



**Ammonia emissions
from agricultural
fertilizer in China**

P. Xu et al.

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High resolution inventory of re-estimating ammonia emissions from agricultural fertilizer in China from 1978 to 2008

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Abstract

The quantification of ammonia (NH_3) emissions is essential to the more accurate quantification of atmospheric nitrogen deposition, improved air quality and the assessment of ammonia-related agricultural policy and climate mitigation strategies. The quantity, geographic distribution and historical trends of these emissions remain largely uncertain. In this paper, a new Chinese agricultural fertilizer NH_3 (CAF_ NH_3) emissions inventory has been compiled that exhibits the following improvements: (1) a 1 km \times 1 km gridded map on the county level was developed for 2008, (2) a combined bottom-up and top-down method was used for the local correction of emission factors (EFs) and parameters, (3) the spatial and temporal patterns of historical time trends for 1978–2008 were estimated and the uncertainties were quantified for the inventories, and (4) a sensitivity test was performed in which a province-level disaggregated map was compared with CAF_ NH_3 emissions for 2008. The total CAF_ NH_3 emissions for 2008 were 8.4 Tg $\text{NH}_3 \text{ yr}^{-1}$ (a 6.6–9.8 Tg interquartile range). From 1978 to 2008, annual NH_3 emissions fluctuated with three peaks (1987, 1996 and 2005), and total emissions increased from 3.2 to 8.4 Tg at an annual rate of 3.0 %. During the study period, the contribution of livestock manure spreading increased from 37.0 to 45.5 % because of changing fertilization practices and the rapid increase in egg, milk and meat consumption. The average contribution of synthetic fertilizer, which has a positive effect on crop yields, was approximately 38.3 % (minimum: 33.4 %; maximum: 42.7 %). With rapid urbanization causing a decline in the rural population, the contribution of the rural excrement sector varied widely between 20.3 and 8.5 %. The average contributions of cake fertilizer and straw returning were approximately 3.8 and 4.5 %, respectively, thus small and stable. Collectively, the CAF_ NH_3 emissions reflect the nation's agricultural policy to a certain extent. An effective approach to decreasing $\text{PM}_{2.5}$ concentrations in China would be to simultaneously decrease NO_x , SO_2 and NH_3 emissions.

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1 Introduction

NH₃ is a colorless alkaline gas with high reactive ability and solubility in the atmosphere, where its presence has undesirable consequences. The gas reacts with HNO₃ and H₂SO₄ in the air to form ammonium salts (NH₄NO₃, (NH₄)₂SO₄ and (NH₄)HSO₄) (Pinder et al., 2007), which further contribute to visibility degradation and regional haze and have adverse health effects (Kim et al., 2006; Ye et al., 2011; Langridge et al., 2012). Such salts could account for 7.1–57 % of the total quantity of atmospheric fine particulate matter (PM_{2.5}: aerodynamic diameter of particle size ≤ 2.5 μm) (Yang et al., 2011; Huang et al., 2014; F. Zhang et al., 2014). NH₃ comprises nearly half of all reactive nitrogen released into the atmosphere and plays a key role in soil acidification, eutrophication and the disruption of ecosystems by dry deposition (Vanbreemen et al., 1984; Hellsten et al., 2008; Bouwman et al., 1997). In addition, although NH₃ exerts a cooling effect on the planet as a result of radiation forcing by aerosol particles (Martin et al., 2004), it is an indirect source of the major greenhouse gas nitrous oxide (IPCC, 2006) Therefore, efforts to decrease NH₃ emissions could have the triple benefit of slowing global climate change, decreasing regional air pollution and protecting human health (Zheng et al., 2012; Erisman et al., 2013).

Agriculture in China utilizes approximately 7 % of the world's cultivated land area to support 22 % of the global human population. To meet the demand for food of China's large and increasing population (30–50 % more food will be required over the next two decades) (Zhang et al., 2013; Cui et al., 2014; Ma et al., 2013), the government has initiated a series of agricultural policies that aim to increase the will of farmers, including farmers who overuse agricultural fertilizers (including synthetic and organic fertilizers), to increase yields. Policy-driven measures to increase the use of fertilizer and low nitrogen-use efficiency have resulted in continually increasing NH₃ emissions (Vitousek et al., 2009; Gu et al., 2012; W. Zhang et al., 2014). Therefore, to achieve food and environmental security, NH₃ emissions must be accurately estimated in a manner that reflects the spatial and temporal pattern of their sources.

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Previous studies mainly estimated NH_3 emissions in China based on EFs and activity. In the 1990s, China's NH_3 emissions were estimated based on uniform or overseas EFs for the entire country (Sun and Wang, 1997; Wang et al., 1997; Olivier et al., 1998; Xing and Zhu, 2000; Streets et al., 2003; Yan et al., 2003), which decreased the accuracy of these estimates because of the differences in regional environmental conditions. Subsequent studies, in which national or provincial statistical data on the rural population, fertilizers and agricultural production were used to estimate NH_3 emissions, generally downscaled to realize higher spatial resolution (e.g., Yamaji et al., 2004; Huang et al., 2012, and EDGAR v.4.2, 2013). Thus, biases could occur. Paulot et al. (2014) improved the bottom-up emission inventory to incorporate sector-resolved information on global agricultural activities known as MASAGE_ NH_3 , which is still limited to specific sectors (Paulot et al., 2014). Additionally, the previous emission inventories provide no or only coarse temporal distributions, which could result in underestimation during summer and overestimation during winter (Wang et al., 1997; Yan et al., 2003).

