f}(T_{j,p})$ represents a function of climate effect on NH_3 emission; T_j is the annual average temperature (°C), T_0 represents the fifteen years (2000-2015) average temperature (9.7°C); $Q_{10,j,p}$ stands for a temperature effect on the NH_3 volatilization potential; $AL_{i,j,p}$ is the activity data; $ATOUT_{Dep}$ is the NH_x deposition on land, including both dry and wet deposition. $ATOUT_{Export}$ is the NH_x transferred to surrounding areas (mainly to ocean) through atmospheric circulation that advects NH_x away from China.

Uncertainty analysis. Monte Carlo simulation was used to quantify the variability of the NH_3 fluxes. In Monte Carlo simulations, random numbers are selected from the normal or uniform distribution of input variables and a variation range is attributed to the emission inventory. In order to thoroughly test the accuracy of the simulation results, 10,000 Monte Carlo simulations were executed to estimate the range of NH_3 emission

uncertainties for different sources. Meanwhile, the mass balance approach used in the CHANS model constrains the uncertainty ranges and helps to refine the overall uncertainty.[7] Details about the uncertainty assessment can be found in the SI text and tables.

RESULTS

Total NH_3 emission in 2015. The total NH_3 emission into the atmosphere was estimated at 15.6±0.9 Tg N in 2015 for China. Agricultural sources were the largest contributors (13.6±0.8 Tg N), accounting for 88% of total NH_3 emissions, with 5.8±0.3 Tg N and 6.6±0.5 Tg N stemming from cropland and livestock, respectively. The majority of the NH_3 emissions from cropland originated from the application of mineral fertilizers (5.4±0.2 Tg N), and the rest from irrigation, agricultural soils, N-fixing crops, and composting of crop residues (Figure S3). An estimated 3.8±0.2 Tg N was emitted from livestock housing and manure storage. In addition, 2.8±0.3 Tg N was emitted from manure application to cropland, which in some studies has been attributed to cropland emissions.[35, 36]

The spatial distribution of NH_3 emissions on a provincial scale in 2015 is shown in Figure 1a, revealing a strong spatial variability and association with the distribution of arable land. High NH_3 emission densities above 50 kg N ha^{-1} were concentrated across the North China Plain, where intensive agriculture for both crop production and animal husbandry are located. Sichuan Basin, Middle South China and Northeastern China also show high NH_3 emission densities. In contrast, low NH_3 emission densities of less than 10 kg N ha^{-1} were primarily found across western China, e.g. Tibet, Qinghai, Inner Mongolia and Gansu, which are characterized by dry regions with low agricultural production and little N fertilizer use.[29]

Temporal trends of NH_3 emissions. Total NH_3 emissions increased from 12.1±0.8 Tg N in 2000 to about 15.6±0.9 Tg N in 2015 with an annual rate of 1.9% (statistically significant). ~85% of the inter-annual variations could be well explained by the changes of human activity levels (Figure S4), and the remaining 15% were attributed to air temperature changes during this period (Figure S5). Agriculture is the main emission source, accounting for ~87% of the total NH_3 emission in China, with on average, 43.1% (41.6-44.1%) contributed by livestock manure and 36.4% (35.0-38.3%) by fertilizer application. Non-agricultural sources account for only about 13% of total national NH_3 emissions, including humans responsible for 6.6% (5.1-8.8%), other sources (fuel combustion, waste disposal, traffic sources, chemical industry, urban green land) contributed to less than 2% (Figure S4).

Uncertainty assessment. The range of NH_3 emissions with a 95% confidence interval for 2000, 2005, 2010, 2015 was estimated at 11.4-12.7, 12.3-13.8, 13.3-14.9, and 14.5-16.6 Tg N yr^{-1}, respectively (Figure 2). Uncertainty contribution and variation ranges of

different emission source sectors are presented in Table S14. Livestock manure and fertilizer application were identified as the key sources, with contributions to overall uncertainty of 43.1% and 36.4%, respectively.

