Atmos. Chem. Phys. Discuss., 15, 26959–26995, 2015
www.atmos-chem-phys-discuss.net/15/26959/2015/
doi:10.5194/acpd-15-26959-2015


Atmospheric Chemistry and Physics Discussions
Open Access



# High-resolution ammonia emissions inventories in China from 1980–2012

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Received: 7 August 2015 – Accepted: 10 September 2015 – Published: 6 October 2015

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Published by Copernicus Publications on behalf of the European Geosciences Union.

## Abstract

Ammonia ($NH_3$) can interact in the atmosphere with other trace chemical species, which can lead to detrimental environmental consequences, such as the formation of fine particulates and ultimately global climate change. China is a major agricultural country, and livestock numbers and nitrogen fertilizer use have increased drastically since 1978, following the rapid economic and industrial development experienced by the country. In this study, comprehensive $NH_3$ emissions inventories were compiled for China for 1980–2012. In a previous study, we parameterized emissions factors (EFs) considering ambient temperature, soil acidity, and the method and rate of fertilizer application. In this study, we refined these EFs by adding the effects of wind speed and new data from field experiments of $NH_3$ flux in cropland in northern China. We found that total $NH_3$ emissions in China increased from 5.9 to 11.2 Tg from 1980 to 1996, and then decreased to 9.5 Tg in 2012. The two major contributors were livestock manure and synthetic fertilizer application, which contributed 80–90 % of the total emissions. Emissions from livestock manure rose from 2.87 Tg (1980) to 6.17 Tg (2005), and then decreased to 5.0 Tg (2012); beef cattle were the largest source followed by laying hens and pigs. The remarkable downward trend in livestock emissions that occurred in 2007 was attributed to a decrease in the numbers of various livestock animals, including beef cattle, goats, and sheep. Meanwhile, emissions from synthetic fertilizer ranged from 2.1 Tg (1980) to 4.7 Tg (1996), and then declined to 2.8 Tg (2012). Urea and ammonium bicarbonate (ABC) dominated this category of emissions, and a decline in ABC application led to the decrease in emissions that took place from the mid-1990s onwards. High emissions were concentrated in eastern and southwestern China. Seasonally, peak $NH_3$ emissions occurred in spring and summer. The inventories had a monthly temporal resolution and a spatial resolution of 1000 m, and thus are suitable for global and regional air-quality modeling.





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## 1 Introduction

Ammonia ($NH_3$) is an important reactive nitrogen (N) compound, and has wide impacts on both atmospheric chemistry and ecosystems. As an alkaline gas in the atmosphere, it can readily neutralize both sulfate and nitric acid to form ammonium sulfate and ammonium nitrate, which are the major constituents of secondary inorganic aerosols (Behera and Sharma, 2012). Kirkby et al. (2011) found that atmospheric $NH_3$ could substantially accelerate the nucleation of sulfuric acid particles, thereby contributing to the formation of cloud condensation nuclei. The total mass of secondary ammonium salts accounts for 25–60 % of particulate matter less than or equal to 2.5 µm in aerodynamic diameter ($PM_{2.5}$) (Ianniello et al., 2011; He et al., 2001; Fang et al., 2009), and large amounts of this fine PM not only cause air pollution but also have a significant effect on radiative forcing (Charlson et al., 1992; Martin et al., 2004). In addition, the sinking of $NH_3$ into terrestrial and aquatic ecosystems can directly or indirectly cause severe environmental issues, such as soil acidification, eutrophication of water bodies, and even a decrease in biological diversity (Matson et al., 2002; Pearson and Stewart, 1993). When deposited into soils, $NH_3$ compounds can be converted into nitrate ($NO_3^-$) through nitrification, simultaneously releasing protons into the soil, resulting in soil acidification (Krupa, 2003).

Livestock manure and synthetic fertilizer represent the most important sources of $NH_3$ emissions, jointly accounting for more than 57 % of global emissions and more than 80 % of total emissions in Asia (Bouwman et al., 1997; Streets et al., 2003; Zhao and Wang, 1994). Previous studies have verified that China emits a considerable proportion of the total global $NH_3$ emissions budget due to its intensive agricultural activities (Streets et al., 2003). A major agricultural country, China has undergone rapid industrialization and urbanization since the Chinese government implemented its economic reform in 1978. The rapid economic development and rise in living standards over the last 30 years has resulted in a sharp increase in grain output and meat production. The use of synthetic fertilizers, which are applied by Chinese farmers to promote





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the growth of crops, has also undergone a considerable, sustained increase. According to figures from the International Fertilizer Industry Association (Zhang et al., 2012), synthetic fertilizer production has increased 3-fold during the past three decades, from 10 million t in 1980 to 43 million t in 2012. Several factors have contributed to the dramatic rise in the use of synthetic fertilizers. First, their use grew dramatically in the latter half of the 20th century in most parts of the world, as farmers increasingly expected to achieve higher crop yields. Second, N over-fertilization has been common, resulting in higher $NH_3$ volatilization loss, especially in the North China Plain and Taihu region (Xiong et al., 2008; Ju et al., 2009). In addition, due to farmers' increasing labor costs and income from off-farm activities, traditional farmyard manure has been gradually eliminated in much of China and replaced by synthetic fertilizers (Ma et al., 2009; Zhang et al., 2011). As a consequence, the surge in $NH_3$ emissions from synthetic fertilizer application during this period has been inevitable. Meanwhile, since 1980, when China began developing a series of policies to support livestock production, the industry has undergone rapid growth driven by the increasing demand for beef, pork, mutton, milk, and wool (Zhou et al., 2007). For example, in 2006, there were 56 million head of slaughtered cattle in China, representing a 16-fold increase from the number in 1980; the number of poultry in production increased 12-fold during this same period (EOCAIY, 2007). The flourishing livestock industry has produced large volumes of manure that releases gaseous $NH_3$ through N hydrolyzation and volatilization. In conclusion, a marked increase in $NH_3$ emissions from livestock manure and synthetic fertilizer are expected from 1980 to the present, but specific data on annual emissions and variation in emissions are lacking.

Changes induced by anthropogenic activities can significantly influence the global N cycle (Vitousek et al., 1997). Therefore, to better understand the evolution of the global N budget and the impacts on living systems, it is essential to quantify $NH_3$ emissions during recent decades in China. Moreover, the compilation of multi-year regional and national $NH_3$ emissions inventories would also help elucidate the causes of severe air pollution in China.