In this study, a $1 \text{ km} \times 1 \text{ km}$ gridded new CAF_ NH_3 emission inventory based on county-level activity data was developed for 2008 and historical time series of NH_3 emissions based on province-level activity data from 1978 to 2007. An effort was made to improve accuracy and decrease uncertainty by considering more comprehensive emission sources. In addition, a combined bottom-up and top-down method was used for the local correction of EFs and parameters. We analyzed the emission totals, source apportionment, spatial and temporal patterns and uncertainty and compared our results with the findings of previous studies. Then, we compared the 2008 emission map with a province-level disaggregated map. We also provided a clear description of the change in NH_3 emissions in CAF historical emissions between 1978 and 2008. Finally, the implications of the higher spatial and temporal resolution NH_3 emission inventory are discussed, with a focus on the control of N deposition, the improvement of air quality, NH_3 emission-related agricultural policy and climate mitigation strategies.

2 Methodology and data sources

2.1 NH₃ emission sources

Five NH₃ emission sources, including synthetic fertilizers (i.e., chemical and compound fertilizers) and organic fertilizers (i.e., rural excrement, livestock manure spreading, cake fertilizer and straw returning; sludge is not considered to be an agricultural fertilizer, and the quantity of green manure that is applied is limited) (Gao et al., 2011), are included in our emissions model. China's NH₃ emissions from agricultural fertilizer (E_{NH_3} , Tg NH₃ yr⁻¹) are calculated with the following equation:

$$E_{\text{NH}_3} = \sum_i A_{nij} \cdot \text{EF}_{ij} \cdot f(\text{RP}_{nijm}, C_m) \quad (1)$$

where subscripts n , i , j and m are the year, the emission source, the region (county in 2008 and province in 1978–2007) and the parameter, respectively; A is the activity data; EF is the region-specific emission factor (EF); RP_{nijm} represents a region-specific emission controlling parameter m for activity data or EF; $f(\bullet)$ represents a function whose shape depends on the source type; and C stands for a coefficient. More detailed descriptions of the equations used for each source (Sect. S1 in the Supplement), the activity data, the RPs, and the EFs (Sect. S2) can be found in Supplement and are briefly summarized below.

2.2 Data sources

In this paper, county-level data for the annual quantities of synthetic fertilizer (5), livestock types (8), crop types (17), and the 2008 rural population were derived for 2376 counties from 329 municipal statistical registers in Mainland China, Hong Kong, Macau and Taiwan. (There were no data for Hong Kong and Macau primarily because they have no agriculture). Annual above-province-level data for Mainland China from 1978 to 2008 were obtained from the China Agriculture Yearbook

(NBSC, 2009a) and the China Compendium of Statistics (NBSC, 2009b) Taiwanese data for 1991 to 2008 were collected from the Yearly Report of Taiwan's Agriculture (COA, 1992–2009) We obtained the 2008 unavailable fertilizer, crop, and livestock data for 332 counties in Mainland China. Taiwanese annual above-province-level data for 1978–1990 were based on temporal interpolation (Zhou et al., 2014). We disaggregated these activity data into 1 km × 1 km maps based on China's land use pattern to determine their spatial distribution. In addition, regarding activity data collected in international statistics databases for 2008, the synthetic fertilizers data were obtained from the IFA (<http://www.fertilizer.org/>), the rural population data were obtained from the World Bank (<http://data.worldbank.org/>) and the crop and livestock data were obtained from the FAO (<http://faostat.fao.org/>). The meteorological data for Mainland China were provided by the China Meteorological Data Sharing Service System (<http://cdc.nmic.cn/home.do>), and the Taiwanese meteorological data were provided by the Central Weather Bureau (<http://www.cwb.gov.tw>). Complementary gridded activity data include soil pH (1 km × 1 km) (the Harmonized World Soil Database v1.2, <http://web.archive.iiasa.ac.at/Research/LUC/External-World-soildatabase/HTML/>) and the secondary classification of land-use data (1 km × 1 km) (Liu et al., 2010b).

2.3 EFs for NH₃

The EFs reported in the literature are associated with large uncertainty because of differences in ambient temperatures, planting practices, soil properties, and other crucial influential factors. In this paper, for synthetic fertilizer sources, a previously developed EF correction model was used. Because direct measurements of EFs are limited in number, the EFs were adjusted for soil pH, fertilization method, application rate, precipitation and local temperature conditions to establish their spatial and temporal variations (i.e., the county or provincial level; monthly emissions) according to the top-down NASRSES model (Webb et al., 2006) (for detailed information, see Xu et al., 2015). For livestock manure spreading sources, the original NH₃ EFs compiled to develop the bottom-up RAINS model were found in the EEA's inventory guidebook (EEA, 2009)

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However, the values for region-specific N excretion in livestock manure management and the feeding days for livestock species/categories are based on published measurements for China and the results for the Livestock Manure Sector in the National Pollution Source Survey Database (NPSS) (Huang et al., 2012; MEP, 2008; SCC, 2013).