DISCUSSION

Validation by NH_x deposition. In contrast to the bottom-up estimates of NH_3 emission in current inventories, NH_x deposition can be comparatively well constrained based on data from field monitoring. Based on our NH_3 emission estimations, we could derive the range of NH_x deposition in mainland China (Dep_derived = NH_3 emission – exported + imported, where exported NH_3 flux accounts for ~20% of NH_3 emission[14], and imported N flux from outside contributed by foreign anthropogenic emissions is ~1 Tg N yr^{-1}[10]). The derived NH_x deposition was calculated at 12.6 Tg N in 2010 (Figure 2).

To be directly comparable with the observation data of NH_x deposition, Figure 2 integrated the NH_x deposition on land for China (data extracted from Liu et al.[17] and Xu et al.[6]). Liu et al.[17] and Xu et al.[6] conducted a comprehensive evaluation of N deposition dynamics across China (Figure S6) based on a Nationwide Nitrogen Deposition Monitoring Network (NNDMN). Result indicated a NH_x deposition around 11-14 Tg N yr^{-1} over land areas in China during 2000-2015, with a 12.9 Tg N in 2010. A range of additional studies based on N deposition monitoring data also support the estimated range of NH_x deposition over China (Table 2). Therefore, the derived NH_x deposition from our estimate of NH_3 emission agreed well with the observed NH_x deposition over land areas in China.

It is noticeable from Figure 2 that the difference between NH_3 emissions and terrestrial NH_x deposition increased in recent years. We assume that this can be attributed to the increased long-distance transport of NH_x deposition to the ocean. Elevated NH_x deposition rates found in the North and Northwestern Pacific Oceans[11, 13] demonstrates that the formation of ammonium sulphate and –nitrate extended the lifetime of NH_3 in the atmosphere, promoting its long-range transport to ocean.[10] This is in line with the observation that, despite the most recent reductions of NO_x and SO_2 emissions [37] in China, the molar amount of ($2SO_2$+NO_x) still substantially exceeded that of NH_3 at least until 2015 (Figure 3), suggesting that NH_3 emissions presented the limiting factor to the formation of ammonium aerosols. Thus, the increase of NH_3 emission would increase the formation of ammonium aerosols and the long-range transport to ocean during 2000-2015 in China. It should be noted that while Figure 3 presents a mass balance limit to the relative components of each species, other factors, i.e. temperature, relative humidity, and aerosol composition could affect the limitations of aerosol formation,[27] and a full atmospheric chemistry transport model assessment would be needed to further assess limiting reagents.

Validation by satellite observations. Ground-based measurements of ambient NH_3 are

sparse and not always representative for a larger area. Satellites provide an alternative and are ideal to measure NH_3 spatial and temporal variability on global scale.[38, 39] Figure 1 presents the spatial distributions of NH_3 emission and the NH_3 vertical column densities (VCDs) measured with the Infrared Atmospheric Sounding Interferometer (IASI) satellite for 2015 over China.[40] The spatial variability of our estimates of the NH_3 emission densities agree well with the IASI-NH_3 VCDs distribution, with the largest NH_3 emission density found in the North China Plain, Sichuan Basin and Northeastern China. Relatively high NH_3 VCDs could also be observed in Xinjiang by satellite IASI instrument, which is not captured by our emission map. There is little emission of SO_2 and NO_x from industrial sources in Xinjiang compared to that in Eastern China.[41] Therefore, a reduced conversion rate to aerosol is expected. This, and the dry climate is probably responsible for a longer lifetime of NH_3 in the atmosphere in Xinjiang compared to other regions in China. This will lead, in those regions to a larger buildup of NH_3, and hence larger observed columns and a larger qualitative discrepancy with the emission inventory.

