Response to Referee #1

The study develops high-resolution NH_3 emission inventories in China during 1980–2012 based on the bottom-up estimates. The authors provide annual trend of NH_3 using region-specific and temporal-varied emission factors. As a result, the authors found the significant annual trend which increased from 1980 to 1996 and then fluctuated from 1997 to 2006, but went down a sharp decrease after 2006. They clarified that this downward trend is mainly caused by a change in the relative contributions of urea and ABC consumption and by an improvement of rearing system for livestock manure. These are new findings. Additionally, the interannual variations of spatial distribution of NH_3 emissions are discussed. This article is the first study in which the interannual and spatial variations of NH_3 emissions in China are estimated based on the top-down methodology using detailed regional information. Consequently, the reviewer recommends publishing this paper and expects that the gridded emission data is opened as early as possible.

Response: Thank the referee for comments on our manuscript and we appreciate the referee's hard work and kindly concern.

Response to Referee #2

This is an interesting paper describing an extension of the earlier inventory published in 2012 (Huang et al. in Global Biogeochemical Cycles) towards a longer time period and with a few modifications of the emission factors used. The approach uses gridded emission factors on a monthly basis, and that is an important improvement compared to annual emissions. This way atmospheric chemistry-transport models can better simulate the fate of the atmospheric ammonia and aerosols. Hence, I think this paper is an important contribution. However, there are a few issues that need to be improved in terms of method description. The paper as it is now does not provide sufficient information to understand how the inventory has been constructed, and only by going back to the 2012 publication readers can understand how the monthly emission factor shave been calculated.

Response: We would appreciate the referee very much for providing the insightful comments, which help us to improve the manuscript greatly.

For example, it is not clear how the emission factor for synthetic fertilizer use has been calculated. My guess is that the authors have a crop calendar with data on timing of fertilizer application. The 2012 paper mentions that a range of different crops are considered, but this information is missing in the current text. Also, it is not clear how then, for fertilizer use in a specific month, emissions are calculated; is it a flush, or is the emission extended over a longer period (the same comment is for all other sources)?

Response: Accepted. It was reworded in Lines 163−181. A crop calendar covering timing of the growth of various crops in each province was used to derive the monthly changes of fertilizer application. 16 kinds of crops cultivated were involved in the activity data and their fertilization timings are generally fixed. Consequently, monthly condition-dependent emission factors were calculated based on timing of crop growth and practice of fertilizer application. In this work, the monthly emissions for all sources were the product of monthly activity data and corresponding EFs. We supposed that these emissions were produced instantaneously from related activities in current month and couldn't extend to next month. More descriptions about the calculation of monthly emissions are listed in Lines 278−289.

Revision in Lines 163–181 on Page 7: "A crop calendar, which involves the type of crop cultivated at specific region and corresponding fertilization timing was used to derive the monthly fertilizer application. We considered 16 kinds of crops in the activity data that are wildly cultivated in different seasons in China, including early rice, semi-late rice, late rice, non-glutinous rice, wheat, maize, bean, potato, peanut, oil crop, cotton, beet, sugarcane, tobacco, vegetables and fruits. We derived monthly condition-specific EFs for synthetic fertilizer volatilization by introducing several influencing factors like the type of fertilizers, soil pH, ambient temperature, fertilization method, and application rate (see Table 2 in (Huang et al., 2012b)). Briefly, EFs are characterized by fertilizer types with ABC and urea more volatile than the other fertilizers. Liner relationships between the volatilization of mineral fertilizers and soil pH were developed to correct EFs (Fan et al., 2005; Bouwman et al., 2002). A threshold of 200 kg N ha^{-1} was defined as the high fertilization rate and when the local fertilization rate exceeded this value, we multiplied EFs by 1.18 (Fan et al., 2006). We derived the relationship between the emission rate and temperature for various fertilizers from EEA (2009) and Lv et al. (1980). Compared to Huang et al. (2012b)，the effects of wind speed and in-situ measurements of NH_3 flux conducted by our research group in a typical cropland were involved to further refine the EFs for synthetic fertilizer emissions in this study."

Revision in Lines 278–289 on Page 12: "The seasonal NH_3 emission estimation for fertilizer application could be calculated as the product of condition-specific EFs derived from meteorological factors (average monthly temperature and wind speed) and monthly fertilizer consumption associated with agricultural timing. For livestock emissions, we assumed that the number of each livestock category per month remains constant, because the monthly fluctuation in the production of meat, eggs and milk is very small (http://www.caaa.cn/). The monthly EFs were distinguished by average monthly temperature and wind speed from NCEP. Besides, the emission from biomass burning also shows a temporal fluctuation. MCD45A1 (monthly burned area product), MOD14A2 and MYD14A2 products (8-day thermal anomalies/fire products) were utilized to ascertain the timing of different kinds of biomass. For other minor sources, the emissions were equally divided into 12 months."

In section 2.2 on livestock waste there is the same problem of lack of information to understand the approach. In addition, I wonder if wind speed is also used for this source, since it is also soil-borne for grazing and spreading –related emissions. I also wonder how the authors can assume that the parameters used to compute TAN have not changed. I guess that the feeding situation has changed in the inventory period, so that the composition and amount of manure or N excretion per kg of product or per animal probably has changed significantly. So a brief discussion on the impact of this assumption is needed.

Response: Accepted. We add more detail description. Please see Lines 223−250. Actually, wind speed was not involved for grazing and manure spreading in the previous manuscript. In this revised version, we recalculated the livestock emissions within the grazing and spreading stages by multiplying an exponent term with original EFs, similar with the modification in the EFs for mineral fertilizers (please see Lines 257−262). Finally, the livestock emissions have been updated in the revised text.
The feed situation in Chinese agricultural has been changed, e.g. animal housing conditions or feedstuff types, and how to parameterize these changes is difficult. However, based on the statistics year book for husbandry, we still introduced an interannual ratio between the free-range system and the intensive one to reflect the change of animal rearing types in the inventory periods. It could represent the changes of Chinese livestock practice in the inventory period to some degree. They were discussed in the manuscript (Lines 627−640).

Revision in Lines 223–250 on Pages 9−10: "In this study, TAN inputted into manure management was the product of the daily amount of urine and faece produced (kg/[day*capita]), N content (%), and TAN content (%) (see Table 3 in (Huang et al., 2012b)). We assumed that these parameters have not changed during the 30-year period and some uncertainties from this assumption would be discussed in Sect. 3.5. 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 corresponding EFs. In the outdoor stage, the excreta were directly deposited in the open air without any treatment after that while animals' excrete inside buildings would release emissions during housing, storage and spreading stages. The periods spent in buildings in a year for different livestock classes were used to determine the portion of excrement indoors or outdoors. After a proportion of TAN was depleted through some processes like immobilization, discharge of NH_3, N_2O and N_2, or the leaching loss of nitrogen, the rest TAN would flow into next stage (EEA, 2013). 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 (except grazing) was larger than a certain value (Table S1). Under this definition, an interannual ratio between the free-range system and the intensive one was introduced to reflect the change of animal rearing types in the inventory periods. It could represent the changes of Chinese livestock practice in the inventory period to some degree."

<u>Revision in Lines 257–262 on Page 11:</u> "Temperature-dependent volatilization rates were considered by using specific EFs at different temperature intervals in the manure housing stage (Koerkamp et al., 1998). We also implemented wind speed and temperature adjustment in the stages of manure spreading and grazing, based on model results reported by Gyldenkaerme et al. (2005). Ambient temperature and wind speed data were also extracted from NCEP final analysis dataset."

<u>Revision in Lines 627–640 on Page 26:</u> "For example, the feed situation in Chinese agricultural has been changed, e.g. animal horsing conditions, feedstuff types or feeding periods. Zhou et al. (2003) conducted rural household surveys on the Chinese household animal raising practices. They found that in some provinces like Zhejiang, industrial processed feed had become a major animal feed. The industry processed feed is easy to digest and absorb, showing more use efficiency than traditional farm-produced forage. Therefore, the amount of N excreta per animal feed by industry forage should be less than that by farm forage. But Li et al. (2009) investigated that compared with 1990s, the average N content in manure from pig, chicken, beef and sheep has little change in recent years according to nationwide 170 samples analysis. On the other hand, rearing periods for animals like poultry were significantly reduced during recent years along with the development of breeding technology, that is, manure excreted per animal per year was supposed to be declining. However, this change was not considered in this study and it may result in overestimation of livestock emissions in recent years."

Finally, I wonder how the authors can use monthly temperatures and monthly wind speed as a factor in the calculation of the emission factors. How representative are monthly mean wind speed and temperature, while perhaps maximum day temperature and variability of wind speed are better predictors of NH3 emissions. In addition, there may be an interaction between temperature and wind speed that is not represented in the emission factor approach.

Response: Yes. It could be better to use daily temperature and wind speed for estimation. However, in this study, the daily activity data is not available (we could not quantify the synthetic fertilizer use each day), or we could not identify the exact date of fertilizer application and the timing varies annually. We adopted monthly mean temperature to parameterize the emission factor according to EEA (2009). Despite the uncertainties, we still use the monthly mean temperature and wind speed to produce monthly emission inventories. Please see Lines 208−215 in the revised text. We admit there might be some interactions between temperature and wind speed. Some discussion on this uncertainty in Section 3.5 in the revised manuscript (Please see Lines 607−616).