Although the age and growth stage of livestock are likely to cause a certain degree of variation in the quantity of fecal excretion, this effect is only reflected by specific parameters on the farm scale (Ross et al., 2002). The activity levels and the EFs of the national- or regional-scale emission inventories do not distinguish according to the factors included in this study because EFs are restricted by activity level, i.e., animal industry statistical data. In addition, although Chinese statistics can be approximate, theoretically, they can be refined, as a number of inventories that have previously considered these factors have attempted (Huang et al., 2012). However, Chinese statistics currently contribute little to emissions inventories because of a lack of functionality and practical significance. It was assumed that the number of livestock is same during each month of the year. The proportion of livestock EFs for different seasons, which were used to establish the annual livestock EFs, was derived from Huang et al. (2012), and Hutchings et al. (2001) reported that the EFs for the different seasons are equivalent (Huang et al., 2012; Hutchings et al., 2001). These principles were applied in this study. Regarding the remaining sources, a literature review was aimed at collecting relevant EFs, whereby the arithmetic mean of different experiments was used. Additionally, we performed only regional correction for activity data when calculating the NH_3 emission of cake fertilizers and straw returning. The emissions from these sources were equally divided into 12 months because of their smaller application and EFs. All of the EFs and parameters used in this inventory are listed in Tables S1 and S2 in the Supplement.

2.4 Uncertainty analysis

A Monte Carlo simulation that consisted of 10 000 calculations of the NH_3 emission inventory was run to characterize the uncertainty caused by the variations in the activity data, the EFs and related parameters. The coefficient of variation (CV) of each activity

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data is assumed to be equal to the absolute value of the average difference between a given dataset for China used in to determine CAF_NH₃ and a default global dataset (e.g., IFA, FAO, World Bank) for 2008. In addition, the CV of each activity data in 1978–2007 is assumed to be equal to the CVs of the 2008 data. The CV values for sugarcane, highland barley, alfalfa, peanuts, other oil crops, other beans and other tubers were set at 0.2 because they are absent from the global datasets. The CV values for the EFs and related parameters were based on values found in the literature (Wang et al., 2012; Huang et al., 2012; Zhou et al., 2014; Xu et al., 2015). For activity data, uniform distributions were assumed. Normal distributions were adopted for the EFs and other parameters. The precise CV values are summarized in Table S3. Medians and the R_{50} were aimed at estimating the emissions and representing the uncertainties.

3 Results

3.1 NH₃ emissions in China in 2008

The total NH₃ emissions (E_{total}) from CAF for 2008 were estimated as 8.4 Tg NH₃ yr⁻¹ (median, 6.6–9.8 Tg as R_{50}), and the contribution of synthetic fertilizer application, live-stock manure spreading, rural excrement, cake fertilizers and straw returning were 3.3, 3.8, 0.7, 0.3, and 0.3 Tg, respectively. Detailed information on the contribution of each source to E_{total} from CAF for 2008 is presented in Fig. 1. Regarding the synthetic fertilizer contribution, 1.9, 1.3, 0.02, 0.003, and 0.05 Tg could be attributed to ammonium bicarbonate (ABC), urea, ammonium nitrate (AN), ammonium sulfate (AS) and others, respectively. Among the various sources of livestock manure (Fig. S1 in the Supplement), cattle were the largest emitter (30.2%), followed by pigs (28.9%), poultry (26.2%) and dairy cattle (7.9%). Rural excrement (8.5%), cake fertilizer (3.2%) and straw returning (3.9%) were of less importance but non-negligible.

3.2 Spatial and temporal distribution

Figure 2 shows the 1 km × 1 km and county-level geographic distributions of NH₃ emissions in 2378 counties for 2008. The mean per-unit cultivated area NH₃ emission was 5.9 tNH₃ km⁻² yr⁻¹. Using county-level data to create this NH₃ emissions map reveals the strong spatial association of the emissions with the distribution of arable land. The average emission density over western, central and eastern China is 4.7, 6.4 and 6.5 tNH₃ km⁻² yr⁻¹, respectively. The three regions are defined in Fig. S2. Eastern China (36.7 % of China's cultivated area) was the largest contributor of NH₃ emissions and responsible for approximately 41.6 % of the total. In central China, synthetic fertilizer was the largest contributor (44.4 %). This contribution was substantially higher than that of western (34.0 %) or eastern (36.3 %) China. However, the contribution of livestock manure spreading (37.6 %) in central China was substantially less than in western (50.3 %) or eastern (50.2 %) China. In addition, high emission densities were presented in the North China Plain, the Northeast Plain, the Huaihe River Basin, the Lianghu Plain, the Sichuan Basin, the Tarim Basin and the Weihe Plain. Most of China's grain and livestock production is concentrated in these areas. We compared our results with the global NH₃ column distribution using satellite monitoring (Clarisse et al., 2009; Van Damme et al., 2014). A number of global hotspots resemble those of China, such as the Tarim basin, the North China Plain and western Heilongjiang province and Jilin province. However, the higher emission areas were not observed by satellite monitoring because of clouds, water vapor, the surface temperature, high SO₂ emissions (Kharol et al., 2013; Wang et al., 2013; Garcia et al., 2008), land surface variation and the retrieval methods of NH₃ total columns (Xu et al., 2015). High NH₃ emission densities were also found in western China, such as Tibet, Sichuan and Qinghai, where livestock is raised on a large scale and less cropland exists.

