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Non-agricultural ammonia emissions in urban China

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Abstract

The non-agricultural ammonia (NH_3) emissions in cities have received little attention but could rival agricultural sources in term of the efficiency in PM formation. The starting point for finding credible solutions is to comprehensively establish a city-specific Non-agricultural Ammonia Emission Inventory (NAEI) and identify the largest sources where efforts can be directed to deliver the largest impact. In this paper, I present a NAEI of 113 national key cities targeted on environmental protection in China in 2010, which for the first time covers NH_3 emissions from pets, infants, smokers, green land, and household products. Results show that totally 210 478 Mg, the NH_3 emissions from traffic, fuel combustion, waste disposal, pets, green land, human, and household products are 67 671 Mg, 56 275 Mg, 44 289 Mg, 23 355 Mg, 7509 Mg, 7312 Mg, and 4069 Mg, respectively. The NH_3 emission intensity from the municipal districts ranges from 0.08 to 3.13 $\mathrm{Mg\,km^{-2}\,yr^{-1}}$, with a average of 0.84 $\mathrm{Mg\,km^{-2}\,yr^{-1}}$. The high NH_3 emission intensities in Beijing-Tianjin-Hebei region, Yangtze River Delta region and Pearl River Delta region support the view that non-agricultural NH_3 sources play a key role in city-scale NH_3 emissions and thus have potentially important implications for secondary PM formation (ammonium-sulfate-nitrate system) in urban agglomeration of China. Therefore, in addition to current SO_2 and NO_x controls, China also needs to allocate more scientific, technical, and legal resources on controlling non-agricultural NH_3 emissions in the future.

1 Introduction

Ammonia (NH_3) is the most abundant atmospheric base with the ability to neutralize sulphuric and nitric acids (oxidation products of SO_2 and NO_x ($NO+NO_2$), respectively) to form secondary particulate matter (PM) with an aerodynamic diameter equal to or less than 2.5 µm ($PM_{2.5}$) in the form of $(NH_4)_2SO_4$, NH_4HSO_4, and NH_4NO_3 (Seinfeld and Pandis, 2012). Current ambient air quality standards in many developed countries and some developing countries covered $PM_{2.5}$ in the book because of its adverse effects on human health, including premature mortality, chronic bronchitis, hospital admissions, and asthma attacks (Dockery et al., 1993; Nel, 2005; Pope et al., 2002; van Donkelaar et al., 2010). NH_3, NO_x and SO_2 emissions reduction is the key to lower ambient levels of $PM_{2.5}$ (Megaritis et al., 2013; Pathak et al., 2009). Major world economies have targeted reducing NO_x and SO_2 emissions. However, there are no regional emission ceilings set for NH_3 (with the exception of the EU27 levels for European countries arising from the Gothenburg protocol, Reis et al., 2012) despite the fact that control technologies are cost-effective compared to NO_x and SO_2 (Pinder et al., 2007). China has over-fulfilled the national goal of a 10 % reduction in SO_2 emissions from 2005 to 2010 by 14.3 %, and NO_x emissions are planned to be cut by 10 % during the 12th Five-Year Plan (2011–2015). Therefore, NH_3 is expected to play an increased role in $PM_{2.5}$ formation during the coming years (Chang et al., 2012).

It is well known that agricultural sources, notably animal manure and fertilizer application, contribute the most to NH_3 emissions (Cui et al., 2013; B. Gu et al., 2012). However, researchers have found that there are a myriad of important but frequently overlooked anthropogenic non-agricultural activities (e.g., vehicles and landfill) contributing to NH_3 emissions (Battye et al., 2003; Pierson and Brachaczek, 1983; Sutton et al., 2000, 2008; Wilson et al., 2004), and many countries do not report emissions for all these terms. In the UK, the non-agricultural emissions of NH_3 accounts for around 15 % of the total national NH_3 emissions, in which the transport sector is the main source (16 %), followed by sewage emissions (12 %) (Dragosits et al., 2008; Sutton et al., 2000). This clearly indicates that the emissions and sources of NH_3 deserve a more comprehensive discussion in scientific community. Although compared to dominant agricultural NH_3 source sectors such as livestock operations, these non-agricultural sources form a small part of the global NH_3 emissions (Bouwman et al., 1997), and they might be more locally concentrated, particularly at an urban level. Moreover, those strong rural NH_3 emissions can hardly make a long-range transport in gaseous phase to influence urban areas unless reacting locally to form particulate NH_4^+.