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In a previous study, we developed a comprehensive $NH_3$ inventory for 2006 to show the monthly variation and spatial distribution of $NH_3$ emissions in China based on a bottom-up method (Huang et al., 2012). Our method had several advantages over previous inventories. First, emissions factors (EFs) characterized by ambient temperature, soil acidity, and other crucial influences based on typical local agricultural practices were used to parameterize $NH_3$ volatilization from synthetic fertilizer and animal manure. In addition, we included as many different types of emission sources as possible, such as vehicle exhaust and waste disposal. Our $NH_3$ emissions inventory was compared with some recent studies to show its reliability. Paulot et al. (2014) used the adjoint of a global chemical transport model (GEOS-Chem) to optimize $NH_3$ emissions estimation in China; the results were similar to our previous study (Huang et al., 2012). In addition, the distribution of the total $NH_3$ column in eastern Asia retrieved from measurements of the Infrared Atmospheric Sounding Interferometer (IASI) aboard the European METeorological OPerational (MetOp) polar orbiting satellites (Van Damme et al., 2014) was also in agreement with the spatial pattern of $NH_3$ emissions calculated in our previous study. Hence, our bottom-up emissions inventory appears to be reliable, and the method can be used to estimate $NH_3$ emissions in China.

In this study, we made further improvements based on this bottom-up method and considered the following sources of $NH_3$: (1) farmland ecosystems (synthetic fertilizer application, soil and N fixing, and crop residue compost); (2) livestock waste; (3) biomass burning (forest and grassland fires, crop residue burning, and fuelwood combustion); and (4) other sources (excrement waste from rural populations, the chemical industry, waste disposal, $NH_3$ escape from thermal power plants, and traffic sources). The interannual variation and spatial patterns of $NH_3$ emissions from 1980 to 2012 on the Chinese mainland (excluding Hong Kong, Macao, and Taiwan) are discussed in this paper.

## 2 Methods and data

$NH_3$ emissions were calculated as a product of the activity data and corresponding condition-specific EFs, according to the following equation:

$$E(NH_3) = \sum_i \sum_p \sum_m (A_{i,p,m} \times EF_{i,p,m}) \,, \quad (1)$$

where $E(NH_3)$ is the total $NH_3$ emissions; $i$, $p$, and $m$ represent the source type, the province in China, and the month, respectively; $A_{i,p,m}$ is the activity data of a specific condition; and $EF_{i,p,m}$ is the corresponding EF. The emissions were allocated to a 1 km × 1 km spatial resolution on the basis of land cover, rural population, and other proxies. Further details on the estimation methods and gridded allocation of the various sources are presented in Huang et al. (2012).

### 2.1 Synthetic fertilizer application

$NH_3$ volatilization from synthetic fertilizers represents an important pathway of N release from the soil, resulting in large losses of soil and plant N (Harrison and Webb, 2001). We classified the synthetic fertilizers used in Chinese agriculture as urea, ammonium bicarbonate (ABC), ammonium nitrate (AN), ammonium sulfate (AS), and others (including calcium ammonium nitrate, ammonium chloride, and ammonium phosphates). $NH_3$ emissions were estimated by multiplying gridded (1 km × 1 km) EFs for five types of fertilizer by synthetic fertilizer use, which was calculated as the product of cultivated area and the application rate to crops (EOCAY, 1981–2013; Zhang et al., 2012; NBSC, 2003–2013b). In our previous inventory, we introduced the type of fertilizer, soil pH, ambient temperature, fertilization method, and application rate as parameters to develop gridded EFs for specific conditions. In the present study, the effects of wind speed and recent results from field experiments of $NH_3$ flux in cropland were added to further refine the EFs used to estimate $NH_3$ emissions from synthetic fertilizers, which is described below.

### 2.1.1 Wind speed

In addition to temperature, wind speed is a meteorological parameter that affects the partial pressure of $NH_3$ by regulating the exchange of $NH_3$ between the soil/floodwater and the air, thereby influencing $NH_3$ volatilization (Bouwman et al., 2002). Several previous studies have shown that high winds significantly influence $NH_3$ volatilization (Denmead et al., 1982; Fillery et al., 1984; Freney et al., 1985). In this study, we followed the approach of Gyldenkaerne et al. (2005) to introduce the effects of wind speed on $NH_3$ volatilization from synthetic fertilizer application. The original EFs were multiplied by a factor that was an exponential function of wind speed. The monthly average wind speeds for a 1 km × 1 km grid were based on the final analysis dataset of the National Centers for Environmental Prediction (NCEP).

### 2.1.2 Improvement of the EF for soil pH

Huang et al. (2012) used the measurements of Cai et al. (1986) and Zhu et al. (1989) to develop a linear relationship between $NH_3$ volatilization and soil pH that was applied to correct the EFs for soil acidity in different regions of China. To refine the correction so that it was nationally representative, we referred to recent results from field experiments to further adjust this relationship. Huo et al. (2015) conducted fertilization experiments in a representative area of farmland in the North China Plain in Hebei Province from 25 March to 6 May 2012, and measured a soil pH and average air temperature of 8.2 and 15 °C, respectively. The results yielded an $NH_3$ EF for urea of 12 % ± 3 %. We combined this condition-specific EF with that from Cai et al. (1986) and Zhu et al. (1989) to refine the correction for soil pH.

## 2.2 Livestock waste

A mass-flow approach has been widely used to estimate $NH_3$ emissions from livestock waste (Beusen et al., 2008; Velthof et al., 2012). Livestock waste can produce





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ammoniacal N (TAN), which can be converted into gaseous $NH_3$ or lost through other pathways during manure management (Webb and Misselbrook, 2004; Webb et al., 2006). We estimated livestock emissions by multiplying TAN at four different stages of manure management: outdoor, housing, manure storage, and manure spreading onto farmland (Pain et al., 1998) with the corresponding EFs. TAN is the product of the daily amount of manure produced (kg day$^{-1}$ capita$^{-1}$), N content (%), and TAN content (%) (Table S1), all of the parameters for which were assumed to have not changed during the 30 year period. In addition, we also considered three main animal-rearing systems in China: free-range, intensive, and grazing. The first two systems are extensively implemented in most rural areas of the country. The free-range system is characterized by small-scale rearing belonging to individual families and has been rapidly developed over recent decades (http://www.caaa.cn/). Based on animal husbandry yearbooks, we defined an intensive rearing system as that where the number of a single livestock class on a single farm was larger than a certain value (Table S2).

The number of livestock in each class from 1980 to 2012 was provided by official statistical data and husbandry industry reports (EOCAIY, 1999–2013; EOCAY, 1981–2013). We applied specific EFs in different situations and took the ambient temperature from the NCEP final analysis dataset into account in the stage in which the manure was stored indoors.