To test the sensitivity of the NH₃ emissions spatial patterns to input activity data, an emissions inventory (PRO-NH₃ (China)) was developed using the same methods that were employed to create CAF_NH₃ except county-level activity data for provin-

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cial disaggregation using regression models (Zhang et al., 2007; Zhou et al., 2014). The E_{total} of PRO-NH₃ is 7.3 TgNH₃yr⁻¹, which is 12.5% less than the CAF_NH₃ value. For a more detailed comparison, the relative difference was defined as $RD = (E_1 - E_2) / ((E_1 + E_2) / 2)$ (Wang et al., 2012), where E_1 and E_2 are the E_{total} for agricultural fertilizer of the counties for CAF_NH₃ and for PRO-NH₃ for each county, respectively. Figure 3 shows all counties' frequency and spatial distributions of the RDs. The spatial bias of the provincial disaggregation increases as the absolute RDs. A negative (positive) RD suggests an overestimation (underestimation) of a county's emissions by utilizing the provincial disaggregation approach (PRO-NH₃). The mean absolute RD was 48.7% for all counties. In 37% of the countries, the absolute RDs were found higher than 50%. In addition, the PRO-NH₃ emission pattern is lowly correlated with the CAF_NH₃ pattern ($R = 0.49$, $p < 0.01$). These results indicate that spatial bias can be substantially reduced using the county-level activity data and that provincial disaggregation using regression models cannot determine the county-scale structure of the spatial distribution of activity data within provinces. Large RDs were often observed in provinces and regions in which the development status significantly varies, such as Sichuan, Qinghai, Inner Mongolia and Tibet.

Figure 4 shows the monthly NH₃ emissions in 2008 from various sources, which are generally in agreement with the local climate, planting time and cultivation practices. Higher emissions occurred during the summer (June to August) and accounted for 39.7%. The peak value was found for July (1.2 TgNH₃yr⁻¹). This value was approximately 3.1 times larger than the smallest value (January). Regarding synthetic fertilizer sources, NH₃ emissions increased significantly in April, peaked in July, and then decreased. This pattern could be partly attributed to an increased application of synthetic fertilizer and higher temperatures. In China, winter wheat and oilseed rape are typically seeded in late September and early October with the base fertilizer application. The basal dressing and topdressing of summer maize occur in June and August. In addition, early rice sowing, late rice sowing and transplanting typically occur in April and July and are accompanied by base fertilization. For these crops, topdressing is performed in late

June and late September, respectively. A total of 50 ~ 80 % of the synthetic fertilizer is applied at or around planting (Zhang et al., 2011). The largest livestock manure spreading emissions also occurred in summer and accounted for nearly 28.8 % of livestock manure spreading emissions. This result might be explained by larger EFs related to the substantial increase in ambient temperature and little variation in the livestock population among the different months (Huang et al., 2012). The NH₃ emissions in winter (December to February) were lower due to the relatively lower temperature and infrequent agricultural activities. The spatial distributions of CAF_NH₃ for January, April, July and October are shown in Fig. S3. In western China, rural excrement's monthly contribution proportions were higher than in eastern China, particularly during winter (1.6 times); In central China, synthetic fertilizer's monthly contribution proportions began to exceed livestock manure in April (Fig. S4). However, this condition occurred in May in eastern and western China because of temperature variations. In addition, NH₃ emissions in central China (i.e., in Guangdong, Guangxi and Hainan) were typically more stable than in eastern China (i.e., in Jilin, Liaoning and Heilongjiang) because of less dramatic temperature fluctuations and less intensive agricultural activities.

3.3 Historical time trend for NH₃ emissions in China

Annual CAF_NH₃ emissions were estimated from the activity data, the EFs and related parameters as described in the Methodology section for 1978 to 2008. Figure 5 shows the annual variations in E_{total} and the distributions of each sector during the study years. The emissions increased from 3.2 to 8.4Tg (2.6 times) during the period 1978–2008. Fertilizer has been promoted as an important means to improve crop yields because the overall grain response to fertilizer and to technological and institutional changes is viewed as crucial to Chinese agricultural production (Wang et al., 1996). In addition because the initiation of reform and opening-up in 1978, the government has implemented a subsidy policy with respect to fertilizer and urged farmers to apply additional fertilizer to increase grain yields. From 1978 to 2008, E_{total} fluctuated, with peaks in 1987, 1996 and 2005. The NH₃ emissions growth rates in 1978–1987 and