Revision in Lines 208–215 on Page 9: "It should be note that in this study, we used monthly weather values in the adjustment of EFs rather than the daily maximum since the daily activity data was not available (we could not quantify the synthetic fertilizer use each day) or we could not identify the exact date of fertilizer application and the timing varies annually. On the other hand, we adopted the parameterization of temperature adjustment provided by EEA (2009), which is also based on mean temperature. Despite the uncertainties, we still used the monthly mean temperature and wind speed to produce monthly emission inventories."

Revision in Lines 607–616 on Page 25: "In this study, the impacts of wind speed and ambient temperature on the EFs in agricultural ammonia emissions were isolated but in real condition, there might be some interactions between temperature and wind speed. Ogejo et al. (2010) indicated that parameter interactions may play a significant role in emission estimation with a process-based model for ammonia emissions but they also didn't consider the interaction between temperature and wind velocity. Actually, previous studies generally examined the respective effect of wind speed and temperature on ammonia volatilization according to controlled experiments (Sommer et al., 1991) and we expect more experimental evidences for the interaction effect."

In relation to this I wonder if this inventory is better than the one published earlier. Have the authors tested this claim. I am asking this, because the emission estimates are quite close, and given the uncertainties I wonder if the modifications are really improvements. Also the claim that the approach of this paper is better or more realistic than emission factors that are based on less factors needs some more thinking. I wonder if the authors can show that this is the case. Does the approach of this paper result in a better comparison with Paulot et al. (2014) and Van Damme et al. (2014) than the previous version of Huang et al. (2012) and of less sophisticated emission factor approaches? In my opinion the claim that a model is better needs to be supported by evidence.

Response: Accepted. Indeed, the emission amounts are close between the two results though some modifications have been done in the present study. It is not surprising because we mostly followed Huang et al.'s method. It should be noted that in our study, we mainly focused on compiling a long-term emission inventory rather than developing a new methodology for NH_3 emission. It covered more than 30 years. These comprehensive multi-year inventories are very necessary for global and regional air quality modeling, especially for studying the impacts of NH_3 emissions on air quality.

Moreover, some modifications are still needed to improve the emissions as some potential problems existed in Huang et al.'s method (listed in Lines 124−129 in the introduction part). First, we used wind speed in the adjustment of emission factors. Secondly, we applied the NH_3 EF which was measured by using micrometeorological method for whole a year in a typical farmland in North China Plain by our research group (Huo et al., 2014, 2015). The in-situ results could represent better than those used in Huang et al. which were derived from studies in early years.

Paulot et al. (2014) estimated the average annual ammonia emission of 10.4 Tg in the period of 2005−2008. For the same period, our result 10.2 Tg, and Huang et al. (2012b) estimated 9.8 Tg in 2006. The three results are close. The excellent qualitative agreement for spatial distributions could be found between our estimations and IASI sensor (Van Damme et al., 2014). The comparisons have been discussed in the revised manuscript (please see Lines 591−598).

Revision in Lines 124–129 on Pages 5−6: "However, there were still some problems in this method; for example, Huang et al. generally adopted EFs reported in early years and up-to-date in-situ measurement was needed; moreover, wind speed that could be of importance in emission estimation was not considered in that study. Though Huang et al. have involved as many NH_3 emitters as possible, some minor sources may be neglected like fertilization in orchard, NH_3 escape from thermal power plants."

Revision in Lines 591–598 on Page 24: "Paulot et al., (2014) estimated the annual NH_3 emissions of 10.4 Tg using a global 3D chemical transport model averagely in 2005−2008 while our result was 10.2 Tg for the same period and Huang et al. (2012b) estimated 9.8 Tg in 2006. The three results are quite close. We also found excellent qualitative agreement for spatial distribution between our estimation and the global NH_3 column retrieved by IASI sensor (Van Damme et al., 2014). Several emissions hotspots shown in this study, 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."

Finally, I wonder why the authors have tried to generate monthly emissions, but nowhere discuss the temporal variation (likewise, the 2012 Huang et al. paper also lacks such a discussion). It would also be interesting to test if the temporal patterns are changing with the shifts in the different sources?

Response: Accepted. A subsection discussing the temporal variation has been added in Sect. 3.3 (please see Lines 477−540).

Revision in Lines 477–540 on Pages 19−22: "The temporal patterns of NH_3 emissions in 1980, 1990, 2000, and 2012 are clearly presented in Fig. 6 (a). The emissions were primary concentrated during April to September due to the intensive agricultural activities and higher temperatures. Specifically, the different sources showed the diverse distribution characteristics.

Figure 6 (b) describes the temporal distributions of NH_3 emissions from synthetic fertilizer application in 1980, 1990, 2000, and 2012, respectively. It is obvious that the monthly emissions from fertilizer exhibited similar seasonal distribution among different years. Generally, the largest emissions were occurred in summer (June to August), accounting for 44.8–47.7% of annual emissions from synthetic fertilizer, which are attributed to denser fertilization and higher temperatures during this time. Conversely, because of the less NH_3 volatilization related to lower temperatures and relatively rare cultivation during the winter (December to February), the NH_3 emissions reduced to 7.7–11.1% of annual fertilizer emissions. In China, the new spring seeding begins in April and is accompanied by corresponding fertilizer application. In the following 1–2 months, due to application of top fertilizer and warming temperatures, particularly in eastern and central provinces such as Jiangsu, Anhui and Henan, the NH_3 emissions continuous increase to August. In the North China Plain, the winter wheat-summer maize rotation system has practiced as a characteristic farming practice. The high emission rates in June and August could be attributed to the basal dressing and top dressing of summer plants, such as maize. From autumn on, most of the crops begin to harvest, which lead to the decline of emissions during this time. In particular, winter wheat is usually seeded in September with the application of basal dressing, and the top dressing is applied two months later, which could be responsible for the peak emissions occurred during September and November. Besides, owing to more temperature fluctuations and fertilizer application, the monthly distribution of emissions in the northern regions was more remarkable than that in the southern.

The significant seasonal dependence of NH_3 emissions from livestock wastes in different years can be clearly seen in Fig. 6 (c). The monthly distributions of NH_3 emissions were highly consistent with the variations in temperature under the premise of the constant animal population among the different months we assumed above. The major emissions occurred in warmer months (May to September), and more than 45% of the annual livestock emissions, which could be explained by more NH_3 volatilization related to substantial increase of temperature. In contrast, the lowest NH_3 emissions from livestock wastes were estimated in winter (December to February), and this is attributed to relatively smaller EFs linked to lower temperatures. Apart from the two major sources, the NH_3 emissions from biomass burning also had distinctly temporal disparities in spite of the relatively small contribution of total emissions.

The temporal variation of emissions from crop burning in fields from 2003 to 2012 (when the annual MODIS thermal anomalies/fire products (MOD/MYD14A1)) were available) is described in Fig. 6 (d). The occurrences of crop burning in fields were concentrated in March to June with another smaller peak in October, which are consistent with local sowing and harvest times (Huang et al., 2012a). The highest emissions rates occurred in June are mostly attributed to the burning of winter wheat straw that fertilizes the soil after the harvest (in the end of May) in the North China Plain. The peak in October can be partly explained by the burning of maize straw after the harvest (in the end September) in the North China Plain. In addition, the south China including Guangdong and Guangxi provinces have two or three harvest times every year. The sowing time for crops here begins in March, when crop residues would be burned to increase the soil fertility. Simultaneously, in northeast China, there is the local farming practice of clearing the farmland before sowing in April, which may emit corresponding NH_3 during the spring. In winter, the mature period of late rice in south China lead to a certain amount of NH_3.

Figure 6 (e) displays the seasonal distribution of NH_3 emissions from forest and grass fires from 2001 to 2012 (when the annual MODIS burned area product (MCD45A1) was available) in China. The weather and vegetation conditions are regard as dominate factors that regulate the fire activity (Perry et al., 2011). The fire emissions were primarily concentrated in February to April and August to October, because of scarce precipitation, high wind speed and gradually rising temperature during early spring and late winter, especially in the southwestern regions. Simultaneously, the lower moisture content of vegetation is favor of burning. In addition, the abundant fallen leaves and crop residues in autumn could make contributions to the fires dramatically."