## 2.3 Other sources

The other minor $NH_3$ emission sources included agricultural soil, N-fixing plants, the compost of crop residues, biomass burning, excrement waste from rural populations, the chemical industry, waste disposal, traffic sources, and $NH_3$ escape from thermal power plants. The data sources and EFs for each source type are summarized in Table 1. Further details on the estimation methods appear in Huang et al. (2012). $NH_3$ escape, which was not included in previous inventories, is a new source of $NH_3$ emissions that has emerged during the past 10 years in China, and refers to the $NH_3$ derived from the incomplete reactions of $NH_3$ additives used in $NO_x$ abatement in thermal





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power plants (EEA, 2013). We roughly estimated the amount of $NH_3$ escape by multiplying the total flue gas released in power plants nationwide by the maximum allowable concentration of $NH_3$ carried in flue gas (NEA, 2011; CAEPI, 2013).

## 3 Results and discussion

### 3.1 Annual $NH_3$ emissions

Over the past 30 years, China has undergone dramatic changes and significant economic development, and the annual variation in $NH_3$ emissions has changed correspondingly. Figure 1 illustrates the trends in total $NH_3$ emissions, which are divided into fertilizer application, livestock waste, and other minor sources. Total emissions increased from 5.9 to 11.2 Tg between 1980 and 1996, then decreased to 9.5 Tg in 2012. The most important contributor was livestock manure management, accounting for approximately 50 % of the total budget. Due to the extremely high consumption and high volatility of ABC and urea, synthetic fertilizer application was responsible for 30–43 % of the total emissions, second only to livestock manure. However, in Europe and the United States, where less-volatile synthetic fertilizers such as AN and AS are more popular (Bouwman and VanderHoek, 1997), livestock manure overwhelmingly dominate the $NH_3$ emissions inventory (Ferm, 1998). These two primary sources combined accounted for 80–90 % of the total emissions budget, with other minor sources accordingly accounting for 10–20 %.

#### 3.1.1 Emissions from livestock waste

Livestock waste was the largest source of $NH_3$ emissions in China from 1980 to 2012, contributing approximately 50 % of total emissions each year. Since the 1980s, rapid economic development in China has driven the large increment of livestock production. The total number of the major livestock animals, namely, beef cattle, sheep, pigs, and poultry, increased from approximately 70 to 140 million, 180 to 370 million, 420 to 1400





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million, and 0.9 to 10 billion respectively, from the 1980s to mid-2000s (Fig. S1 in the Supplement). Correspondingly, $NH_3$ emissions increased from 2.9 Tg in 1980 to 6.2 Tg in 2005, more than doubling during this period, and then decreased to 5.0 Tg in 2012 (Fig. 2). We divided livestock $NH_3$ emissions from 1980 to 2012 into four phases. In the first phase (1980–1990), emissions steadily increased with a mean growth rate of approximately 3 %. Free-range production contributed most of the emissions (the population of free-range animals represented more than 90 % of the major livestock animals; EOCAY, 1991). The second phase (1991–1996) saw the most rapid increase in emissions, and the growth rate rose to 10 % between the years 1994 and 1995. In 1992, China began to implement a reform of the socialist market economic system, which had previously driven livestock production (CAAA, 2009), and accordingly, more $NH_3$ emissions from livestock waste were emitted. However, in 1997, there was a 0.5 Tg decrease in livestock emissions, compared to the those in 1996. This observed decline could be attributed to the Asian financial crisis, which started in 1997 and may have a detrimental effect on the development of the Chinese livestock industry. From 1998 to 2005, as the third phase, $NH_3$ emissions continually rose in conjunction with an increase in livestock production due to improvements in cultivation technique and increases in market demand (Zhang et al., 2003). Compared to the first two phases, the contribution of intensive rearing systems to total emissions also increased, with the population of intensively reared animals representing nearly 20 % of the major livestock animals (EOCAIY, 2006), because the Chinese government encouraged large-scale intensive methods for livestock production to gradually replace traditional free-range systems (CAAA, 2009). After a peak in 2005, there was a marked decrease, and emissions fluctuated around 5.0 Tg in the fourth phase, significantly lower than those in the mid-2000s, which can be explained by a decrease in several major livestock classes, including cattle and sheep (Fig. S1). Multiple factors inhibited the development of the livestock industry, and thus reduced $NH_3$ emissions from 2007 to 2012. These included a rural labor shortage, increased feeding costs for farmers, and market price fluctuations of meat products (Pu et al., 2008). Moreover, it should be noted that the class-





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specific proportions of intensively reared animals for beef cattle, pigs, and laying hens significantly increased to approximately 30, 40 and 70 % in 2012, respectively, which partly accounted for the reduced livestock emissions due to the lower $NH_3$ EFs of the intensive system compared to the free-range (EEA, 2013). In contrast to the free-range and intensive systems, in recent decades $NH_3$ emissions from grazing systems have demonstrated slight growth, from 0.13 Tg (1980) to 0.20 Tg (2012), without significant changes.

Table S3 presents the interannual emissions of the typical livestock categories. Among them, beef cattle were consistently the largest $NH_3$ emitter, contributing an annual mean 1.9 Tg $NH_3$; pigs, laying hens, goats, and sheep were also major contributors to the total emissions in this period. Poultry had the most rapid growth rate in $NH_3$ emissions. Nevertheless, a marked downward trend has appeared since 2007 for several major $NH_3$ sources, including beef cattle, goats, and sheep, which led to the decrease in total emissions discussed above.

#### 3.1.2 Emissions from synthetic fertilizer application

Figure 3 shows the estimations of $NH_3$ emissions from synthetic fertilizer application for the period 1980–2012. Annual levels consistently increased from 1980 (2.1 Tg) to 1996 (4.7 Tg), and then declined from 1996 to 2012 (2.8 Tg). ABC and urea were the major sources; $NH_3$ release from other synthetic fertilizers, such as AS and AN, made a negligible contribution to emissions (< 0.1 %). In general, the interannual variation in emissions reflects the changes in farming practices in China. First, the relative contribution of urea and ABC has gradually changed over recent decades. During the 1980s, ABC represented a substantial fraction of synthetic fertilizers used in China, and because of its high volatilization (Zhu et al., 1989), emissions from this kind of chemical fertilizer dominated in this period. However, ABC was inefficient for crop production because of the low N content (17 % N) and high N loss. In the mid-1990s, China introduced the technology of urea production, which promoted its widely application (Zhang et al., 2012). Urea, characterized by high N concentration (46 % N), has gradually re-



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placed ABC and become the dominant chemical fertilizer used in cropland over the last 20 years. In 1980, 3.0 million and 5.1 million t of urea and ABC, respectively, were produced; by 2012, these values had changed to approximately 28.8 million and 3.4 million t, accounting for approximately 66.7 and 7.9 % of total synthetic fertilizer production in China, respectively (Fig. S2). Because $NH_3$ volatilization from ABC is more than 2-fold that from urea (Roelcke et al., 2002; Cai et al., 1986), the increasing proportion of urea application relative to that of ABC has caused the decrease in total synthetic fertilizer emissions observed from the mid-1990s onwards. As shown in Fig. 3, although emissions from urea application increased by 1.0 Tg from 1996 to 2012, those from ABC fell by nearly 3.0 Tg. In addition, there have been seasonal disparities in $NH_3$ emissions from synthetic fertilizers, caused by variation in both the temperature and timing of fertilizer application for different crops. Generally, $NH_3$ volatilization began to rise in April with increasing temperatures, and the highest emissions occurred in summer (June–August), which can be attributed to the high temperatures and intensive application of fertilizer. The seasonal distribution of synthetic fertilizer emissions was almost constant from 1980 to 2012, corresponding to stability in the seasonal distribution of agricultural activities.