The non-agricultural NH_3 emissions in an urban SO_2- and NO_x-rich atmosphere can be fully neutralized (Behera and Sharma, 2010; X. Huang et al., 2011), while model results showed that over half of NH_3 emissions would be deposited downwind of its source within 10 km depending on local meteorological conditions. Meanwhile, the remaining part of NH_3 in rural areas has less chance to convert to particulate NH_4^+ (Asman et al., 1998). Various studies worldwide suggest that the ambient levels of NH_3 concentration in urban areas are comparable with, or even higher than that of rural areas (Doyle et al., 1979; Cadle et al., 1982; Allen et al., 1989, 2011; Giroux et al., 1997; Perrino et al., 2002; Burgard et al., 2006; Li et al., 2006; Whitehead et al., 2007; Alebic-Juretic, 2008; Cao et al., 2009; Shen et al., 2009; Tanner, 2009; Behera and Sharma, 2010; Bishop et al., 2010; Ianniello et al., 2010; Gong et al., 2011; Meng et al., 2011; Pandolfi et al., 2012; Reche et al., 2012; Ye et al., 2011; Zbieranowski and Aherne, 2012). In addition, the high seasonal variability of agricultural activities in rural areas tends to make pulse emissions of NH_3, but the situation is much better for the case of non-agricultural activities. These evidences mentioned above supporting a hypothesis that the non-agricultural NH_3 emissions contribute to $PM_{2.5}$ formation in urban areas may outweigh the contribution of agricultural NH_3 emissions in a scale of a full year.

China, historically a nation of mostly agriculture activity, is in the vanguard of a wave of urban expansion that is driving the country towards an economic superpower. In the course of intense expansion, China urbanized nearly half of its people in 2010 (49.95 %) compared with 20 % in 1980 (Gong et al., 2012). Beijing, for example, with over 10 million migrant workers and a similar size of local citizens already, is a living experiment in urbanization – and one that is failing to shine largely because of its severe air pollution (Watt, 2005; Zhang et al., 2012). In fact, alarm about the perilous state of $PM_{2.5}$ pollution in Beijing has provoked a huge amount of public outcry and media attention lately (Chang, 2012). Beijing is however by no means unusual in today's China, it is over 50 % cities in China have serious air pollution, and more than 75 % of the urban population are exposed to high concentrations of both primary and secondary PM that does not meet the Chinese NAAQS (national ambient air quality standards)

(Shao et al., 2006). Over the past several years, China has implemented a portfolio of plans to run more on-road vehicles with renewable energy, phase out its coal-fired power plants, and raise standards for auto emissions (Chang et al., 2012). If all goes to plan, China could expect a substantial emission reduction of $PM_{2.5}$ and its precursors (mainly NO_x and SO_2) in the coming years. However, current benefits might be partially offset in the absence of the non-agricultural NH_3 control (Chang et al., 2012).