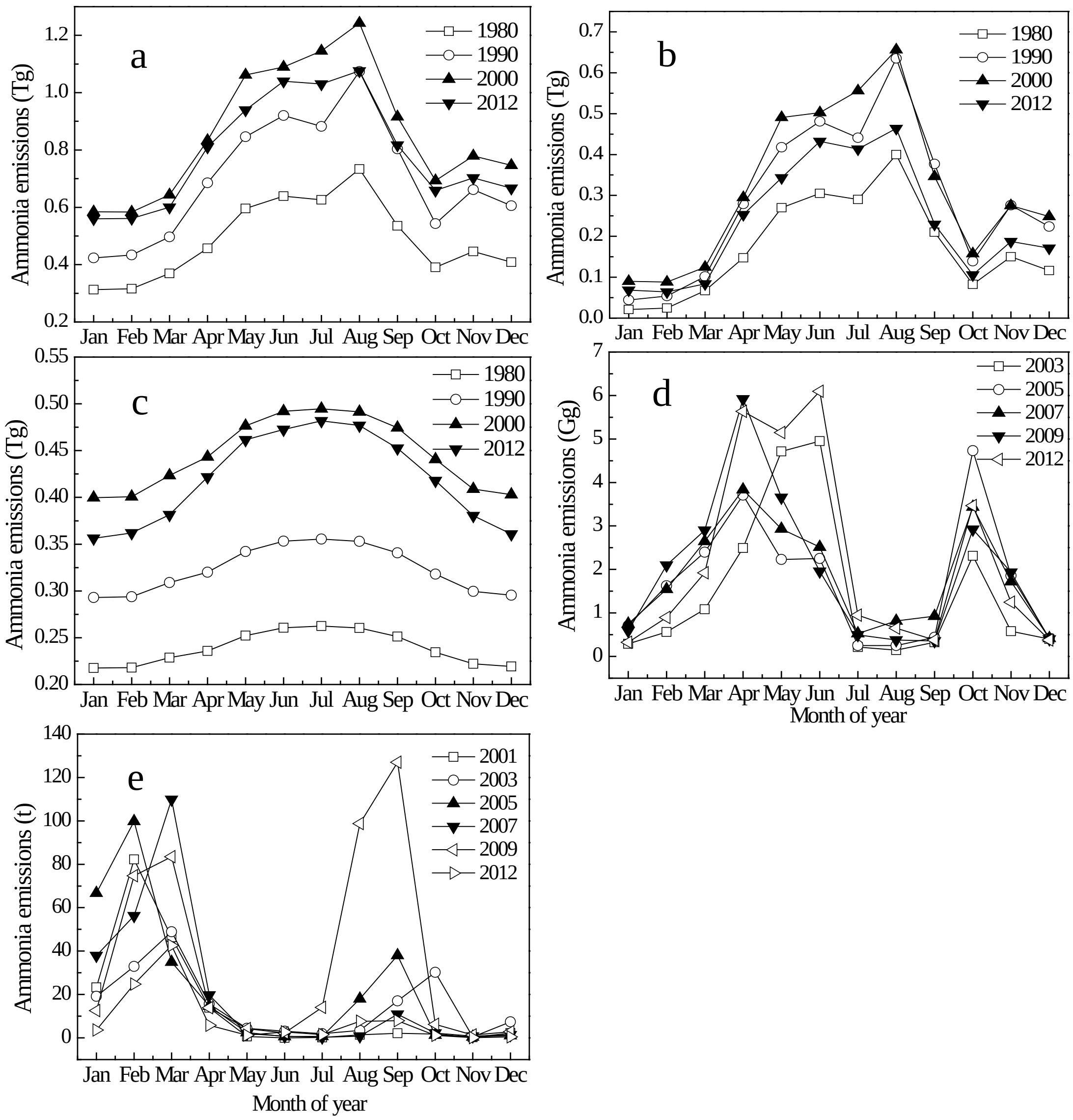

Figure 6. Monthly distribution of NH_3 emissions from different sources in China: (a) all the sources; (b) synthetic fertilizer; (c) livestock wastes; (d) crop burning in fields; (e) forest and grass fires.

Minor comments

-Table S1 is copied from the Huang et al. (2012) paper except for the EEA reference which is now more recent. To avoid problems, this needs to be made clear.

Response: Accepted. We thank the reviewer for the suggestion. Actually, there are almost no differences between recent released EEA inventory guide and the early version for the parameters we used. So we removed this table in the revised manuscript and the detailed values can be looked up in Table 3 in Huang et al.'s paper. We have added more information about estimation method to livestock emissions. Please see Section 2.2.

-Header of section 2.1.2: the soil pH cannot be a source of ammonia.

Response: Accepted. We changed it to "In-situ measurement" in Section 2.1.1 (Line 183).

-The section 2.1.2 on improvement of the EF for soil pH is not clear.

Response: Accepted. We reworded it in Section 2.1.1 (please see Lines 184–195).

Revision in Lines 184–195 on Page 8: "For acquiring up-to-date EFs that could reflect NH_3 volatilization from synthetic fertilizer application in present Chinese agricultural practice, we measured NH_3 EF by using micrometeorological method for a whole year in a typical farmland in the North China Plain and an inverse dispersion model was also used to derive the ammonia EFs (Huo et al., 2014, 2015). The in-situ results could represent better than those used in Huang et al. which were derived from studies in early years. The soil pH and mean air temperature in this farmland was 8.2 and 15°C, respectively. The measurement yielded an NH_3 EF for urea of 12%±3% in this case. Huang et al. (2012b) develop a linear relationship between NH_3 volatilization and soil pH to involve the impact of soil acidity on EFs according to Cai et al. (1986) and Zhu et al. (1989). We applied the condition-specific EF we measured recently to refine this relationship with linear regression analysis."

-A description of the approach for the Monte Carlo analysis is missing.

Response: Accepted. We add a brief description about the Monte Carlo method in Lines 644−651.

Revision in Lines 644–651 on Page 26: "Monte Carlo is an effective method to evaluate the uncertainties in various issues including an emission inventory. In Monte Carlo simulation, random numbers are selected from each distribution (normal or uniform) of input variables and the output uncertainty of an emission inventory is based on the input uncertainties from activity data and emission factors. In this study, 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 Tg/yr, 6.3–11.1 Tg/yr, 8.0–13.4 Tg/yr, and 7.5–12.1 Tg/yr, respectively."

-It is not clear how the temporal distribution of the other sources has been done.

Response: Accepted. We add a brief description about the method of the monthly NH_3 emissions from the other sources in Section 2.4 (please see Lines 278−289).

Revision in Lines 278–289 on Page 12: "The seasonal NH_3 emission estimation for fertilizer application could be calculated as the product of condition-specific EFs derived from meteorological factors (average monthly temperature and wind speed) and monthly fertilizer consumption associated with agricultural timing. For livestock emissions, we assumed that the number of each livestock category per month remains constant, because the monthly fluctuation in the production of meat, eggs and milk is very small (http://www.caaa.cn/). The monthly EFs were distinguished by average monthly temperature and wind speed from NCEP. Besides, the emission from biomass burning also shows a temporal fluctuation. MCD45A1 (monthly burned area product), MOD14A2 and MYD14A2 products (8-day thermal anomalies/fire products) were utilized to ascertain the timing of different kinds of biomass. For other minor sources, the emissions were equally divided into 12 months."

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

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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~~1 Tg from 1980 to 1996, and then decreased to 9.~~5~~7 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~~86 Tg (1980) to 6.~~17~~16 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.

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 the growth of crops, has also undergone a considerable, sustained ~~increase.~~increased. 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 tons in 1980 to 43 million tons 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~~showing 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.

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., 2012b). 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., 2012b). 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. However, there were still some problems in this method; for example, Huang et al. (2012b) generally adopted EFs reported in early years and up-to-date in-situ measurement was needed; moreover, wind speed that could be of importance in emission estimation was not considered in that study. Though Huang et al. (2012b) have involved as many NH_3 emitters as possible in the inventory, some minor sources may be neglected like fertilization in orchard, NH_3 escape from thermal power plants. Nevertheless, this bottom-up emission inventory appears to be reliable, and the method can be used to estimate NH_3 emissions in China.

In this study, we mainly focused on compiling a long-term emission inventories based on Huang et al.'s method and some improvements to this method have been made. Sources of NH_3 in our inventories were listed as follow: 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:

$$E(NH_3) = \sum_{i}\sum_{p}\sum_{m}(A_{i,p,m} \times EF_{i,p,m}), \qquad (1)$$

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~~each 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. (2012b).

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 and consumption, which was calculated as the product of cultivated area and the application rate to crops. (EOCAY, 1981−2013; Zhang et al., 2012; NBSC, 2003−2013a). A crop calendar, which involves the type of crop cultivated at specific region and corresponding fertilization timing was used to identify the monthly fertilizer application. We considered 16 kinds of crops that are wildly cultivated in different seasons in China, including early rice, semi-late rice, late rice, non-glutinous rice, wheat, maize, bean, potato, peanut, oil crop, cotton, beet, sugarcane, tobacco, vegetables and fruits. We derived monthly condition-specific EFs for synthetic fertilizer volatilization by introducing several influencing factors like the type of fertilizer, soil pH, ambient temperature, fertilization method, and application rate (see Table 2 in (Huang et al., 2012b)). Briefly, EFs were characterized by fertilizer types with ABC and urea more volatile than the other fertilizers. Liner relationships between the volatilization of mineral fertilizers and soil pH were developed to correct EFs (Fan et al., 2005; Bouwman et al., 2002). A threshold of 200 kg N ha^{-1} was defined as the high fertilization rate and when the local fertilization rate exceeded this value, we multiplied EFs by 1.18 (Fan et al., 2006). We derived the relationship between the emission rate and temperature for various fertilizers from EEA (2009) and Lv et al. (1980). Compared to Huang et al. (2012b)，the effects of wind speed and in-situ measurements of NH_3 flux conducted by our research group in a typical cropland were involved to further refine the EFs for synthetic fertilizer emissions in this study.