### 3.1.3 Source apportionments

Table 2 lists $NH_3$ emissions at the national level from 1980 to 2012 from various sources. $NH_3$ emissions released by biomass burning generally increased, of which crop-residue burning and housing fuelwood combustion jointly accounted for a large proportion of the biomass burning emissions. In particular, a large forest fire occurring in 1987 in the Greater Khingan Mountains, located in Heilongjiang Province, released more than 40 Gg $NH_3$. $NH_3$ escape derived from the denitrification process in thermal power plants increased substantially, as the implementation of flue gas denitrification has rapidly increased in the past 10 years, especially in 2012. $NH_3$ emissions from human excrement decreased from 362 Gg (6.2 % of total) to 121 Gg (1.3 % of total), due to rural depopulation and improvements in sanitary conditions over the past three





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decades. The contributions of different sources to total $NH_3$ emissions in 1980, 1996, 2006, and 2012 are illustrated in Fig. 4. Emissions from livestock and synthetic fertilizer dominated the total inventories. Specifically, the proportion of emissions from synthetic fertilizers of total emissions peaked at 42.3 % in 1996, and then started to decrease in the following years, which can be attributed to changes in the types of fertilizer used. Furthermore, as mentioned above, the Asian financial crisis and the resulting depression in the livestock market led to a decline in the proportion of emissions from livestock after 1997 and 2006, respectively. The contributions of the chemical industry, waste disposal, traffic, and $NH_3$ escape from thermal power plants reached the peak values of 3.3, 2.8, 1.8, and 1 %, respectively, in 2012.

### 3.2 Spatial distribution of ammonia emissions

Figure 5 displays the spatial patterns of $NH_3$ emissions in 1980, 1990, 2000, and 2012, respectively. Over recent decades, high emission rates of greater than 2000 $kg\,km^{-2}$ were always concentrated in Hebei, Shandong, Henan, Jiangsu, Anhui, and East Sichuan provinces, which form the major areas of intensive agriculture in China. The sum of emissions from these provinces contributed approximately 40 % of national $NH_3$ emissions annually from 1980 to 2012. Emissions rates in northeastern China, consisting of the Liaoning, Jilin, and Heilongjiang provinces, another major grain-producing area that is also important for cattle breeding, showed an increasing trend from the 1980s to the 2000s. Again, synthetic fertilizers and livestock waste dominated the spatial distribution of the total emissions.

As mentioned above, $NH_3$ volatilization from synthetic fertilizer application initially increased rapidly, reaching its peak value in the mid-1990s, and then consistently decreased. This decadal pattern can be observed in the temporal-spatial distribution of $NH_3$ emissions from fertilizers (middle column in Fig. 5). High emission rates, with more than 2000 $kg\,km^{-2}$ released in both 1980 and 1990, occurred mainly in Shandong, Henan, and Jiangsu provinces, where farmers consistently over-applied syn-





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thetic fertilizers (Richter and Roelcke, 2000), as well as in Sichuan, which ranked first in ABC application among all provinces in the 1980s. In 2000, $NH_3$ emission rates from cultivated land in Shandong, Henan, Anhui, and Jiangsu provinces (the North China Plain) generally exceeded 3000 $kg\,km^{-2}$, but decreased to less than 2000 $kg\,km^{-2}$ in 2012 due to a reduction in the use of ABC. The total usage of ABC fertilizer in these provinces in 2000 was 5-fold that in 2012. Hubei, Hunan, Jiangxi, and Guangdong provinces, covering China's major rice-production areas, displayed significant growth in $NH_3$ volatilization from 1980 to 1990, with emissions approximately doubling; however, emissions reduced after the mid-1990s possibly because of the transition of fertilizer usage from ABC to urea in these provinces. In contrast to the variation observed in the areas mentioned above, $NH_3$ emissions in the Northeast Plain encompassing Jilin, Heilongjiang, and Inner Mongolia, and in Xinjiang Province, have consistently increased in recent decades. From the 1990s onwards, grain production in the Northeast Plain entered a rapid growth period, accompanied by an increasing demand for synthetic fertilizers.

The spatial distribution of $NH_3$ emissions from livestock waste was similar to that from synthetic fertilizers, with high emission rates in eastern China, East Sichuan, and parts of Xinjiang. In the 1980s, Sichuan was the largest emitter among all of the provinces, accounting for more than 10 % of emissions from livestock manure management, followed by Inner Mongolia and Henan. More than half of $NH_3$ emissions from livestock in Sichuan originated from cattle rearing. In Henan, both cattle and goats played significant roles in the $NH_3$ emissions, whereas in Inner Mongolia, Qinghai, and Xinjiang, large numbers of sheep were raised and were responsible for 33, 30, and 41 % of livestock emissions in 1980, respectively. From the 1980s to 1990s, emissions in the North China Plain showed more rapid growth than in other areas in China, and almost doubled during this period in Shandong, Hebei, and Anhui, where the contribution of beef cattle, pigs, and poultry increased significantly. Until the 2000s, the North China Plain was the area of highest $NH_3$ emissions, with levels of 3000 $kg\,km^{-2}$ throughout most of Hebei, Shandong, and Henan provinces. The two largest contributors to livestock emis-





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sions in these three provinces were beef cattle and laying hens, which contributed 38 and 19 % in 2000, respectively. Beef cattle and goats were extensively bred in Henan, Shandong, Sichuan, and Hebei provinces, and in 2012 the decrease in their population caused a corresponding decrease in $NH_3$ emissions from livestock manure in these provinces, by approximately 0.14, 0.14, 0.02, and 0.09 Tg, respectively. In addition, although emissions from grazing rearing system were less significant than those from other systems (free-range and intensive) in general, they did become significant in northern Inner Mongolia, central and southern Xinjiang, west-central Qinghai, western Sichuan, and large areas of Tibet.