Recently, Warner et al.[33] showed an increasing trend of NH_3 VCDs in China from 2003 to 2015 with an increment of 0.076 ± 0. 020 ppbv (parts-per-billion by volume) per year (2.3% yr^{-1}) using measurements of the Atmospheric InfraRed Souder (AIRS) satellite. This increasing trend of NH_3 VCDs in China agreed well with our results that showed an annual increase rate of 2.0% during the same period. Generally, the increased NH_3 emissions contributed to the variation in NH_3 VCDs. It suggested that the decreasing trends or trends with turning points of NH_3 emission from 2000 to 2015 found in previous studies are not accurate (Figure 2). For instance, a recent study by Kang et al.[23] estimated that total NH_3 emissions in China showed a downward trend from 8.5 Tg N in 2000 to 8.0 Tg N in 2012, contradictory to the increasing trend found in satellite measurements. However, note that satellite concentrations should not be directly compared with emissions, as NH_3 columns are in addition to sources, determined by transport and sinks[40, 41]. Hence, we suggest such data is primarily used for qualitative comparisons such as temporal trend analysis.

Comparison with other studies. Figure 2 indicates that our results estimated NH_3 emissions to be about 40% higher than previous studies suggested, majority of which estimated a lower amount of NH_3 emission than inference from observed NH_x deposition would yield. Table 3 shows a quantitative comparison with previous studies from five main sources, and the differences are mainly arising from estimates of emissions from fertilizer use and livestock production. In addition, we included additional emission sources compared to other studies, such as grassland, aquaculture, traffic sources, urban vegetation, humans and pets, even though the latter contributed a relatively small proportion of total emissions (SI text).

The total amount of mineral fertilizer applied and emission factors (EFs) for NH_3 volatilization are two key factors that directly affect NH_3 emissions from N fertilizer

application. Compared to other studies, the difference regarding estimated activity data (i.e. amount of fertilizer uses) is less than 5%. However, the final corrected EFs (16.2-18.4%, average 17.0%) used in this study were about 16.6% higher than those used in other studies (Figure S10, average 14.6%). The majority of previous inventories compiled for China used constant European-based emission factors (EFs) (e.g., Dong et al.[21]; Kang et al.[23]), and did not adjust for specific agricultural practices, environmental conditions and climatological factors. Some studies[23, 42] adopted much lower EFs (Figure S10, average 12.8%) because they believed that an anticipated shift of dominant fertilizer types from ABC to urea would substantially reduce the NH_3 emission.[23, 42] In fact, the actual EFs are substantially influenced by the method of fertilizer application used. ABC is normally applied as base fertilizer with deep placement that results in a lower EF, while urea is widely used for top-dressing and surface application[43, 44]. The percentage of topdressing for urea is higher than that for ABC (Table S5) and as such is likely to substantially increase NH_3 emissions in China.[44] Therefore, the gradual shift of fertilizer type from ABC to urea would not significantly change the overall NH_3 EF for fertilizer application in China.

A recent meta-analysis [45] on the topic of NH_3 volatilization from global fertilizer use indicates that the global average percentage of N lost as NH_3 was 17.6%. The NH_3 volatilization rate for China is expected to be higher than this value, given its higher application rate and lower N use efficiency considering the fact that most farming is done on smallholder farms, which rely mainly on family labor and are traditionally slow to adapt technical improvements such as 4R fertilization management.[46]

Table 3 indicates that the total NH_3 emissions from livestock calculated in this study were 1.4 Tg N yr^{-1} higher than other studies on average, such as REAS2.1,[26] Xu et al. [35] and EDGARv4.3.1.[24] The EFs for livestock used in this study are compared with other studies in Figure S6. In fact, due to the absence of closed systems to produce liquid manure, the air-dry process used to produce manure for field application in China results in high emissions of NH_3 from livestock production.[47] In addition, the backyard and small-scale livestock farms still dominate animal production in China, which is difficult to supervise the NH_3 emission from livestock production and further introduce advanced technologies to reduce the NH_3 emission due to high costs.[47] Therefore, even assuming the same excretion rate for livestock, the resulting NH_3 emission rate is expected to be higher in China because manure management systems are not as advanced as developed countries.

Uncertainty and Limitation. To estimate the uncertainty range on a national scale is challenging given the large spatial variability of EFs of NH_3. A simple additive approach for all uncertainties from each region and source would undoubtedly exaggerate the overall uncertainty. The NH_3 emissions in this study were calculated based on a full life-cycle analysis, which is a framework to quantify and track the trajectory of all nitrogen

fluxes in the CHANS.[7, 31] Thus, the uncertainty range inherent to the CHANS model could be well constrained by the mass balance calculation in all the 13 subsystems, combining with Monte Carlo simulations.