2.1.1 In-situ measurement

For acquiring up-to-date EFs that could reflect NH_3 volatilization from synthetic fertilizer application in present Chinese agricultural practice, we measured NH_3 EF by using micrometeorological method for a whole year in a typical farmland in the North China Plain and an inverse dispersion model was also used to derive the ammonia EFs (Huo et al., 2014, 2015). The in-situ results could represent better than those used in Huang et al. which were derived from studies in early years. The soil pH and mean air temperature in this farmland was 8.2 and 15°C, respectively. The measurement yielded an NH_3 EF for urea of 12%±3% in this case. Huang et al. (2012b) develop a linear relationship between NH_3 volatilization and soil pH to involve the impact of soil acidity on EFs according to Cai et al. (1986) and Zhu et al. (1989). We applied the condition-specific EF we measured recently to refine this relationship with linear regression analysis.

2.1.2 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). 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~~Both the average monthly wind ~~speeds~~speed and ambient temperature mentioned above for a 1 km × 1 km grid were derived from the final analysis dataset of the National Centers for Environmental Prediction (NCEP). It should be noted that in this study, we used mean monthly weather values in the adjustment of EFs rather than the daily maximum since the daily activity data was not available (we could not quantify the synthetic fertilizer use each day) or we could not identify the exact date of fertilizer application and the timing varied annually. On the other hand, we adopted the parameterization of temperature adjustment provided by EEA (2009), which is also based on mean temperature. Despite the uncertainties, we still used mean monthly temperature and wind speed to produce monthly inventories.

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~~Ammoniacal N (TAN) produced from livestock waste can ~~produce ammoniacal N (TAN), which can~~ be converted into gaseous NH_3 or lost through other pathways during different process of manure management (Webb and Misselbrook, 2004; Webb et al., 2006). In this study, TAN inputted into manure management was the product of the daily amount of urine and faece produced (kg/[day*capita]), N content (%), and TAN content (%) (see Table 3 in (Huang et al., 2012b)). We assumed that these parameters have not changed during the 30-year period and some uncertainties from this assumption would be discussed in Sect. 3.5. 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*capita]), N content (%), and TAN content (%) (Table S1), all of the parameters for which were assumed to have not changed during the 30-year period.~~ with the corresponding EFs. In the outdoor stage, the excreta were directly deposited in the open air without any treatment after that while animals' excrete inside buildings would release emissions during housing, storage and spreading stages. The periods spent in buildings in a year for different livestock classes were used to determine the portion of excrement indoors or outdoors. After a proportion of TAN was depleted through some processes like immobilization, discharge of NH_3, N_2O and N_2, and the leaching loss of nitrogen, the rest TAN would flow into next stage (EEA, 2013). 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).~~(except grazing) was larger than a certain value (Table S1). Under this definition, an interannual ratio between the free-range system and the intensive one was introduced to reflect the change of animal rearing types in the inventory periods. It could represent the changes of Chinese livestock practice in the inventory period to some degree.

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.~~. We mainly adopted the EFs in each stage for different livestock classes that are listed in Table S2 in Huang et al. (2012b). Temperature-dependent volatilization rates were considered by using specific EFs at different temperature intervals in the manure housing stage (Koerkamp et al., 1998). We also implemented wind speed and temperature adjustment in the stages of manure spreading and grazing, based on model results reported by (Gyldenkaerne et al., 2005). Ambient temperature and wind speed data were extracted from NCEP final analysis dataset.

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. (2012b). 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 NOx abatement in thermal 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).

2.4 Monthly Emissions

The seasonal NH_3 emission estimation for fertilizer application could be calculated as the product of condition-specific EFs derived from meteorological factors (average monthly temperature and wind speed) and monthly fertilizer consumption associated with agricultural timing. For livestock emissions, we assumed that the number of each livestock category per month remains constant, because the monthly fluctuation in the production of meat, eggs and milk is very small (http://www.caaa.cn/). The monthly EFs were distinguished by average monthly temperature and wind speed from NCEP. Besides, the emission from biomass burning also shows a temporal fluctuation. MCD45A1 (monthly burned area product), MOD14A2 and MYD14A2 products (8-day thermal anomalies/fire products) were utilized to ascertain the timing of different kinds of biomass. For other minor sources, the emissions were equally divided into 12 months.

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 NH_3 emissions have 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~~1 Tg between 1980 and 1996, then decreased to 9.~~5~~7 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 1,400 million, and 0.9 to 10 billion respectively, from the 1980s to mid-2000s (Fig. S1). ~~Correspondingly, NH~~$_3$In this period, large quantities of NH_3 derived from livestock waste have been emitted into the atmosphere. As shown in Fig. 2, 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 had 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-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~~S2 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 replaced ABC and become the dominant chemical fertilizer used in cropland over the last 20 years. In 1980, 3.0 million and 5.1 million tons of urea and ABC, respectively, were produced; by 2012, these values had changed to approximately 28.8 million and 3.4 million tons, 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. Other sources (except synthetic fertilizer and livestock) made no notable contribution to the total budget due to their relatively low levels; nevertheless, some of these sources exhibited distinct variation during this period. For example, 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. Particularly, in 1987, 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 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 traffic, chemical industry, waste disposal~~, traffic,~~ and NH_3 escape from thermal power plants reached the peak values of 4.0%, 3.2%, ~~1.8%~~2.8% and 0.9% in 2012, respectively.

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 2,000 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 2,000 kg/km^2 released in both 1980 and 1990, occurred mainly in Shandong, Henan, and Jiangsu provinces, where farmers consistently over-applied synthetic 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 3,000 kg/km^2, but decreased to less than 2,000 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%, 31%, and 42% 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 3,000 kg/km^2 throughout most of Hebei, Shandong, and Henan provinces. The two largest contributors to livestock emissions 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 Tg, 0.14 Tg, 0.02 Tg, and 0.09 Tg, respectively. ~~In addition, although emissions~~Emissions from grazing rearing system were less significant nationally than those from other systems (free-range and intensive~~) in general,~~), but they did become important in northern Inner Mongolia, central and southern Xinjiang, west-central Qinghai, western Sichuan, and large areas of Tibet.

~~3.3~~3.3 Monthly variation in ammonia emissions

The monthly variation in NH_3 emissions in 1980, 1990, 2000, and 2012 are clearly presented in Fig. 6 (a). The emissions were primary concentrated during April to September due to the intensive agricultural activities and higher temperatures. Specifically, the different sources showed the diverse distribution characteristics. Figure 6 (b) describes the temporal distributions of NH_3 emissions from synthetic fertilizer application in 1980, 1990, 2000, and 2012, respectively. It is obvious that the monthly emissions from fertilizer exhibited similar seasonal distribution among different years. Generally, the largest emissions were occurred in summer (June to August), accounting for 44.8–47.7% of annual emissions from synthetic fertilizer, which are attributed to denser fertilization and higher temperatures during this time. Conversely, because of the less NH_3 volatilization related to lower temperatures and relatively rare cultivation during the winter (December to February), the NH_3 emissions reduced to 7.7–11.1% of annual fertilizer emissions. In China, the new spring seeding begins in April and is accompanied by corresponding fertilizer application. In the following 1–2 months, due to application of top fertilizer and warming temperatures, particularly in eastern and central provinces such as Jiangsu, Anhui and Henan, the NH_3 emissions continuous increase to August. In the North China Plain, the winter wheat-summer maize rotation system has practiced as a characteristic farming practice. The high emission rates in June and August could be attributed to the basal dressing and top dressing of summer plants, such as maize. From autumn on, most of the crops begin to harvest, which lead to the decline of emissions during this time. In particular, winter wheat is usually seeded in September with the application of basal dressing, and the top dressing is applied two months later, which could be responsible for the peak emissions occurred during September and November. Besides, owing to more temperature fluctuations and fertilizer application, the monthly distribution of emissions in the northern regions was more remarkable than that in the southern.

The significant seasonal dependence of NH_3 emissions from livestock wastes in different years can be clearly seen in Fig. 6 (c). The monthly distribution of NH_3 emissions was highly consistent with the variation in temperature under the premise of the constant animal population among the different months we assumed above. The major emissions occurred in warmer months (May to September), and more than 45% of the annual livestock emissions, which could be explained by more NH_3 volatilization related to substantial increase of temperature. In contrast, the lowest NH_3 emissions from livestock wastes were estimated in winter (December to February), and this is attributed to relatively smaller EFs linked to lower temperatures.

Apart from the two major sources, the NH_3 emissions from biomass burning also had distinctly temporal disparities in spite of the relatively small contribution of total emissions.

The temporal variations of emissions from crop burning in fields from 2003 to 2012 (when the annual MODIS thermal anomalies/fire products (MOD/MYD14A1)) were available) are described in Fig. 6 (d). The occurrences of crop burning in fields were concentrated in March to June with another smaller peak in October, which are consistent with local sowing and harvest times (Huang et al., 2012a). The highest emissions rates occurred in June are mostly attributed to the burning of winter wheat straw that fertilizes the soil after the harvest (in the end of May) in the North China Plain. The peak in October can be partly explained by the burning of maize straw after the harvest (in the end September) in the North China Plain. In addition, the south China including Guangdong and Guangxi provinces have two or three harvest times every year. The sowing time for crops here begins in March, when crop residues would be burned to increase the soil fertility. Simultaneously, in northeast China, there is the local farming practice of clearing the farmland before sowing in April, which may emit corresponding NH_3 during the spring. In winter, the mature period of late rice in south China lead to a certain amount of NH_3.