### 3.3 Comparison with previous studies

Our $NH_3$ emissions inventories provide a detailed description of interannual variation from 1980 to 2012 in China. A comparison between this study and the Regional Emission Inventory in Asia (REAS) is presented in Fig. 6. The figures from REAS for 1980–2000 and 2000–2008 were derived from version 1.1 (Ohara et al., 2007) and 2.1 (Kurokawa et al., 2013), respectively. Note that the interannual variability in the emissions in our study was generally consistent with that in REAS before 1996. However, after that year the annual trend of emissions in our study differed from those in REAS. In addition, the $NH_3$ emissions in REAS were generally higher than those in our study. These differences are likely attributable to differences in the estimations of synthetic fertilizer emissions, discussed below.

In REAS, $NH_3$ emissions from animal manure applied as fertilizer were included as a category of fertilizer emissions (Yan et al., 2003). $NH_3$ from the application of animal waste onto croplands was 2.8 Tg in 2000 in REAS, accounting for approximately 60 % of the total fertilizer emissions in that year. To render these two inventories comparable, we excluded the application of animal waste from the fertilizer emissions in REAS using the value for 2000. A comparison of the emissions from synthetic fertilizer application is presented in Fig. 7. We found that the REAS values were 20–50 % higher than ours in 2000–2005, and this percentage rose to 100 % by 2008, which could be largely





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responsible for the differences of total emissions between REAS and our study in the 2000s. It should be noted that in REAS 2.1, the agricultural emissions were extrapolated from REAS 1.1 for 2000 using the corresponding activity data of the target year (Kurokawa et al., 2013), which could have resulted in considerable inaccuracies due to various missing parameters. On the other hand, the discrepancy possibly originated from the treatment of the types of fertilizer and the corresponding EFs considered in the estimation methods. As mentioned above, the types of fertilizer applied have changed substantially since 1997. Although the total amount of synthetic fertilizers increased significantly, the proportion of highly volatile ABC consistently decreased, which could be responsible for the marked decline in $NH_3$ emissions. However, REAS considered only the total fertilization activities rather than the change in fertilizer types so the emissions in REAS continued to increase in recent years. Moreover, we took into account the local environmental conditions (soil pH, wind speed etc.) and agricultural practices, and used fields results from Chinese studies to correct the EFs, whereas REAS employed only uniform EFs based on European studies, and applied these across the whole of China. Fu et al. (2015) recently estimated synthetic fertilizer $NH_3$ emissions at approximately 3.0 Tg in 2011 using the bi-directional CMAQ model coupled to an agro-ecosystem model, which is similar to our value of 2.8 Tg for the same year and supports the reliability of our inventories.

Table 3 shows the comparison of the emissions from livestock waste in our study with previous ones. Our results are generally in close agreement with those of Zhao and Wang (1994), Streets et al. (2003), and Yamaji et al. (2004). The majority of the previous inventories used European-based EFs, which could introduce significant inaccuracies. Our study employed a mass-flow approach, and considered three different livestock rearing systems, as well as four phases of manure management based on local agricultural practices. The EFs used in our study were also refined according to environmental conditions. Hence, our estimations employed more realistic parameters, and the differences between the present study and previous ones are expected. For the total $NH_3$ emissions, our result (10.1 Tg) was close to that reported (10.4 Tg) using





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a global 3-D chemical transport model for 2005–2008 (Paulot et al., 2014). The global $NH_3$ column mapped with the observations of IASI sensor also demonstrated excellent qualitative agreement with the spatial distribution of our estimated emissions (Van Damme et al., 2014). More specifically, several emissions hotspots, including the North China Plain, Sichuan and Xinjiang provinces (near Ürümqi and in Dzungaria), and the region around the Tarim Basin were also detected by the IASI sensor.

### 3.4 Uncertainty

Uncertainties in $NH_3$ emissions originated from the values used for both the activity and EFs. Huang et al. (2012) summarized the possible sources of uncertainty in the emissions inventory, including extremely high activity data for fertilizer use and livestock, the numerous parameters involved in the EF adjustment, and large variation ($\geq$ 100 %) in the coefficients of biofuel combustion and chemical industry production. Furthermore, our method used constant EFs for estimating 30 year inventories, which may introduce additional uncertainties. First, the application rate and synthetic fertilization method may have changed during recent decades because Chinese farmers have come to expect higher grain production within limited areas of cropland, which may lead to uncertainties in $NH_3$ loss per unit area. Second, although we considered interannual changes in the percentage of intensive rearing systems to livestock emissions, manure management, which was divided into four phases in our method, could have also changed over time because it was affected by many factors including the N content of the feed, housing structure, manure storage system, spreading technique, and time spent outside or indoors (Zhang et al., 2010). In addition, over recent decades, excessive synthetic fertilizer use has caused significant soil acidification in China (Guo et al., 2010), but our inventories did not consider the influence on $NH_3$ volatilization, which may lead to more deviation in the emissions estimation. We ran 20 000 Monte Carlo simulations to estimate the range of $NH_3$ emissions with a 95 % confidence interval for 1980, 1990, 2000, and 2012. The estimated emission ranges were 4.5–7.4, 6.3–11.1, 8.0–13.4, and 7.5–12.1 $Tg\,yr^{-1}$, respectively.





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## 4 Conclusions

We developed comprehensive $NH_3$ emissions inventories from 1980 to 2012 in China. Generally, emissions increased from 1980 to 1996, reaching a peak value of approximately 11.2 Tg, then fluctuated at around 10.5 Tg from 1997 to 2006, but underwent a sharp decrease after 2006. The interannual variation in the emissions is attributable to changes in the types of synthetic fertilizer applied and livestock manure management. These factors were the two major $NH_3$ sources, accounting for more than 80 % of total $NH_3$ emissions, while demonstrating different temporal trends. Emissions from synthetic fertilizers initially rose, from 2.1–4.72 Tg, in the period 1980–1996, and then decreased to 2.81 Tg by 2012, which was caused by a change in the relative contributions of urea and ABC consumption to total emissions. In contrast to synthetic fertilizer emissions, emissions from livestock, ranging from 2.9–6.1 Tg from 1980 to 2012, rose until 2005, but significantly decreased after 2006. Other sources were insignificant in the total budget, and exhibited distinct variation during this period. $NH_3$ emissions generally peaked in the spring and summer, corresponding to planting schedules and ambient temperatures. At the regional level, the spatial patterns of the total emissions have generally been consistent over recent decades, with high emissions rates of more than 2000 kg km$^{-2}$ concentrated in Hebei, Shandong, Henan, Jiangsu, Anhui, and East Sichuan provinces, which represent the major areas of intensive agriculture in China. Compared to $NH_3$ emissions in REAS, our results are more reliable because we considered more parameters when calculating specific EFs according to local ambient conditions and agricultural practices.