An inventory of NH_3 emission can serve as a baseline toward tracking emission trends, developing mitigation strategies, and assessing progress. Besides, it is necessary to provide detailed inventories combined with spatial mapping of emissions as inputs to atmospheric transport models. It has been estimated that the global NH_3 emission was about 54 Tg in 1990, 70 % of which was related to food production (Olivier et al., 1998; Pinder et al., 2007). There are currently over 10 national emission inventories of NH_3 in China (Sun and Wang, 1997; Wang et al., 1997, 2009; Klimont et al., 2001; Streets et al., 2003; FRCGC, 2007; Dong et al., 2010; Cao et al., 2011; B. Gu et al., 2012; Huang et al., 2012; Li and Li, 2012; Cui et al., 2013), which providing strong evidence that China has experienced a dramatic increase of NH_3 since the late 1970s (Fig. S1). Several regions with dense population such as the North China Plain (Zhang et al., 2010; Zhao et al., 2012), the Yangtze River Delta (Fu, 2009; C. Huang et al., 2011) and the Pearl River Delta (Yin et al., 2012; Zheng et al., 2012) are the NH_3 hotspots. However, previous studies were mainly focused on the agricultural sector with large scales, such as global, Asian, national and regional inventories. As few studies involved transportation, waste disposal, human breath and sweat, etc. (Cao et al., 2011; Huang et al., 2012; Zheng et al., 2012), an inventory of city-scale NH_3 emissions covering all non-agricultural sources is clearly missing. As a consequence, past endeavours have failed to adequately reflect the overall NH_3 emissions status for individual cities, and failed to identify all the sources and activities that are responsible for NH_3 emissions, subsequently hindering target setting for future management or abatement.

In this paper, I develop a comprehensive non-agricultural NH_3 inventory for 113 Chinese cities based on statistical data in the year 2010. The emission sources included seven main categories, with each main category including several subcategories (Table 1). The activity data is based on province- or city-specific statistical data sets, with the exception of the population of pets, which was deduced from urban residents according to a constant proportion. The emission factors (EFs) were derived from a wide range of literature, some of which have been revised to be more representative of the situation in China.

2 Materials and method

2.1 Domain of the study

The Ministry of Environmental Protection of China (MEP) selected 113 key “cities” for environmental protection since 2003, and their daily concentrations of PM_{10}, SO_2, and NO_2 are published on the MEP official website (http://datacenter.mep.gov.cn/). The 113 big cities are selected in terms of both population and economy, and the administrative area of these cities is at or above the prefecture level. In addition to their administrative centres in the urban districts, much of the administrative areas (e.g., counties) are rural. Therefore, it is not appropriate to select the entire administrative area since the assessment of non-agricultural activities is merely on the urban area. In this sense, the municipal district areas of the 113 cities were chosen as the domain of current study (Fig. 1). In 2010, the 113 cities totally accounted for 2.6 % (250 600 km^2), 17.4 % (238 million), 39.8 % (15 827 billion RMB) of China’s land area, population and GDP, respectively. Table S1 presents detailed information regarding the social and economic index for each city.

2.2 Emission inventory estimate

An emission inventory is generally calculated as a product of activity data and its corresponding emission factor. For the purposes of this study, the term "non-agricultural sources" excludes emissions from the agricultural sector (e.g., fertilizer application, livestock operations), as well as natural sources (e.g., soils, wild animal populations). The primary census dataset was mostly obtained from several official yearbooks, which is currently the most authoritative data source in China. The main categories, subcategories and their representative EFs are listed in Table 1.

2.2.1 Traffic

The over-reduction of NO_x by three-way catalytic converters (TWCs) in vehicles has been shown to produce NH_3 in addition to N_2 and water (Cadle et al., 1982; Fraser and Cass, 1998; Kean et al., 2000), and these are fitted to all newly manufactured light-duty vehicles in China (Huo et al., 2009). In 2009, China has surged past the United States to become the world's largest automobile market – including buses, trucks and the small commercial vans that powered much of previous year's growth – rose 46.2 % to 13.6 million units. The NH_3 emissions from vehicles for each city were calculated as the product of the number of operating vehicles, mileage and ammonia emissions per kilometre for each vehicle type. The original data on the provincial population of vehicles in 2010 were obtained from China Automotive Industry Yearbook 2011 (http://tongji.cnki.net/kns55/Navi/YearBook.aspx?id=N2011090109&floor=1). In this work, these vehicles were divided into five different types, and the annual total mileages were based on previous work (Che et al., 2009). The EFs were quoted from the Emission Inventory Improvement Program (EIIP) database (Roe et al., 2004), which have been previously validated in the context of China (Huang et al., 2012; Zheng et al., 2012; Chao et al., 2014). Given that the important role of traffic to urban ammonia emissions, data like the composition of vehicle fleet, annual total mileages, and oil usages were also verified through various