Figure 6 (e) displays the seasonal distribution of NH_3 emissions from forest and grass fires from 2001 to 2012 (when the annual MODIS burned area product (MCD45A1) was available) in China. The weather and vegetation conditions are regard as dominate factors that regulate the fire activity (Perry et al., 2011). The fire emissions were primarily concentrated in February to April and August to October, because of scarce precipitation, high wind speed and gradually rising temperature during early spring and late winter, especially in the southwestern regions. Simultaneously, the lower moisture content of vegetation is favor of burning. In addition, the abundant fallen leaves and crop residues in autumn could make contributions to the fires dramatically.

3.4 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~~7. 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~~8. 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 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 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)~~Paulot et al. (2014) estimated the annual NH_3 emissions of 10.4 Tg using a global 3D chemical transport model averagely in 2005−2008 while our result was 10.2 Tg for the same period and Huang et al. (2012b) estimated 9.8 Tg in 2006. The three results are quite close. We also found excellent qualitative agreement for spatial distribution between our estimation and the global NH_3 column retrieved by IASI sensor (Van Damme et al., 2014). Several emission hotspots shown in this study, 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~~5 Uncertainty

Uncertainties in NH_3 emissions originated from the values used for both the activity and EFs. Huang et al. (2012b) 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 (≥100%) in the coefficients of biofuel combustion and chemical industry production. We may miss some possible sources like NH_3 loss from fertilization in orchard and also overestimated the emissions in agricultural soils covered with plastic shed. In this study, the impacts of wind speed and ambient temperature on the EFs in agricultural ammonia emissions were isolated but in real condition, there might be some interactions between temperature and wind speed. Ogejo et al. (2010) indicated that parameter interactions may play a significant role in emission estimation with a process-based model for ammonia emissions but they also didn't consider the interaction between temperature and wind velocity. Actually, previous studies generally examined the respective effect of wind speed and temperature on ammonia volatilization according to controlled experiments (Sommer et al., 1991) and we expect more experimental evidences for the interaction effect.

Furthermore, our method mostly used constant ~~EFs~~ parameters for estimating 30-year inventories rather than the time-varying, 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, that 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). For example, the feed situation in Chinese agricultural has been changed, e.g. animal horsing conditions, feedstuff types or feeding periods. Zhou et al. (2003)

conducted rural household surveys on the Chinese household animal raising practices. They found that in some provinces like Zhejiang, industrial processed feed had become a major animal feed. The industry processed feed is easy to digest and absorb, showing more use efficiency than traditional farm-produced forage. Therefore, the amount of N excreta per animal feed by industry forage should be less than that by farm forage. But Li et al. (2009) investigated that compared with 1990s, the average N content in manure from pig, chicken, beef and sheep has little change in recent years according to nationwide 170 samples analysis. On the other hand, rearing periods for animals like poultry were significantly reduced during recent years along with the development of breeding technology, that is, manure excreted per animal per year was supposed to be declining. However, this change was not considered in this study and it may result in overestimation of livestock emissions in recent years. In addition, over recent decades, excessive synthetic fertilizer use has caused significant soil acidification in China (Guo et al., 2010), but our inventories didn't consider the influence on NH_3 volatilization~~, which may lead to more deviation in the emissions estimation. We~~. Monte Carlo is an effective method to evaluate the uncertainties in various issues including an emission inventory. In Monte Carlo simulation, random numbers are selected from each distribution (normal or uniform) of input variables and the output uncertainty of an emission inventory is based on the input uncertainties from activity data and emission factors. In this study, 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 Tg/yr, 6.3–11.1 Tg/yr, 8.0–13.4 Tg/yr, and 7.5–12.1 Tg/yr, respectively.

4 Conclusions

We developed comprehensive NH_3 emission inventories from 1980 to 2012 in China. Generally, emissions increased from 1980 to 1996, reaching a peak value of approximately 11.~~2~~1 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~~7 Tg, in the period 1980–1996, and then decreased to 2.~~81~~8 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~~,~~ but they could play a role in specific region or periods like vehicles on road in big cities, crop residue burning and ~~exhibited distinct variation during this period.~~large wild fires due to agricultural timing and climate conditions. NH_3 emissions generally peaked in the spring and summer, corresponding to planting schedules and relatively high temperature that were the two determining factors for the monthly variation of mineral fertilizer and livestock emissions, respectively. The emissions from crop residue burning were generally concentrated from March to June and October when major crops like winter wheat and corn are harvested. 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 2,000 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 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, 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.

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Table captions

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

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

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

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 N	EEA (2006)
Compost of Crop Residues	EOCAY (1981-2013)	0.32 kg NH_3/ton	Stephen et al. (2004)
Biomass burning			
Forest fires	MODIS Burned Area (2000-2012), CMF (1990), CMF (1989-1998) and SFA (1999-2000)	1.1 g NH3/kg	Andreae and Merlet (2001)
Grassland fires	MODIS Burned Area (2000-2012)	0.7 g NH_3/kg	Seiler and Crutzen (1980)
Crop residues burning	EOCAY (1981-2013) and NBSC (1985-2013)	0.37(wheat)g NH_3/kg 0.68(maize) 0.52(others)	Li et al. (2007)
Fuelwood combustion	NBSC (1985-2013)	1.3 g NH_3/kg	Andreae and Merlet (2001)
Human excrement	NBSC (1981-2013b) and NBSC (2003-2013b)	0.787 kg NH_3/year/cap	Buijsman et al. (1987), Moller and Schieferdecker (1989), EPBG (2005)
Chemical industry			
Synthetic ammonia	NBSC (1981-2013a)	0.01 kg NH_3/ton	EEA (2013)
N fertilizers production	NBSC (1981-2013a)	5 kg NH_3/ton	Stephen et al. (2004)
Waste disposal			
Wastewater	NBSC (2003-2013b),	0.003 kg NH_3/m^3	EPBG (2005)
Landfill	Du et al. (2006)	0.560 kg NH_3/ton	Stephen et al. (2004)
Compost		1.275 kg NH_3/ton	Stephen et al. (2004)
Incineration		0.210 kg NH_3/ton	Sutton et al. (2000)
Traffic			
Light-duty gasoline vehicles	CAAM (1983-2013)	0.023 g NH_3/km	Liu et al. (2014)
Heavy-duty gasoline vehicles	CAAM (1983-2013)	0.028 g NH_3/km	Stephen et al. (2004)
Light-duty diesel vehicles	CAAM (1983-2013)	0.04 g NH_3/km	Stephen et al. (2004)
Heavy-duty diesel vehicles	CAAM (1983-2013)	0.017 g NH_3/km	Stephen et al. (2004)
Motorcycles	CAAM (1983-2013)	0.007 g NH_3/km	Stephen et al. (2004)
Ammonia escape	CAEPI (2013)	2.3 mg/m^3	NEA (2011)

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

	Synthetic Fertilizer	Agricultural Soil	N-fixing Crop	Compost	Livestock	Biomass Burning	Human Excrement	Chemical Industry	Waste Disposal	Traffic	Ammonia Escape	total
1980	2,103	175	20	42	2,862	214	362	61	5	7		5,851
1981	2,077	175	19	43	2,888	214	368	60	5	8		5,858
1982	2,368	175	20	48	3,010	220	375	62	6	8		6,290
1983	2,616	175	18	51	3,028	219	383	68	7	9		6,574
1984	2,812	175	17	54	3,111	223	389	74	7	10		6,872
1985	2,686	175	18	52	3,257	218	397	70	8	12		6,893
1986	2,880	175	18	54	3,403	226	405	71	7	14		7,252
1987	3,015	175	18	57	3,509	267	413	82	8	16		7,559
1988	3,349	174	17	56	3,693	231	420	83	9	18		8,050
1989	3,562	174	17	57	3,799	224	430	87	10	20		8,381
1990	3,474	174	17	63	3,872	234	432	89	17	21		8,395
1991	3,861	174	16	63	3,908	234	435	92	28	23		8,835
1992	3,808	174	16	63	4,011	234	438	96	36	27		8,902
1993	3,803	173	18	66	4,259	237	442	93	43	32		9,166
1994	4,007	173	19	65	4,672	236	424	106	45	37		9,783
1995	4,329	173	17	67	5,170	242	404	113	57	40		10,613
1996	4,720	174	15	74	5,330	255	377	130	60	43		11,177
1997	4,528	174	16	72	4,844	246	353	126	69	47		10,476
1998	4,391	174	16	76	5,055	255	327	132	75	51		10,553
1999	4,331	174	19	76	5,120	257	309	139	80	56		10,562
2000	3,797	237	21	76	5,349	249	283	146	96	63	0.02	10,317
2001	3,835	237	22	69	5,391	278	271	154	92	53	0.04	10,403
2002	3,957	237	21	69	5,527	308	269	171	97	81	0.07	10,738
2003	3,692	237	22	65	5,783	310	253	173	103	94	0.09	10,733
2004	3,683	237	21	70	5,970	324	234	203	111	105	0.12	10,958
2005	3,492	237	21	76	6,159	303	209	232	110	122	0.12	10,962
2006	3,319	237	21	77	5,867	313	200	238	113	160	0.29	10,545
2007	3,258	222	19	79	4,992	305	195	258	128	165	0.60	9,621
2008	3,105	221	20	79	5,024	306	185	264	140	191	0.96	9,536
2009	3,244	221	20	84	5,202	315	169	277	152	231	1.77	9,917
2010	2,967	221	20	85	5,104	309	182	271	168	284	3.14	9,654
2011	2,804	221	19	91	4,928	326	131	296	186	335	4.98	9,342
2012	2,811	221	18	95	5,026	332	121	308	268	388	86.63	9,674

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

	Base year	Total	Synthetic Fertilizer	Husbandry	Biomass burning	Others
Zhao and Wang (1994)	1990	13.6/8.4	6.4/4.0	4.2/3.9		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			5.5/5.3		
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.