It should be noted that gaps still exist in these inventories due to uncertainties in the activity data, EFs, and related parameters, especially for earlier years. As many samples as possible should be used in statistical censuses, and more local field studies should be implemented for better estimates of the EFs to reduce uncertainties. Such high-resolution inventories can be used in global and regional modeling to simulate atmospheric aerosol formation, explore the impacts of $NH_3$ emissions on air quality,





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and understand the evolution of the N cycle and atmospheric chemistry during recent decades. In addition, we expect our results to be validated by top-down estimates in future studies.

**The Supplement related to this article is available online at doi:10.5194/acpd-15-26959-2015-supplement.**

*Acknowledgement.* The MCD45A1 burned area product was provided by the University of Maryland. The MODIS NDVI and VCF were provided by the Land Process Distributed Active Archive Center (LPDAAC), USA. The China Land Cover product was provided by the Environmental & Ecological Science Data Center for West China, National Natural Science Foundation of China. The 1 km population distribution dataset was developed by the Data Center for Resources and Environmental Sciences Chinese Academy of Sciences (RESDC). This study was supported by the Public Welfare Projects for Environmental Protection (201309009 and 201409002), National Natural Science Foundation of China (41275155 and 41121004) and Strategic Priority Research Program of the Chinese Academy of Sciences (No. XDB05010300).

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**Table 1.** Activity dataset and EFs of other minor $NH_3$ sources used in our study.

| Sources | Activity dataset | EFs | Reference |
|---|---|---|---|
| **Nitrogen-fixing plants** | EOCAY (1981–2013) | $0.01\,kg\,NH_3\,kg^{-1}$ N | EEA (2006) |
| **Compost of Crop Residues** | EOCAY (1981–2013) | $0.32\,kg\,NH_3\,t^{-1}$ | Roe et al. (2004) |
| **Biomass burning** | | | |
| Forest fires | MODIS Burned Area (2000–2012), CMF (1990), CMF (1989–1998) and SFA (1999–2000) | $1.1\,g\,NH_3\,kg^{-1}$ | Andreae and Merlet (2001) |
| Grassland fires | MODIS Burned Area (2000–2012) | $0.7\,g\,NH_3\,kg^{-1}$ | Seiler and Crutzen (1980) |
| Crop residues burning | EOCAY (1981–2013) and NBSC (1985–2013) | 0.37 (wheat)$g\,NH_3\,kg^{-1}$ 0.68(maize) 0.52(others) | Li et al. (2007) |
| Fuelwood combustion | NBSC (1985–2013) | $1.3\,g\,NH_3\,kg^{-1}$ | Andreae and Merlet (2001) |
| **Human excrement** | NBSC (1981–2013a) and NBSC (2003–2013a) | $0.787\,kg\,NH_3\,yr^{-1}\,cap^{-1}$ | Buijsman et al. (1987), Moller and Schieferdecker (1989), EPBG (2005) |
| **Chemical industry** | | | |
| Synthetic ammonia | NBSC (1981–2013b) | $0.01\,kg\,NH_3\,t^{-1}$ | EEA (2013) |
| N fertilizers production | NBSC (1981–2013b) | $5\,kg\,NH_3\,t^{-1}$ | Roe et al. (2004) |
| **Waste disposal** | | | |
| Wastewater | NBSC (2003–2013a), | $0.003\,kg\,NH_3\,m^{-3}$ | EPBG (2005) |
| Landfill | Du et al. (2006) | $0.560\,kg\,NH_3\,t^{-1}$ | Roe et al. (2004) |
| Compost | | $1.275\,kg\,NH_3\,t^{-1}$ | Roe et al. (2004) |
| Incineration | | $0.210\,kg\,NH_3\,t^{-1}$ | Sutton et al. (2000) |
| **Traffic** | | | |
| Light-duty gasoline vehicles | CAAM (1983–2013) | $0.026\,g\,NH_3\,km^{-1}$ | Roe et al. (2004) |
| Heavy-duty gasoline vehicles | CAAM (1983–2013) | $0.028\,g\,NH_3\,km^{-1}$ | Roe et al. (2004) |
| Light-duty diesel vehicles | CAAM (1983–2013) | $0.04\,g\,NH_3\,km^{-1}$ | Roe et al. (2004) |
| Heavy-duty diesel vehicles | CAAM (1983–2013) | $0.017\,g\,NH_3\,km^{-1}$ | Roe et al. (2004) |
| Motorcycles | CAAM (1983–2013) | $0.007\,g\,NH_3\,km^{-1}$ | Roe et al. (2004) |
| **Ammonia escape** | CAEPI (2013) | $2.3\,mg\,NH_3\,m^{-3}$ | NEA (2011) |