However, we have identified several limitations of this study, especially for the major emission sources of N fertilizer and livestock. The actual EFs for fertilizer application are substantially influenced by many parameters, including meteorological conditions, soil properties, fertilizer application methods, application rate, fertilizer type and so on.[48] However, generating a matching dataset of human activities data (e.g., N fertilizer application rate) with the same degree of detail and resolution on national scale is outside the scope of this study. At the same time, the influence of these factors on NH_3 emission at a large scale has not yet been well studied and quantified, to our knowledge. Therefore, it is difficult to integrate all the factors into the CHANS and comprehensively quantify the effects of those factors. Future work to build comprehensive datasets including both spatial and temporal variations of the key influencing factors would help to refine the results.

In addition, we assume there is no significant inter-annual change in the percentage of intensive rearing systems, manure management practice which is typically affected by many factors, including the housing structure, manure storage system, spreading technique, and so on[49]. However, manure management practices in China have been subject to great changes over the time period of 2000-2015,[29] hence additional uncertainties may be introduced due to using constant parameters for livestock manure during the study period[23].

Some uncertainties may still exist in the observed NH_x deposition used in this study due to the relatively limited number of sampling sites. In addition, NH_3 deposition may be overestimated at rural sites with relatively high canopy compensation points as a result of fertilized croplands or vegetation[6]. Furthermore, there may also be large uncertainties arising from comparison with satellite data, because observed differences between ammonia emission and NH_3 VCDs remain unclear to some extent, for instance, in locations such as Xinjiang. These uncertainties may affect the validation of NH_3 emissions in this paper.

Outlook. Refining the NH_3 emission inventory datasets with high spatiotemporal resolution is crucial for the assessment of future policy implications with regard to mitigation options. This paper highlights that the overall amount of NH_3 emissions may be substantially underestimated in China (Figure 2). However, we still lack a substantial amount of information on spatial resolution at regional to local scales, as well as future changes, which are decision making. Further work is still required to increase the reliability and accuracy of NH_3 emission inventory datasets, underpinned by high-resolution observations, process-based experiments and model-data assimilation to fully quantify more realistic NH_3 emissions. This can enhance our understanding on the variation of NH_3 emission and its driving forces.

Furthermore, NH_3 plays a crucial role in the formation of secondary inorganic aerosols (SIAs) that are the predominant components of fine particles in China.[20]

However, until now no control strategies have yet been implemented for NH_3 emissions in China. Effective strategies for the reduction of NH_3 emissions in China are thus urgently needed given a 40% higher NH_3 emission than previous thoughts. In support of such strategies, a comprehensive and accurate NH_3 emission inventory is also vital to future air pollution prevention and control in China, which can be integrated into air quality model and serve as a baseline toward tracking emission trends, developing mitigation strategies, and assessing progress.

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Note

The authors declare no competing financial interest.

Supporting Information Available

The Supporting Information includes is available free of charge on the ACS Publications website.

Text S1−4, Table S1−16 and Figure S1−9 (PDF).

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546 **Table 1 N input, NH_3 emission and deposition in the world and China**

	World			China
	1860[a]	early-1990s[a]	2010[b]	2010[c]
Total terrestrial N input	**135**	**263**	**278**	**61.3**
Fertilizer production	0	100	120	37.1
NBNF	120	107	58	7.1
CBNF	15	31.5	60	4.6
NO_x-FF	0.3	24.5	40	6.6
NH_3 emission	**20.5**	**58.2**	**69**	**14.0**
Terrestrial	14.9	52.6	60	14.0
Marine	5.6	5.6	9	-
NHx deposition	**18.8**	**56.7**	**64**	**13.8**
Terrestrial	10.8	38.7	40	13.1[d]
Marine	8	18	24	0.7[e]