Figure captions

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.

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

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).

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

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).

Figure 6. ~~Comparison of total NH_3 emissions between this study and REAS.~~ Monthly distribution of NH_3 emissions from different sources in China: (a) all the sources; (b) synthetic fertilizer; (c) livestock wastes; (d) crop burning in fields; (e) forest and grass fires.

~~Figure 7.~~Figure 7. Comparison of total NH_3 emissions between this study and REAS.

Figure 8. Comparison of NH_3 emissions from synthetic fertilizers between this study and REAS.

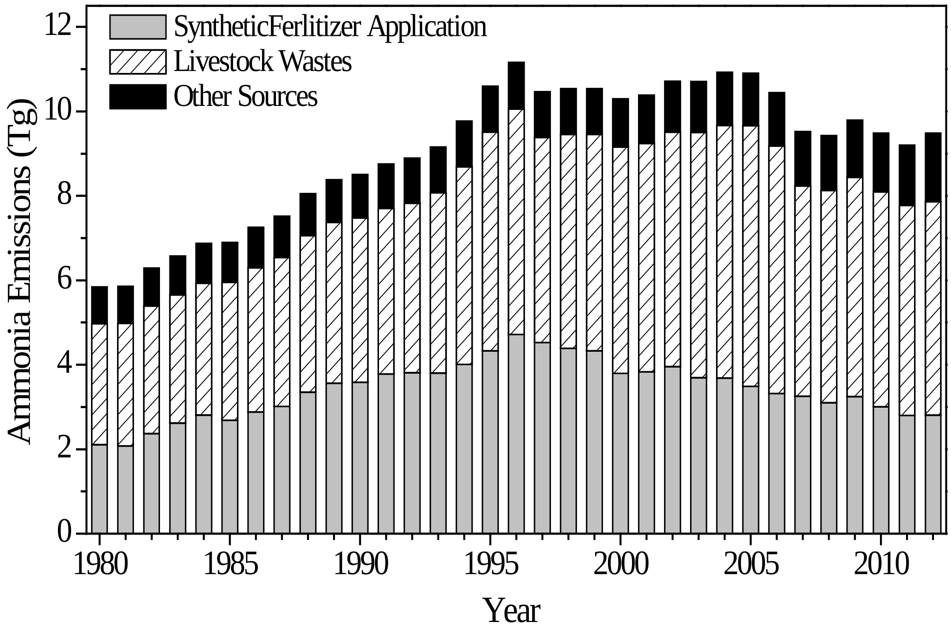
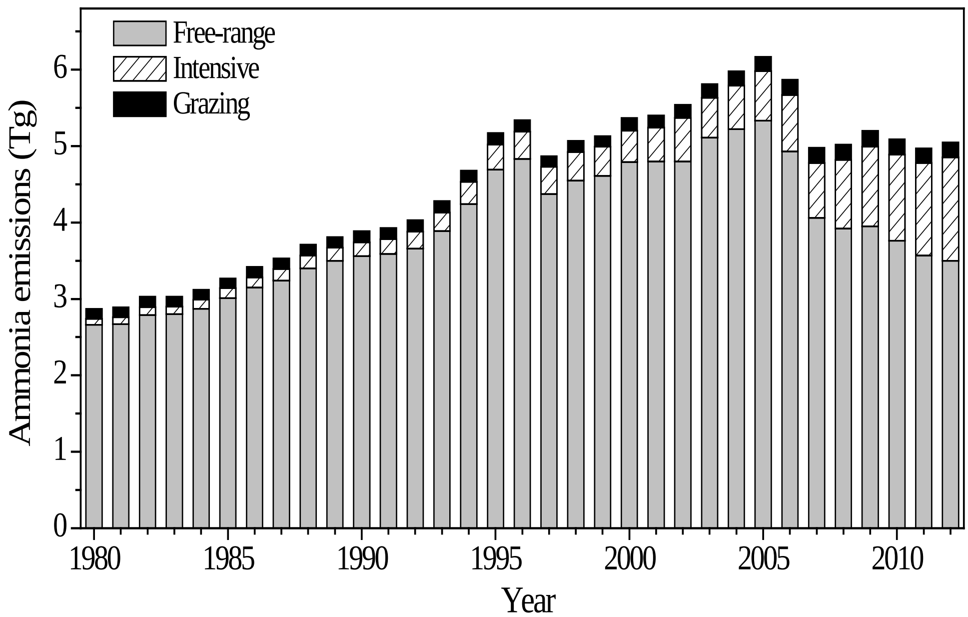


Figure 1. Interannual variation in total NH_3 emissions in China from 1980 to 2012; the sources of the emissions were categorized as synthetic fertilizer application, livestock manure, and other sources.



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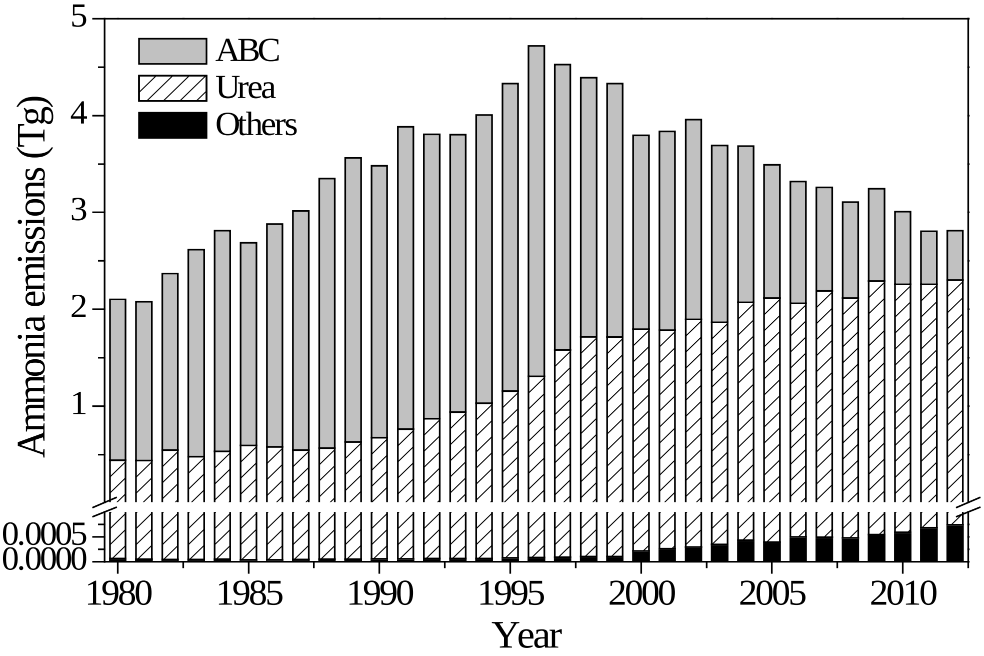