**Table 2.** Contributions to $NH_3$ emissions (Gg) from various sources from 1980 to 2012.

| Year | Synthetic fertilizer | Agricultural soil | N-fixing crop | Compost | Livestock | Biomass burning | Human excrement | Chemical industry | Waste disposal | Traffic | Ammonia escape | Total |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1980 | 2103 | 175 | 20 | 42 | 2874 | 214 | 362 | 61 | 5 | 3 | | 5859 |
| 1981 | 2077 | 175 | 19 | 43 | 2900 | 214 | 368 | 60 | 5 | 4 | | 5865 |
| 1982 | 2368 | 175 | 20 | 48 | 3023 | 220 | 375 | 62 | 6 | 4 | | 6299 |
| 1983 | 2616 | 175 | 18 | 51 | 3041 | 219 | 383 | 68 | 7 | 4 | | 6582 |
| 1984 | 2812 | 175 | 17 | 54 | 3124 | 223 | 389 | 74 | 7 | 5 | | 6880 |
| 1985 | 2686 | 175 | 18 | 52 | 3271 | 218 | 397 | 70 | 8 | 6 | | 6900 |
| 1986 | 2880 | 175 | 18 | 54 | 3418 | 226 | 405 | 71 | 7 | 7 | | 7260 |
| 1987 | 3015 | 175 | 18 | 57 | 3523 | 267 | 413 | 82 | 8 | 8 | | 7565 |
| 1988 | 3349 | 174 | 17 | 56 | 3707 | 231 | 420 | 83 | 9 | 9 | | 8055 |
| 1989 | 3562 | 174 | 17 | 57 | 3812 | 224 | 430 | 87 | 10 | 10 | | 8383 |
| 1990 | 3474 | 174 | 17 | 63 | 3886 | 234 | 432 | 89 | 17 | 11 | | 8397 |
| 1991 | 3861 | 174 | 16 | 63 | 3920 | 234 | 435 | 92 | 28 | 12 | | 8835 |
| 1992 | 3808 | 174 | 16 | 63 | 4023 | 234 | 438 | 96 | 36 | 13 | | 8900 |
| 1993 | 3803 | 173 | 18 | 66 | 4270 | 237 | 442 | 93 | 43 | 16 | | 9160 |
| 1994 | 4007 | 173 | 19 | 65 | 4684 | 236 | 424 | 106 | 45 | 18 | | 9775 |
| 1995 | 4329 | 173 | 17 | 67 | 5179 | 242 | 404 | 113 | 57 | 20 | | 10 601 |
| 1996 | 4720 | 174 | 15 | 74 | 5339 | 255 | 377 | 130 | 60 | 21 | | 11 164 |
| 1997 | 4528 | 174 | 16 | 72 | 4860 | 246 | 353 | 126 | 69 | 23 | | 10 467 |
| 1998 | 4391 | 174 | 16 | 76 | 5065 | 255 | 327 | 132 | 75 | 25 | | 10 537 |
| 1999 | 4331 | 174 | 19 | 76 | 5133 | 257 | 309 | 139 | 80 | 28 | | 10 546 |
| 2000 | 3797 | 237 | 21 | 76 | 5355 | 249 | 283 | 146 | 96 | 31 | 0.02 | 10 298 |
| 2001 | 3835 | 237 | 22 | 69 | 5367 | 278 | 271 | 154 | 92 | 25 | 0.04 | 10 383 |
| 2002 | 3957 | 237 | 21 | 69 | 5546 | 308 | 269 | 171 | 97 | 39 | 0.07 | 10 715 |
| 2003 | 3692 | 237 | 22 | 65 | 5714 | 310 | 253 | 173 | 103 | 45 | 0.09 | 10 715 |
| 2004 | 3683 | 237 | 21 | 70 | 5988 | 324 | 234 | 203 | 111 | 50 | 0.12 | 10 922 |
| 2005 | 3492 | 237 | 21 | 76 | 6181 | 303 | 209 | 232 | 110 | 58 | 0.12 | 10 906 |
| 2006 | 3319 | 237 | 21 | 77 | 5863 | 313 | 200 | 238 | 113 | 67 | 0.29 | 10 448 |
| 2007 | 3258 | 222 | 19 | 79 | 4982 | 305 | 195 | 258 | 128 | 78 | 0.60 | 9524 |
| 2008 | 3105 | 221 | 20 | 79 | 5024 | 306 | 185 | 264 | 140 | 90 | 0.96 | 9435 |
| 2009 | 3244 | 221 | 20 | 84 | 5200 | 315 | 169 | 277 | 152 | 107 | 1.77 | 9791 |
| 2010 | 2967 | 221 | 20 | 85 | 5090 | 309 | 182 | 271 | 168 | 130 | 3.14 | 9486 |
| 2011 | 2804 | 221 | 19 | 91 | 4967 | 326 | 131 | 296 | 186 | 153 | 4.98 | 9198 |
| 2012 | 2811 | 221 | 18 | 95 | 5053 | 332 | 121 | 308 | 268 | 174 | 86.63 | 9487 |

**Table 3.** Comparison of $NH_3$ emissions ($Tg\,yr^{-1}$) from our study with other published results*.

| Reference | Base year | Total | Synthetic fertilizer | Husbandry | Biomass burning | Others |
|---|---|---|---|---|---|---|
| Zhao and Wang (1994) | 1990 | 13.6/9.8 | 6.4/4.0 | 4.2/4.7 | | 3.0/0.9 |
| Yan et al. (2003) | 1995 | | 4.3/4.3 | | | |
| Streets et al. (2003) | 2000 | 13.6/10.3 | 6.7/3.8 | 5.0/5.3 | 0.8/0.25 | 1.1/0.95 |
| Yamaji et al. (2004) | 1995 | | | 5.1/5.2 | | |
| | 2000 | | | 5.5/5.4 | | |
| Ohara et al. (2007) | 2000 | | | | 0.5/0.24 | |
| Zhang et al. (2011) | 2005 | | 4.3/3.5 | | | |
| Zhao et al. (2013) | 2010 | | 9.8/3.0 | | | |
| Paulot et al. (2014) | 2005–2008 | 10.4/10.1 | | | | |
| Fu et al. (2015) | 2011 | | 3.0/2.8 | | | |

* Before and after the slash represent other studies and this study, respectively.





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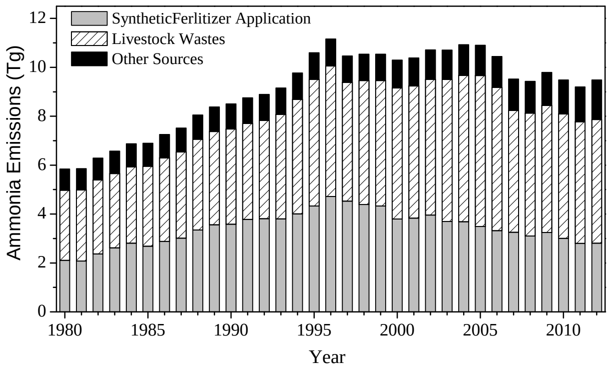


**Figure 1.** Interannual variation in total $NH_3$ emissions in China from 1980 to 2012; the sources of the emissions were categorized as synthetic fertilizer application, livestock manure, and other sources.



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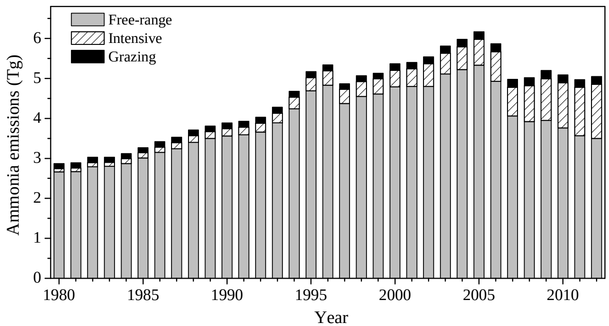


**Figure 2.** Interannual variation in $NH_3$ emissions from livestock manure for three different rearing systems.





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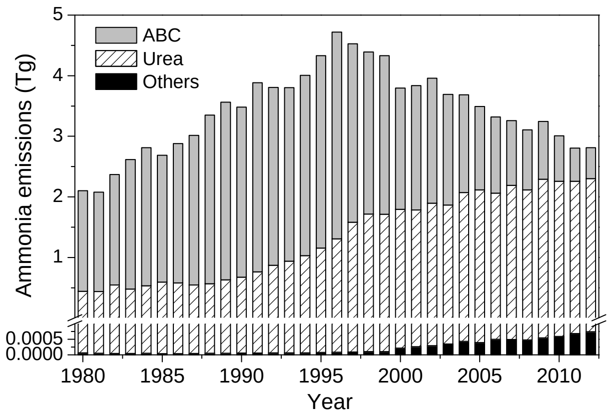


**Figure 3.** Interannual variation in $NH_3$ emissions from synthetic fertilizer in China from 1980 to 2012; types of synthetic fertilizer were categorized as urea, ABC, and others (AN, AS, and others).