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1988–1996 were 41.7 and 51.3 %, respectively, primarily as a result of the improvement of the unified food price in 1979 to increase the enthusiasm of farmers after the establishment of the Householder's Responsibility System. The system enhanced China's agricultural intensification degree and continuously increased the intensity of fertilizer use. In 1988, NH_3 emissions appeared to decrease. The primary reason for this decrease was the shortage and/or inflated price of supplies and equipment required for agricultural production, particularly the substantially higher prices of synthetic fertilizer and fodder, which increased agricultural production costs and thus affected production output. In addition, a severe drought occurred during the entire year, which seriously affected agricultural production (Ma and Zhao, 1989). In 1997–2005, the NH_3 emission growth rate was 17.4 %. This rate reflects a steady growth trend although one that was slower than that of the first two periods because following the grain yield peak in 1998 (512.3 Tg, CAY, 1980–2009) grain prices decreased as a result of oversupply. These events sharply reduced the enthusiasm of farmers engaged in agricultural production. In addition, with China's accession to the World Trade Organization in 2001, lower overseas grain prices restrained the increase of domestic grain prices, which encouraged the rural population transfer to non-agricultural production. The national government did not recognize the problem's severity until the end of 2003 and then issued a series of favorable and preferential agricultural policies, including the repeal of the agriculture tax. This policy approach reversed the damaging decline of crop yields. However, influenced by the Asian financial crisis and El Nino, in 1997, emissions decreased. Since 2005, grain yields have continuously increased, and the quantity of synthetic fertilizer applied has decreased overall as a result of the dissemination of new technologies for soil testing and fertilizer formulation. E_{total} first decreased and then increased. Natural disasters and livestock and poultry disease caused a marked decline in livestock breeding stock, which is the primary reason for the sudden decrease in NH_3 emissions in 2007. That is, livestock manure emissions decreased 18.8 % compared with 2006.

Regarding the contribution by each sector in China, the contribution of livestock manure spreading increased from 37.0 % in 1978 to 45.5 % in 2008 because fertilization

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practices changed from organic to inorganic fertilizer and then to a combination of these types. To encourage farmers to use more organic fertilizer and to spur the development of organic fertilizer resources across the country, in 1988, the Chinese State Council published “With respect to instruction of valuing and reinforcing organic fertilizer.” Subsequently, farmers realized that organic fertilizer could play a significant role in water conservation, soil fertilization and soil improvement. In addition, livestock and poultry breeding techniques were improved as a result of the rapid increase in egg, milk and meat consumption. The contribution of livestock manure spreading (from pigs, poultry and dairy cattle) has been increasing during the past 31 years, others have observed the opposite trend. However, the largest contributors are cattle and pigs (46.6 and 23.3 %, respectively, on average) (Table S4). The average contribution of synthetic fertilizer to E_{total} is approximately 38.3 % (minimum: 33.4 %; maximum: 42.7 %), also during the past 31 years. Generally, synthetic fertilizer application exhibits a strong positive correlation with crop yields ($R^2 = 0.89$) (Fig. S5). In addition, because of the growth effect of synthetic fertilizer in agriculture, the high demand for synthetic fertilizer will not change. Synthetic fertilizer application will continue to increase as the optimization of the domestic agricultural planting structure and the cash crop planting area increase. The contribution of the rural excrement sector substantially decreased from 20.3 % in 1978 to 8.5 % in 2008 as a result of the decline in China’s rural population that accompanied rapid urbanization. The contributions of cake fertilizer and straw returning were small and remained stable during the study period. Their average contributions were approximately 3.8 and 4.5 %, respectively. Collectively, these findings support the hypothesis that in addition to the limitation of climate conditions, agricultural production suffered as a consequence of the co-ordination and control of the country’s agricultural policy, which directly affected the NH_3 emissions of fertilizers used in agricultural production. That is, to a certain extent, the NH_3 emissions attributable to agricultural fertilizer reflect the country’s agricultural policy. Overall, our findings are in substantial qualitative agreement with the analysis of China’s fertilizer policies by Li et al. (2013).

4 Discussion

4.1 Comparison with previous NH₃ emissions inventories

Table 1 presents a comparison of the 2004–2008 NH₃ emission inventories for China of this study with other inventories that investigated the same emission sources. Our estimate is 22.8 % less than that of EDGAR v.4.2 (2013), 20.4 % less than that of Cao et al. (2010), and 43.1 % less than that of Dong et al. (2010). These differences primarily result from differences in synthetic fertilizer emissions. The previous estimates employed uniform EFs for the entire country, which were derived from foreign expert evaluations or European rather than local data. However, despite using corrected EFs (Zhang et al., 2011), our estimate is 23.3 % lower than that of Zhang et al. (2011). This difference can be partly attributed to the choice of parameters used in the EF corrections. Our estimate is completely based on local measurements, whereas the results of Zhang et al. (2011) were primarily based on measurements performed in Europe. Our estimate is 23.8 % higher than that of Huang et al. (2012), 14.5 % higher than that of Paulot et al. (2014), 19.2 % higher than that of Wang et al. (2009), and 7.7 % higher than that of Li et al. (2012). These differences are explained by the differences in base year and by the use of regional EFs as well as local and high-resolution activity data. The annual NH₃ emissions calculated in this study were compared with previous estimates (Wang et al., 2009; Dong et al., 2010), and the results are shown in Fig. 5a. A year-by-year comparison of the findings of EDGAR v.4.2 (2013) or Wang et al. (2009) with the findings of this study indicates that the growth trends compare well for 1980–2005. Our estimates for 1994 to 2006 are approximately 1.8 times higher than those of Dong et al. (2010) for each year. We compared our temporal distribution for synthetic fertilizer application to the findings of Paulot et al. (2014), Huang et al. (2012) and Zhang et al. (2011). Our estimates agree well with the other inventories for the monthly variation tendency. However, in our study and that of Zhang et al. (2011), emissions peaked in July, whereas in Huang et al. (2012), the emissions peaked in August. In all three studies, the maximum emission occurred during summer. This phe-