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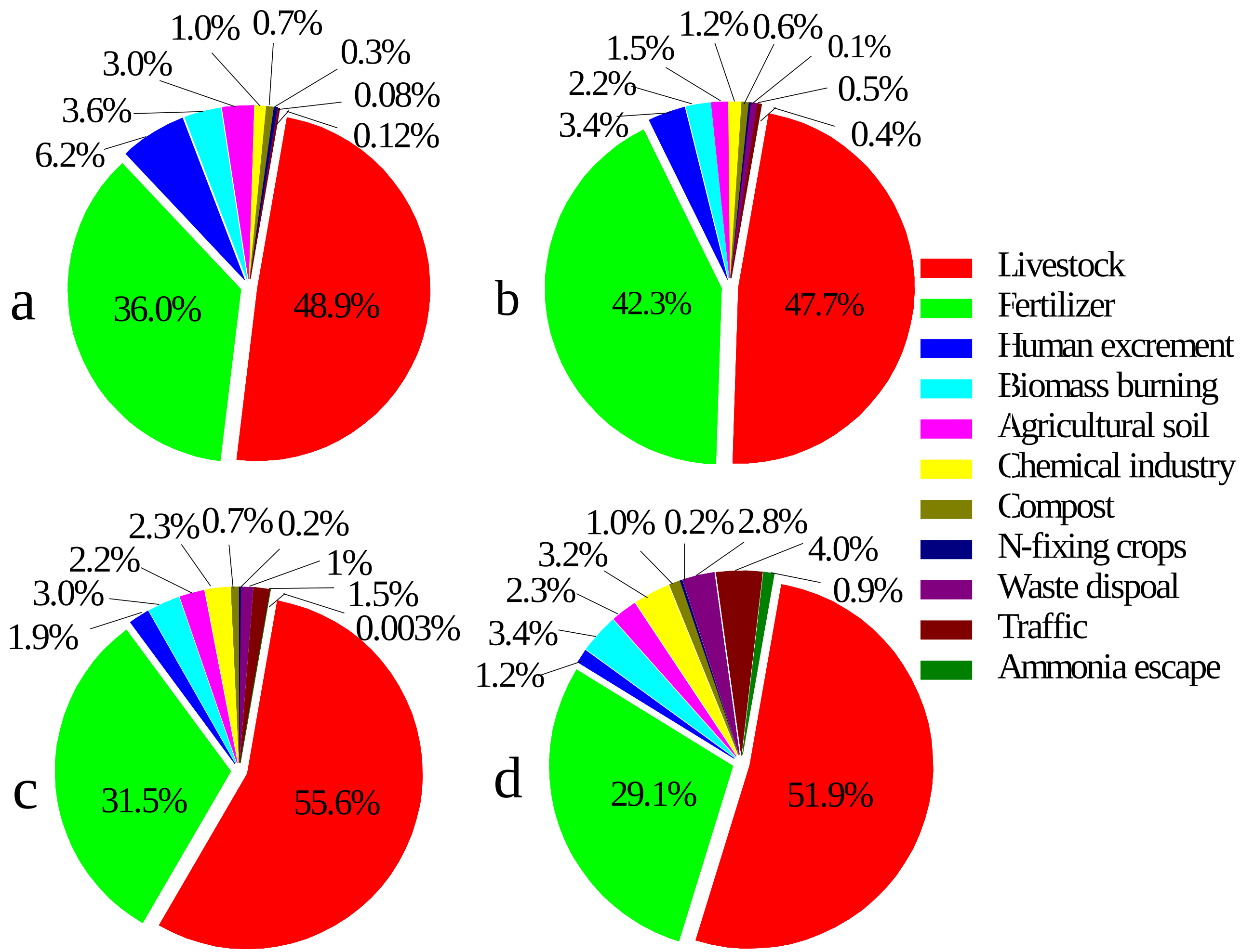
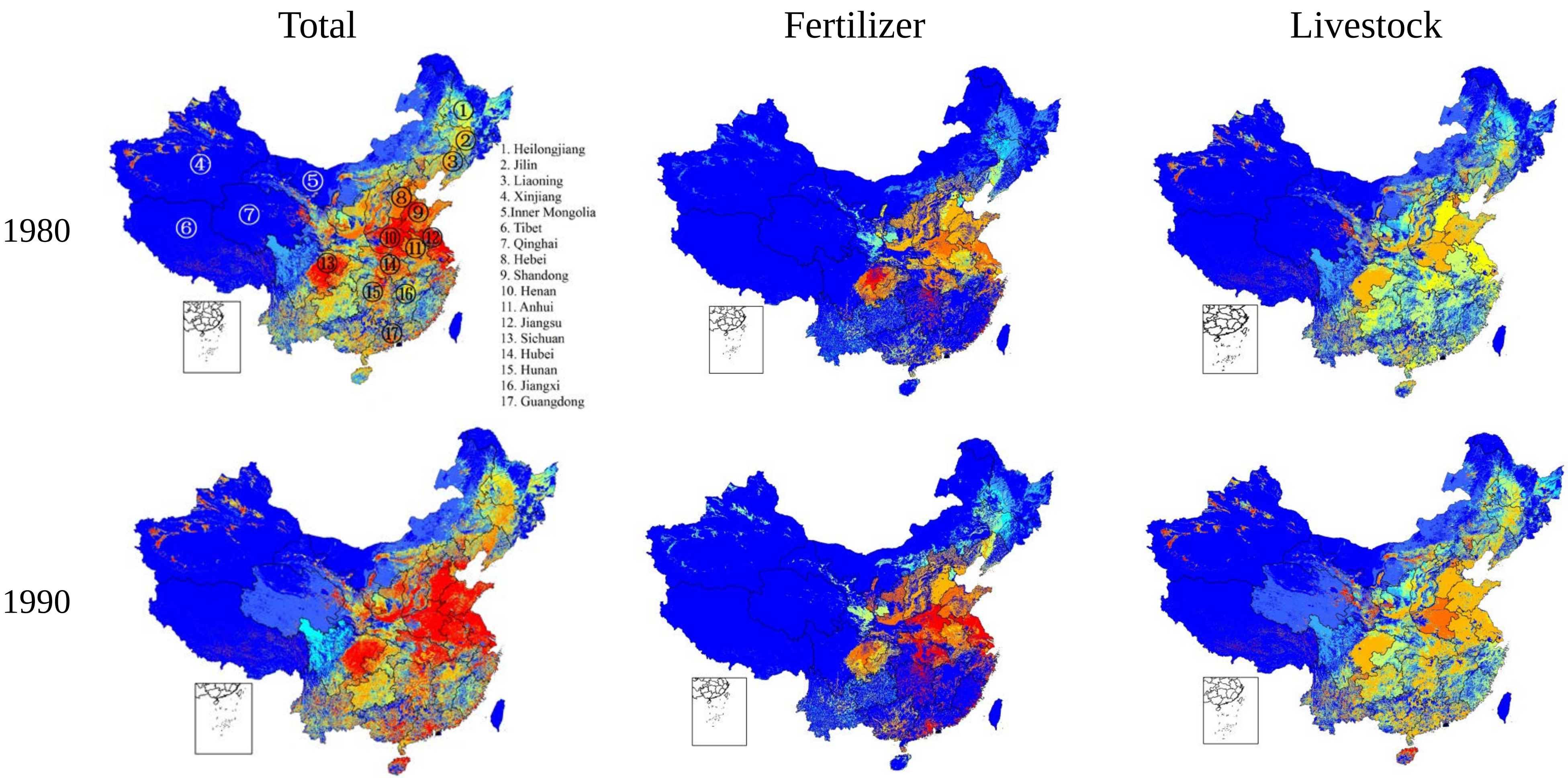


1009

1010 Figure 4. Source contributions (%) to NH_3 emissions in China: (a) 1980; (b) 1996; (c) 2006; (d) 2012.


Total
Fertilizer
Livestock
1980
1990
1. Heilongjiang
2. Jilin
3. Liaoning
4. Xinjiang
5.Inner Mongolia
6. Tibet
7. Qinghai
8. Hebei
9. Shandong
10. Henan
11. Anhui
12. Jiangsu
13. Sichuan
14. Hubei
15. Hunan
16. Jiangxi
17. Guangdong