first-hand investigations. Although NH_3 tailpipe emissions have been subject to a good deal of attention, the largest unknown is the ammonia emission factor for future catalyst systems. Recent technology has tried to reduce the effect (e.g., Tables 3–16 in http://www.eea.europa.eu/publications/emep-eea-guidebook-2013/part-b-sectoral-guidance-chapters/1-energy/1-a-combustion/1-a-3-b-road-transport). Thus, it is possible that ammonia emissions would decrease even further for new catalysts.

2.2.2 Waste treatment

Although the municipal waste treatment such as sewage treatment, landfill, compost and incineration of solid waste are known to emit NH_3, the EFs of them are highly uncertain. For example, the NH_3 EFs of wastewater treatment used in existing inventories vary widely, ranging over several orders of magnitude (Table S3).

Using the relative high EF ($1.93\,g\,m^{-3}$), two research teams provided independent emissions estimates for wastewater treatment in their recent inventories of NH_3 emissions for the Pearl River Delta region (Zheng et al., 2012) and the entire China (Huang et al., 2012), respectively. An intensive investigation of NH_3 EFs for municipal wastewater treatment in China has been conducted in a more recent study, and this study adopted their result ($0.28\,g\,NH_3\,m^{-3}$) in this study (Y. Gu et al., 2012). But then again, one thing should be noted is that using the highest factor to calculate emissions from wastewater treatment still produced an insignificant amount of NH_3 emissions comparing to agricultural sources.

2.2.3 Humans

The proportion of Chinese living in urban areas is close to 50 % mark in 2010 (49.95 %), yet this story overlooks China's special Hukou or household registration system, which may affect the real population living in urban/rural areas. This system divides the population into two types, agricultural and non-agricultural, under which peasants are stringently barred from acquiring an urban registration even though they are allowed to go and work in the city. In 2010, nearly 460 million out of 666 million Chinese urban dwellers had urban registration status (Chan, 2012). The remaining 206 million "floating population", mostly migrant workers (130 million) who seek opportunities in manufacturing centres such as Guangdong and Shanghai, are far less likely to receive pensions, basic health care, and unemployment insurance (Chan, 2012). In brief, there are three different types of population in urban China: resident population, registered population and migrant population.

Human metabolic processes such as respiration, perspiration and excretion can emit NH_3 directly (Lee and Dollard, 1994). In rural China, human excrement is an important ammonia emitter. However, this source was not considered in the current study due to the popularity of flush toilets in urban areas, which means that the resident population waste was discharged into the sewage system instead of being returned to the soil. However, it is noted that the feces and urine in infant nappies do not enter the sewage system and need to be taken into account. The 2010 infant population is the product of the registered population and the birth rate in this year. I calculated human NH_3 emissions of human sweat and breathe by multiplying the resident population and the individual EF. The infant population is the product of the registered population and the birth rate. The NH_3 EFs from infant nappies, human sweat and breath were taken from Sutton et al. (2000).

Cigarette smoking has been proved as a minor source of NH_3. China continues to be the largest producer and consumer of tobacco worldwide. In 2010, an estimated 28.1 % of adults (301 million people with age ≥ 15 years) in China, 26.1 % in urban areas (refers to the registered population here) and 29.8 % in rural areas (refers to the migrant population here) (Li et al., 2011). Of all current smokers, 85.6 % smoked daily. Smokers in China consumed an average of 14.2 cigarettes per day, which is comparable with that in the UK (16 and 14 cigarettes for men and women in 2005, respectively) (Li et al., 2011). The EF for smoking was also taken from Sutton et al. (2000).