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nomenon could be primarily attributed to the local climate conditions, which affected the EFs for the base year. However, in Paulot et al. (2014), the emissions peaked in April because erroneous planting dates were used in the crop model such as the winter wheat-summer corn rotation, corn sown in June instead of April in China (Huang et al., 2012). This study assumes that 60 % of the synthetic fertilizer is used in planting, 20 % in growth and 20 % in harvest. Regarding livestock manure, our estimates are approximately 1.6 times larger than the monthly results of Huang et al. (2012) because of different base years, EF selection and differences in livestock population. In our study, emissions peaked in June–August, which was similar to the findings of Paulot et al. (2014), whereas the monthly emission in winter in our study was nearly 2.2-fold higher than in Paulot et al. (2014) because in the latter study the timing of livestock manure spreading is presumed to be identical with synthetic fertilizer application.

4.2 Impacts of NH₃ emissions on urban air pollution

A key research and policy question is how NH₃ emissions affect China's urban air pollution (in terms of PM_{2.5} and its precursors, e.g., NO_x, SO₂ and NH₃). Because China is a large agricultural country, CAF is the nation's largest emitter of NH₃. However, China is in the midst of an urban expansion necessary to becoming an economic superpower. The nation's urbanization rate rapidly increased from 17.9 % in 1978 to 52.6 % in 2008 (CAY, 1980–2009). In addition to urbanization, the difference between CAF_NH₃ emissions and NO_x and SO₂ emissions from fossil-fuel combustion is being effaced by the development of intensive agricultural and livestock production in marginal zones between rural and urban areas, which results in PM_{2.5} that exacerbates urban air quality because the pollutants react more easily (Gu et al., 2014). Additionally, the high PM_{2.5} levels of 2014 in China occurred in areas that overlap with agricultural areas (Fig. S6). Apparently, the CAF_NH₃ emissions cause urban air pollution by aerial transformation. Using a response surface modeling technique, Wang et al. (2011) revealed that approximately 50–60 % of the increases in NO₃⁻ and SO₄²⁻ aerosol concentrations were caused by the 90 % increase in NH₃ emissions from 1990–2005 in East China. Wang

et al. (2013) utilized GEOS-Chem to examine the impact of precursors of changes in anthropogenic emissions on the change in sulfate-nitrate-ammonium aerosols over China during 2000–2015, and found that the advantage of SO₂ reduction would be totally neutralized if NH₃ emissions increased by 16 % from 2006 to 2015, as anticipated based on China's recent growth rate. Therefore, to decrease PM_{2.5} concentrations and improve urban air quality in China, a more effective approach would be to simultaneously decrease NO_x, SO₂ and NH₃ emissions. However, the determination of the degree to which CAF_NH₃ contributes to the urban PM_{2.5} concentration is a topic for future research.

5 Implications for NH₃ emissions

Using local high-resolution data, spatially and temporally precise EF_{NH₃} and related parameters, in this study, times series of CAF_NH₃ emissions were developed. These time series enabled the creation of high-resolution maps of NH₃ emission densities, the source apportionment, and the spatial and temporal pattern for 2008 as well as a historical time trend analysis of total NH₃ emissions from 1978 to 2008. Additionally, we could distinguish NH₃ emissions hotspots and their spatial and temporal variations as well as identify the influence of national agricultural policy changes on NH₃ emissions because the initiation of reform and opening-up. Fortunately, the rate of NH₃ emissions during the last decade has increased slowly compared with 1978–1996. Because of their high volatility, urea and ABC have been gradually replaced by compound and organic fertilizers in the wake of the country attaching greater importance to the food security problem. Although an increasing portion of the rural population has moved to cities in the current period of rapid urbanization, a large rural population will continue to exist in the next decades (Wang et al., 2012). Agricultural fertilizer will continue to be required to meet the increasing demand for food, and fertilizer application technology will slowly improve (Sutton et al., 2011). To decrease CAF_NH₃ emissions, it is necessary to enhance the efficient use of agricultural fertilizer, reduce the intensity of agricultural

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fertilizer use, improve environmental factors and accelerate abatement strategy development. Liu et al. (2013, 2010a) noted that the accuracy as well as the temporal and spatial resolution of CAF_NH₃ inventories is essential to better quantify atmospheric N deposition and more accurately assess nitrogen flows in cropland (Liu et al., 2013, 2010a). It has been demonstrated that a dependable data-driven approach and local experiments or process-based models can substantially help increase the spatial and temporal resolution and decrease the uncertainties of emissions inventories.

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doi:10.5194/acpd-15-25299-2015-supplement.**

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Table 1. Total NH₃ Emissions (TgNH₃ yr⁻¹); source profile by sector after 2004 and their comparisons with previous studies; Note: SF = Synthetic Fertilizer application; LS = Livestock manure Spreading; RE = Rural Excrement; CF = Cake Fertilizer; SR = Straw Returning. Bold represents the sum of LS and RE.