547 Unit: Tg N yr^{-1}; Fertilizer production, Haber–Bosch N fixation for fertilizer use; NBNF,
548 natural biological N fixation; CBNF, cultivated biological N fixation; NO_x-FF, NO_x
549 emission via fossil fuel combustion.
550 a, adapted from Galloway et al.[8]
551 b, adapted from Fowler et al.[2]
552 c, adapted from Gu et al.[7]
553 d, adapted from Liu et al.[17]
554 e, adapted from Luo et al.[10]
555

556 **Table 2 NH_x deposition on land area of China.**

Studies	Base year	Deposition density (kg N ha^{-1} yr^{-1})			Total deposition (Tg N yr^{-1})
		Dry	Wet	Total	
Jia et al.[50, 51]	2000-2009	-	-	13.9	12.9
Liu et al.[17]	2000-2010	-	-	14.3	13.3
Xu et al.[6]	2010-2014	-	-	14.5	13.5
Liu et al.[18]	2003-2014	-	6.8	-	6.3*
Jia et al.[51]	2005-2014	6.1	-	-	5.7*
Zhu et al.[52]	2013	-	7.3	-	6.8*

557 * Only wet or dry NH_x deposition.

558

559 **Table 3 Comparison of NH_3 emissions with other studies**

Studies	Year	Total	Fertilizer	Livestock	Humans	Burning	Others
REAS	2000	10.3 // +1.8	4.2 // -0.2	4.2 // +0.9	1.2 // -0.1	0.5 // -0.3	0.2 // +1.4
Wang et al.	2005	13.4 // -0.2	4.2 // +0.1	5.8 // -0.1	0.2 // +0.8	-	3.3 // -1.2
Huang et al.	2006	8.1 // +5.1	2.6 // +2.1	4.4 // +1.3	0.2 // +0.7	0.1 // +0.2	0.8 // +0.8
Dong et al.	2006	13.2 // +0.1	7.2 // -2.5	5.4 // +0.3	0.5 // +0.4	-	0.1 // +1.8
Paulot et al.	2007	8.4 // +5.1	3.0 // +1.9	4.8 // +0.9	-	-	0.6 // +2.3
Xu et al.	2008	-	2.7 // +2.0	3.1 // +2.8	0.6 // +0.3	-	-
EDGAR	2010	11.5 // +2.3	4.9 // -0.1	4.2 // +1.9	-	-	0.7 // +2.4
Xu et al.	2010	-	3.7 // +1.2	4.2 // +2.0	0.4 // +0.4	-	-
Fu et al.	2011	-	2.5 // +2.3	-	-	-	-
Kang et al.	2012	8.0 // +6.7	2.3 // +2.7	4.1 // +2.4	0.1 // +0.7	0.3 // -0.1	1.1 // +1.1

560 Note that before the "//" is the previous studies, after the "//" is the difference of our
561 result with previous studies, +xx or –yy in red/green colors in each column represents
562 higher or lower value in our study than previous research; "-" means unavailable data;
563 all the units of NH_3 emission have been converted to the Tg N yr^{-1}.
564 REAS [26]
565 Wang et al.[25]
566 Huang et al.[22]
567 Dong et al.[21]
568 Paulot et al.[16]
569 Xu et al.[35]
570 EDGAR [24]
571 Xu et al.[36]
572 Fu et al.[44]
573 Kang et al.[23]
574