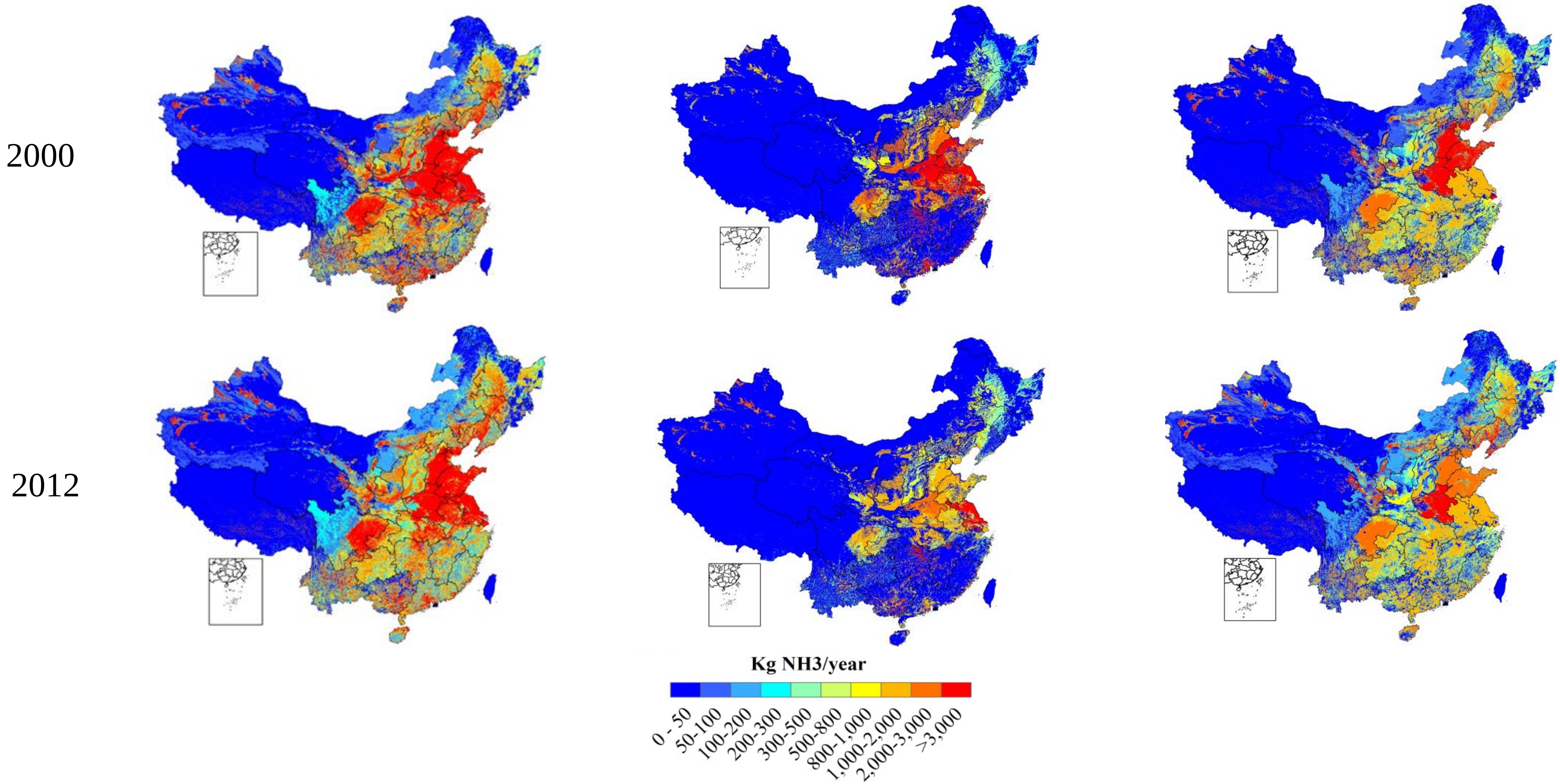
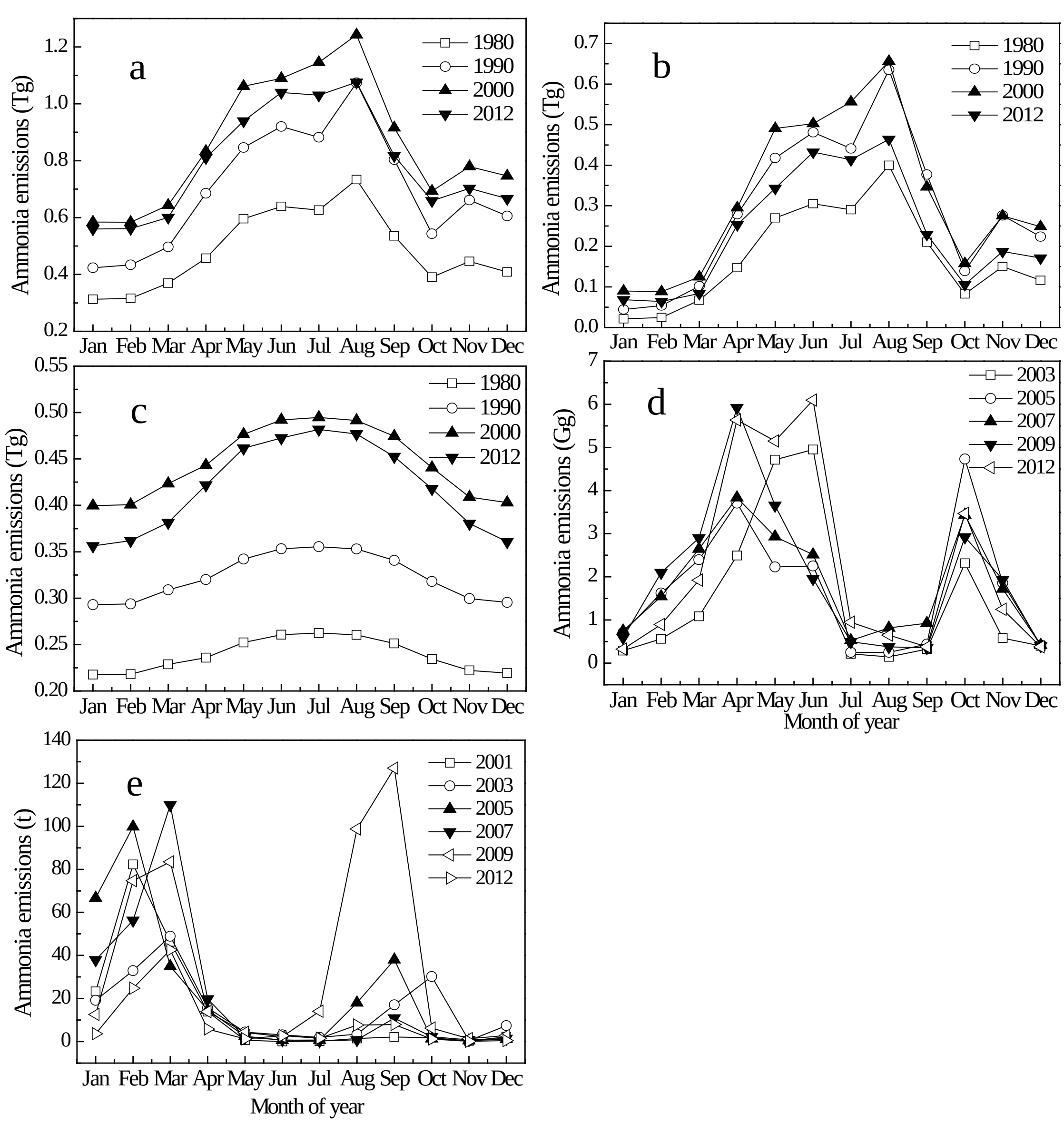


Figure 5. Spatial distribution of ammonia emissions in 1 km grid cell in 1980, 1990, 2000 and 2012 (from left to right: total emissions, synthetic fertilizer emissions and livestock emissions)



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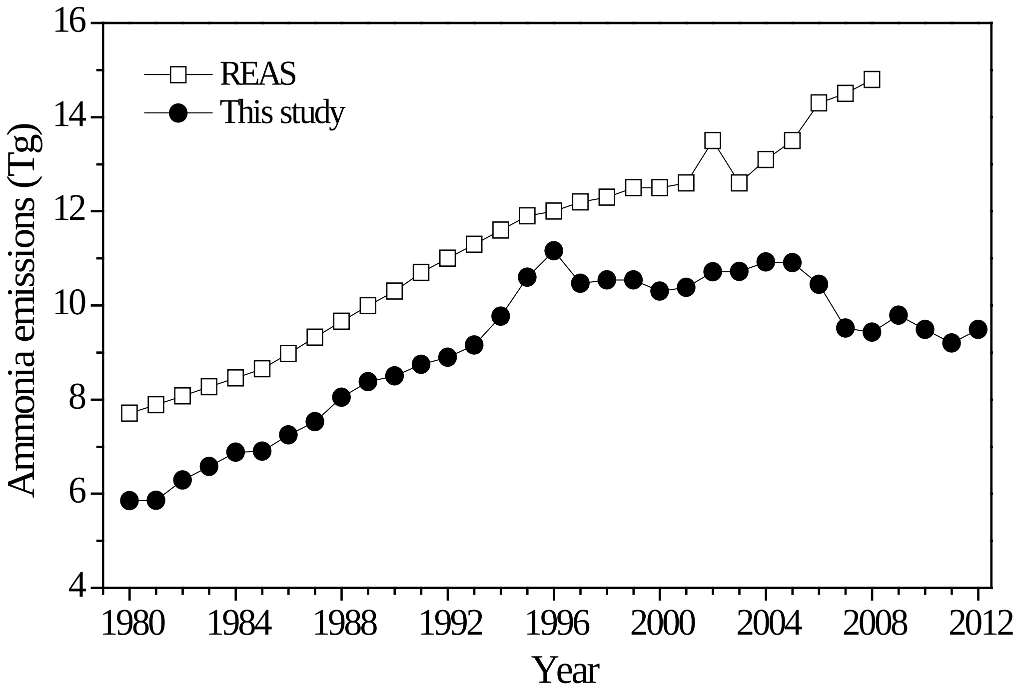

Figure 7. Comparison of total NH_3 emissions between this study and REAS.

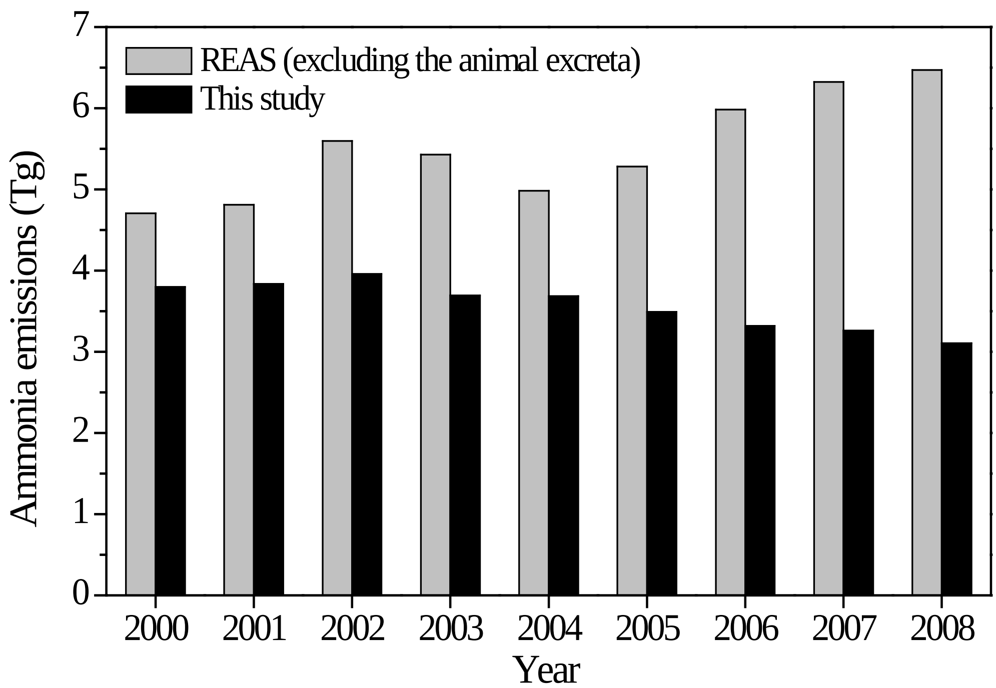

Figure 8. Comparison of NH_3 emissions from synthetic fertilizers between this study and REAS.