References	Base year	EFs	SF	LS	RE	CF	SR	total
This study	2008	Correction EFs	3.3	3.8	0.7	0.3	0.3	8.4
Paulot et al. (2014)	2005–2008	Region-specific EFs	3.6	2.6				6.2
EDGAR (2013)	2008	IPCC	8.1	1.1				9.2
Cao et al. (2010)	2007	EEA	3.6	6.2				9.8
Huang et al. (2012)	2006	Correction EFs	3.2	2.4	0.2	0.3	0.3	6.4
Dong et al. (2010)	2006	EEA	8.7	4.3	0.7			13.7
Wang et al. (2009)	2005	Region-specific EFs	3.5	2.8				6.3
Zhang et al. (2011)	2005	Correction EFs	4.3					4.3
Li and Li (2012)	2004	Region-specific EFs	1.8	3.7	1.7			7.2

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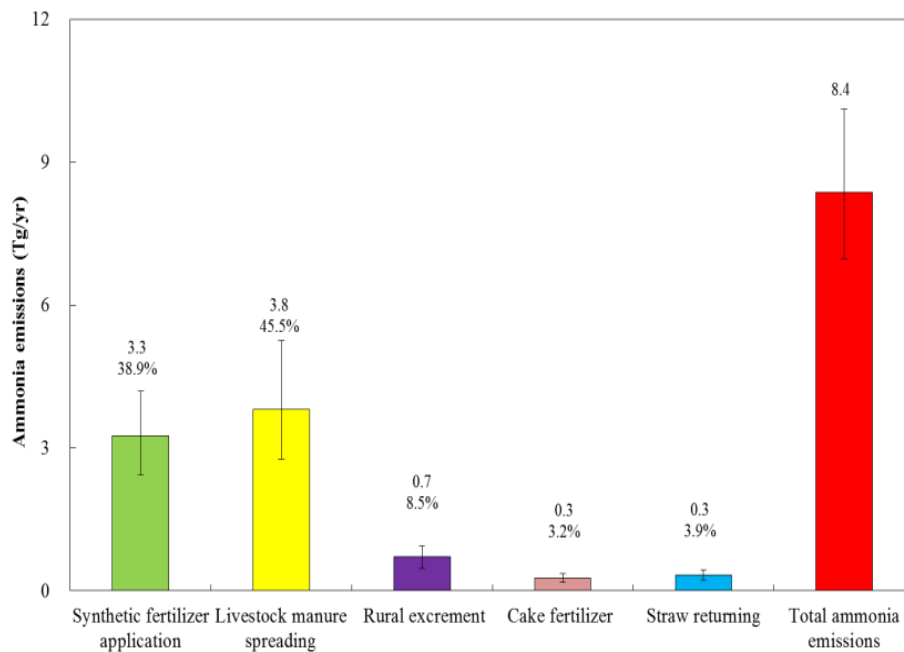


Figure 1. NH_3 emissions from CAF for 2008 by source, and the associated uncertainties.

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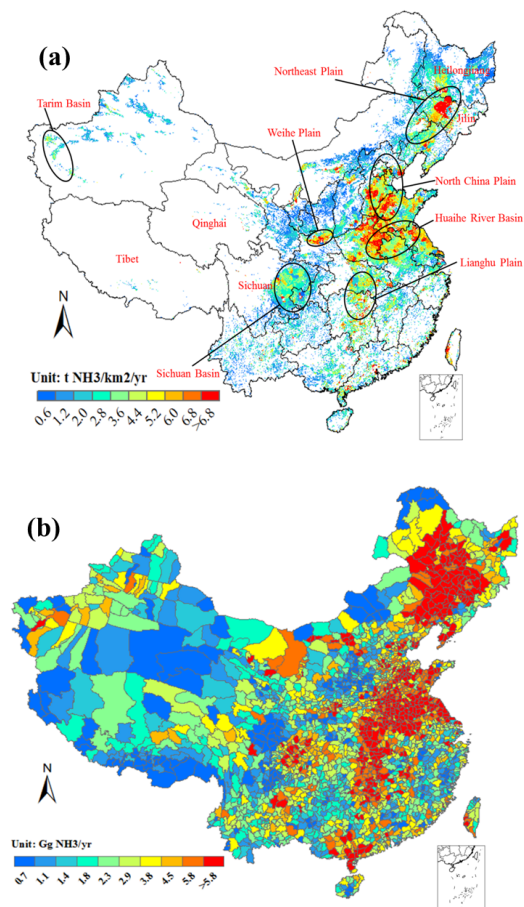


Figure 2. NH_3 emission map of China's agricultural fertilizer at $1 \text{ km} \times 1 \text{ km}$ (a) and the county level (b) for 2008. Major emission areas are circled.