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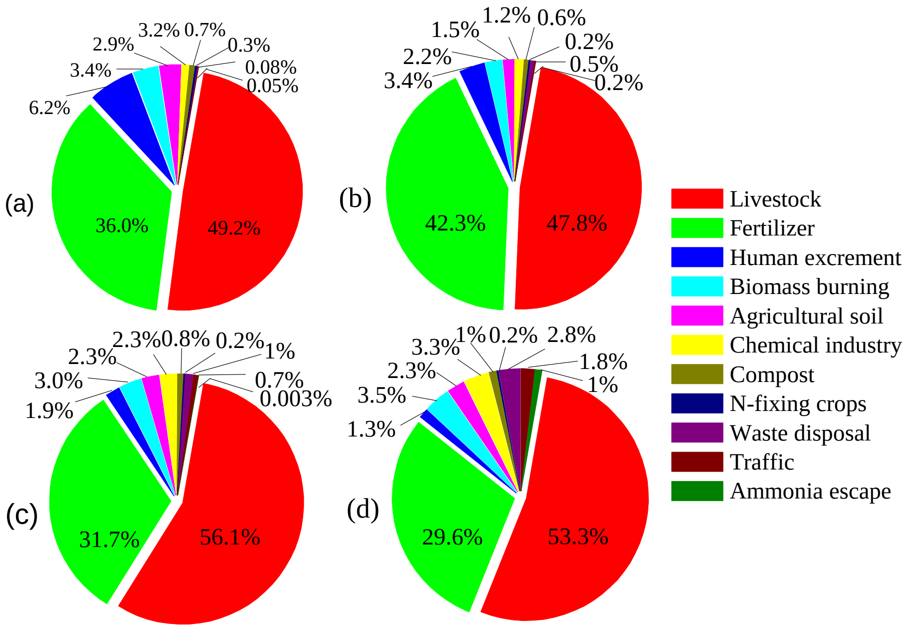


**Figure 4.** Source contributions (%) to $NH_3$ emissions in China in **(a)** 1980, **(b)** 1996, **(c)** 2006, and **(d)** 2012.





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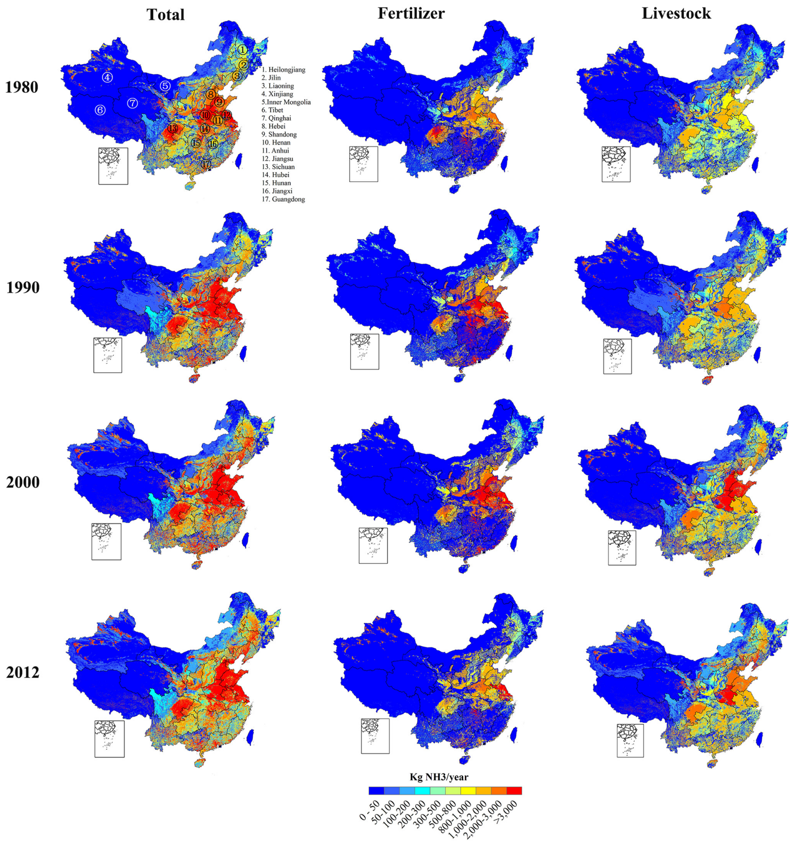


**Figure 5.** Spatial distribution of $NH_3$ emissions in China in 1980, 1990, 2000, and 2012 (from left to right: total emissions, synthetic fertilizer emissions, and livestock emissions).






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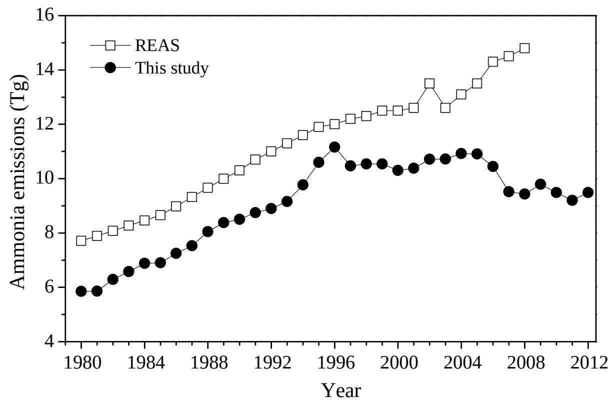


**Figure 6.** Comparison of total $NH_3$ emissions between this study and REAS.





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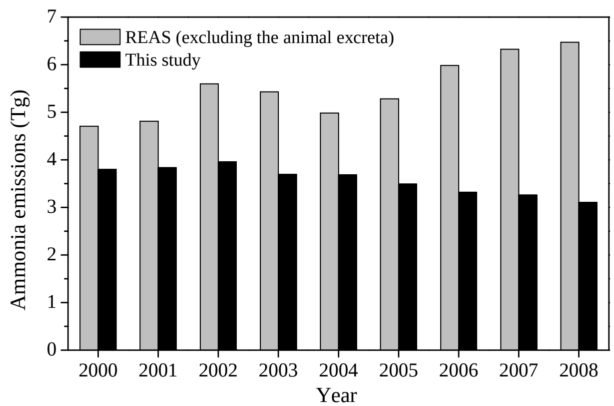


**Figure 7.** Comparison of $NH_3$ emissions from synthetic fertilizers between this study and REAS.





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