Figure Legend

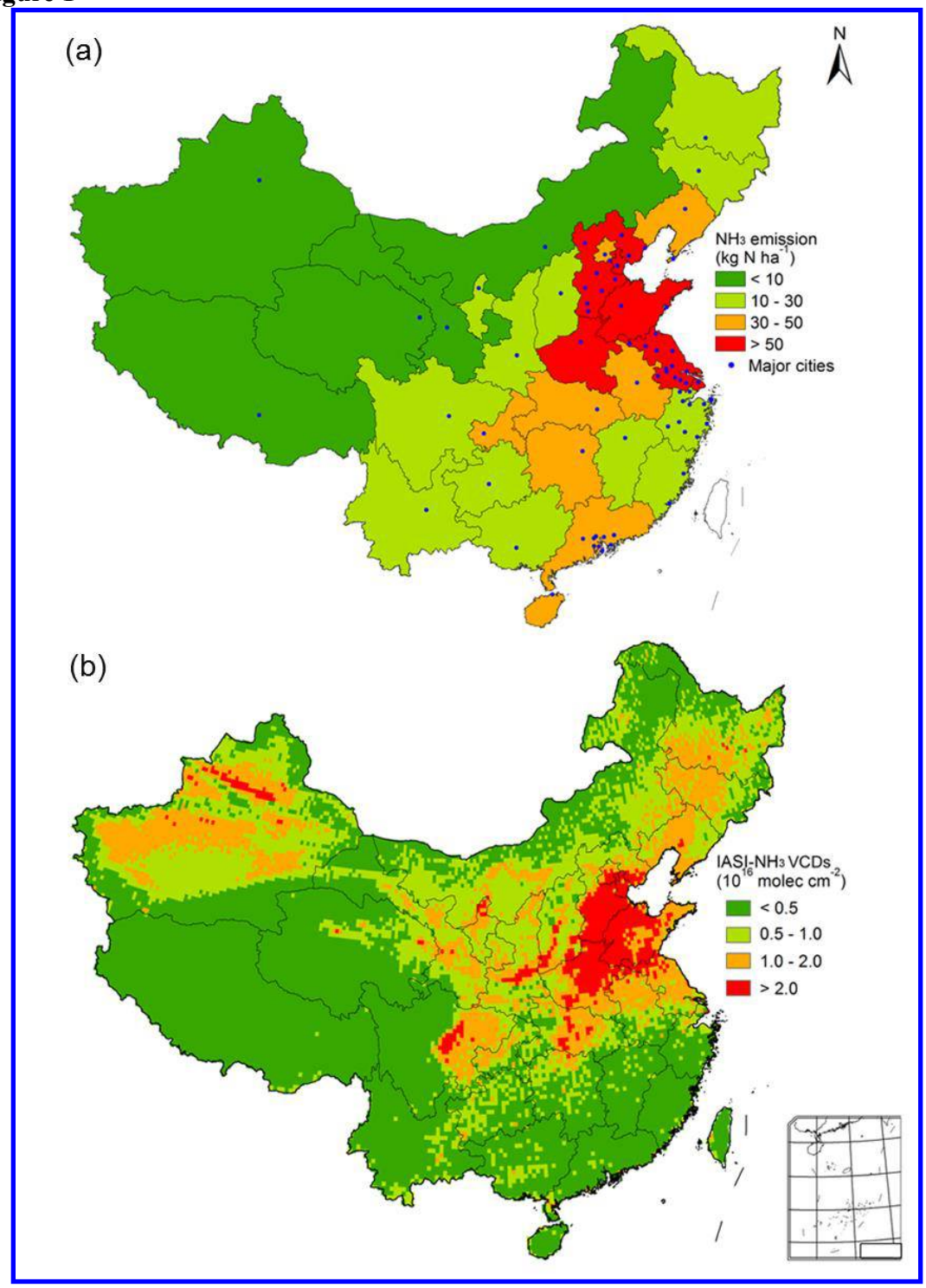
Figure 1 Validation by satellite observations (IASI) on spatial patterns. (a) The spatial patterns of NH_3 emission density in mainland China in 2015; (b) Mean IASI-NH_3 VCDs (10^{16} molec cm^{-2}) distribution for 2015 over China. Data of NH_3 VCDs are derived from an improved version of the IASI dataset presented in Whitburn et al.[40] and Van Damme et al.[53]

Figure 2 Comparison of NH_3 emissions with other published results and NH_x deposition in mainland China during 2000-2015. Studies addressing NH_3 emissions are colored with red symbols and NH_x deposition colored with green symbols (references presented in this figure are listed in the SI TextS4). The red dotted line represents the NH_3 emission estimated in this study, and the green dotted line represents the NH_x deposition synthesized by this study. The blue and grey shaded area indicates the 95% confidence interval of NH_3 emission evaluated by our study and NH_x deposition on land calculated using data provided by Liu et al.[17] and Xu et al.[6], respectively. Error bars of the symbols represent the uncertainties of their estimates, those symbols without error bars mean uncertainties unavailable or yet been discussed.