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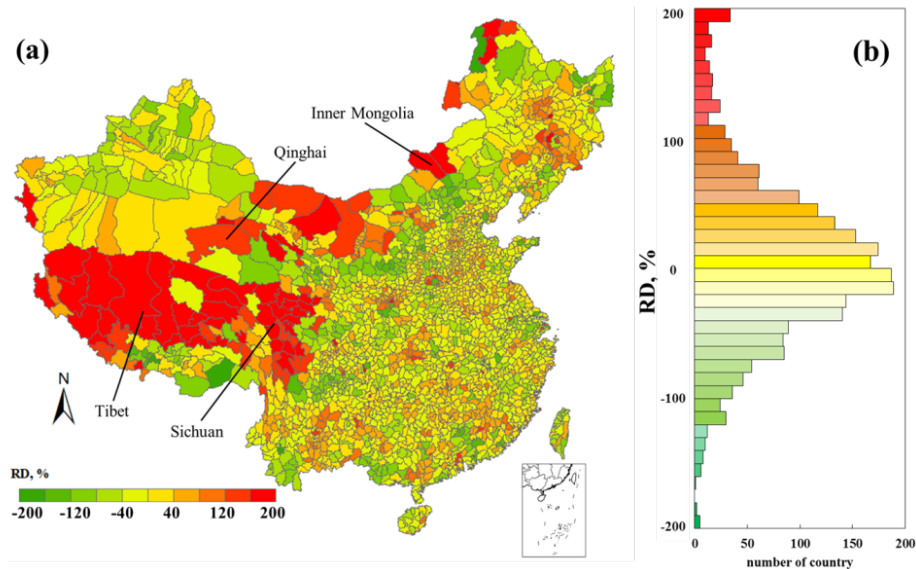


Figure 3. Geographic (a) and frequency (b) distributions of RDs of total NH_3 emissions in 2008 between CAF-NH_3 and PRO-NH_3 .

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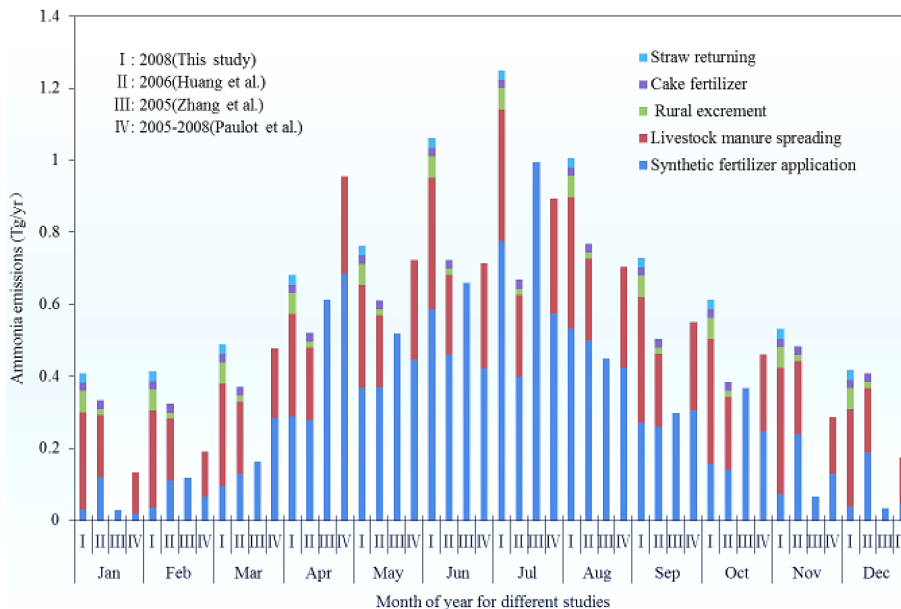


Figure 4. Monthly NH₃ emissions in 2008 and compared with earlier studies.

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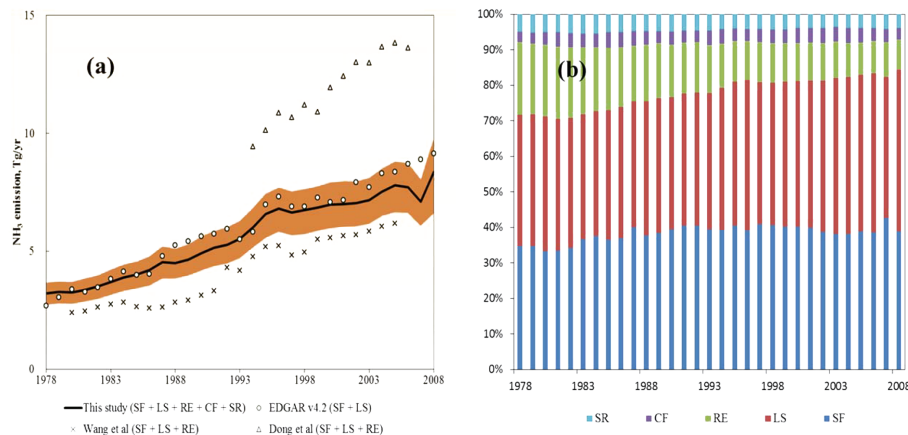


Figure 5. Temporal trend for agricultural fertilizer NH₃ emissions for China **(a)** and sector contributions between 1978 and 2008 **(b)**. The emission estimate and the uncertainty are provided as a median value (black curve) and the R50 (shaded area, for total emissions) derived from a Monte Carlo simulation. Note: SF = Synthetic Fertilizer application; LS = Livestock manure Spreading; RE = Rural Excrement; CF = Cake Fertilizer; SR = Straw Returning.

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