Figure 3 Comparison with SO_2&NO_x emission on temporal trends. The two solid lines show the molar ratios of $(2SO_2+NO_x)/NH_3$ and $2SO_2/NH_3$ in 2000-2015 in China. The dash line represents the molar ratio = 1. A ratio >1 represents NH_3 limitation to form the ammonium aerosol. While a ratio <1 represents NH_3 is available in abundance to form the ammonium aerosol contributing to Secondary Inorganic Aerosol formation (SIA), a substantial contributor to $PM_{2.5}$ concentrations. Data of SO_2, NO_x and NH_3 emission is based on MEPC,[54] Liu et al.[37] and our study, respectively.

Figure 4 Validation by satellite observations (AIRS) on temporal trends. The AIRS-NH_3 VCDs at 918 hPa from 2003 to 2015 are showed in pink dashed curves, with linear fits in solid lines. Data for AIRS-NH_3 VCDs was extracted from Warner et al.[33] and Van Damme et al.[53] The green dotted line represents the NH_3 emission estimated in this study.

608 **Figure 1**



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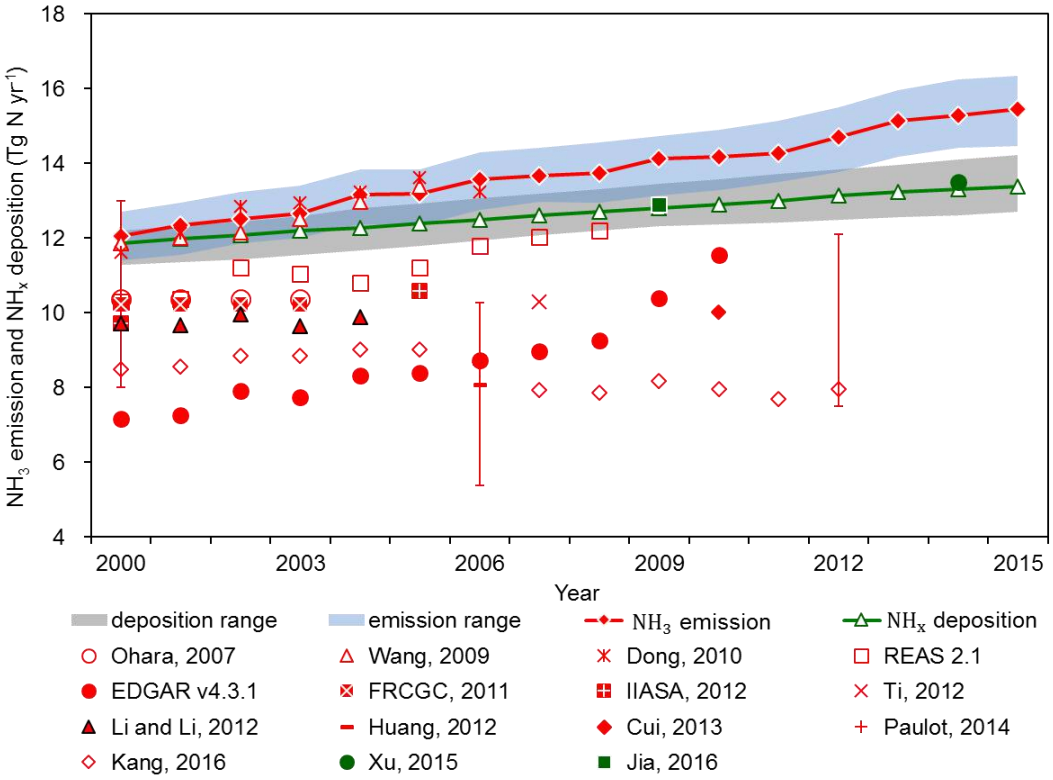
612 **Figure 2**



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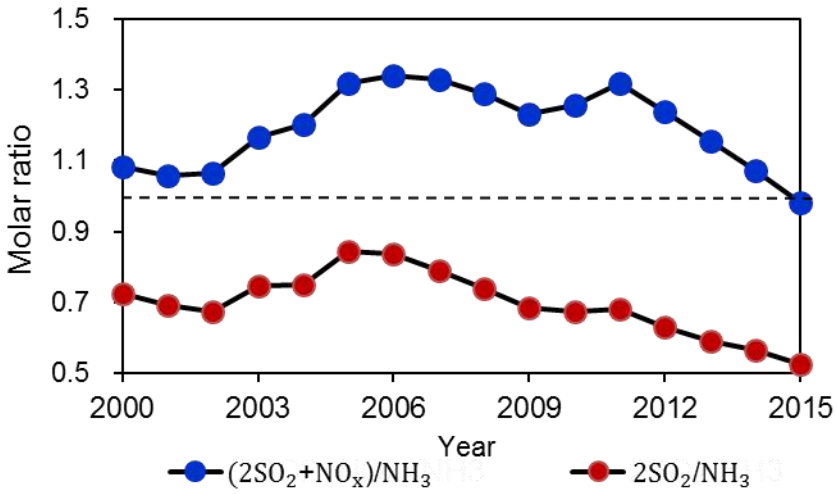
615 **Figure 3**



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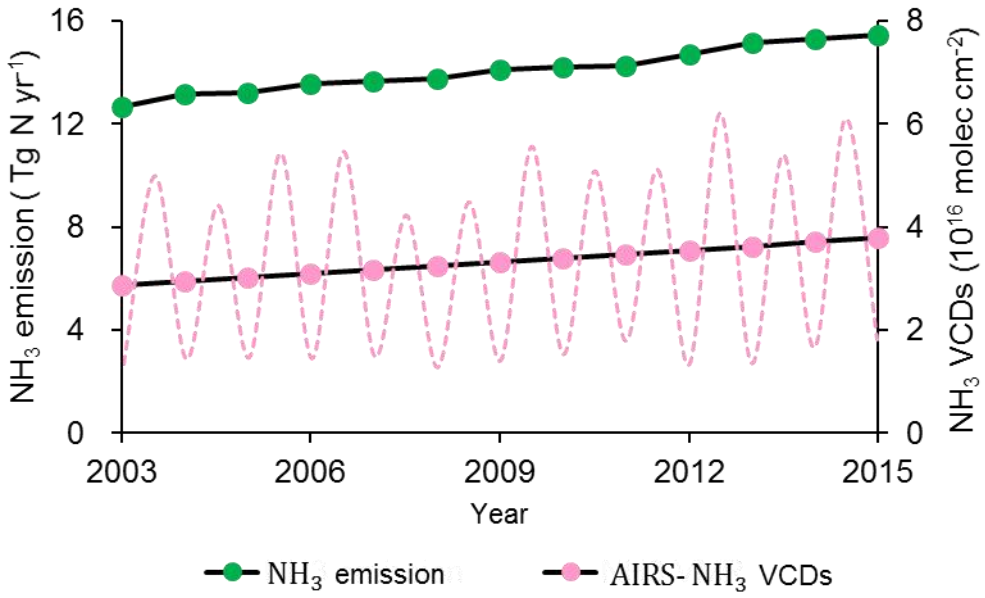
617

618 **Figure 4**



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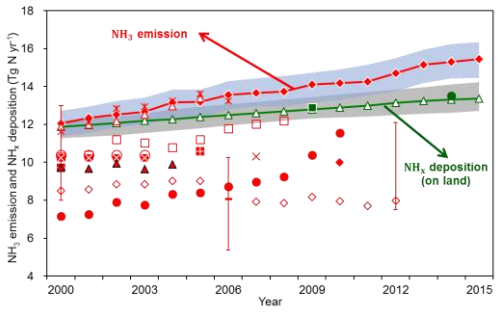
NH3 emission
NHx deposition (on land)
NH3 emission and NHx deposition (Tg N yr-1)
Year
18
16
14
12
10
8
6
4
2000
2003
2006
2009
2012
2015