2.2.4 Fuel combustion

The USEPA developed the Emissions Inventory Improvement Program (EIIP) guidance for estimating NH_3 emissions from industrial sources, combustion sources, and miscellaneous sources (Roe et al., 2004). Zheng et al. (2012) introduced EIIP in developing a NH_3 emissions inventory including domestic, power plant, and industrial fuel combustion sources in the Pearl River Delta region. Ruling out the traffic fuel consumption, the current inventory of fuel combustion sources is in accordance with the work done by Zheng et al. (2012).

2.2.5 Urban land cover

Little information is available on NH_3 emissions from urban green land. However, green land, especially artificial grassland in urban areas needs nitrogen fertilizer to sustain it. In China, according to a report, an average of $200\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$ been applied in urban grassland (Zhang, 2002). In addition, a previous study recommended a volatilization rate of 2.5 % NH_3-N of applied N (Sutton et al., 2000). When combined with the 2.5 % NH_3-N loss and 200 kg N for grassland fertilizer use, an annual NH_3 EF of $6.1\,\mathrm{kg\,NH_3\,ha^{-1}\,yr^{-1}}$ was obtained for China's urban grassland. The data of urban grassland were from China's City Yearbook 2011 (http://tongji.cnki.net/kns55/Navi/YearBook.aspx?id=N2012020070&floor=1).

Although still recognized as a luxury sport by most Chinese people, at present, there is a growing popularity for playing golf in China. The turf grass of golf course typically needs $200–400\,\mathrm{kg\,N\,ha^{-1}\,yr^{-1}}$ as N fertilizer to achieve high performance (Wong et al., 1998, 2002; Zhang, 2002). In 2010 alone, there were 60 new golf facilities opened, expanding the total number of the golf facilities in the country to 395 (equivalent to 490 18-hole courses) (Wang, 2011). According to a survey, every 18-hole equivalent golf course in China had 56.8 ha turf grass on average to be maintenance; this was 43.3 % larger than that of the US (Wang, 2011). Given that the turf grass data for each golf course is not available, this work does not consider the contribution of golf grassland to NH_3 emissions.

2.2.6 Pets

Keeping pets has become increasingly popular in China, especially in the cities where a growing number of people live alone and find it comforting to have a pet at home. As the two most popular pets in China, over 200 million dogs and 100 million cats were raised in 2010 (Yang, 2010). In recent years, there have been problems with unscooped poop and prevalent rabies, so municipal governments in Beijing and Shanghai have therefore proposed a one-dog policy limiting families to one canine per household. Officials revealed that as of 2010 there were about 0.92 and 0.74 million dogs registered in Beijing and Shanghai, respectively, and another 0.5 and 0.6 million dogs that were unlicensed. Unlike dogs, cats do not require a license and their number in cities is currently unavailable. By one estimate there are 0.5 to 0.6 million cats in Beijing (Pets in China, 2011). Information on the population of pets in other cities is extremely sparse in China; however, pet statistics in Beijing and Shanghai, to a large extent, reflect the situation of other cities concerning ownership of pets. Emissions from pets were estimated assuming that for every twenty-four city registered residents in China three dogs (1/8) and one cat (1/24) were owned (J.-F. Zhou, personal communication, a pet expert working at Gerson Lehrman Group, 2013). The NH_3 EFs of pets were derived from Sutton et al. (2000).

2.2.7 Domestic activity

Technically, the domestic sources include pet animal waste, human breath and sweat. In this work, the term “domestic sources” here refers merely to the use of household products and non-agricultural fertilizers. The EFs of these sources are taken from a recent report by Roe et al. (2004). Much of these data reported by Roe et al. came from

the studies in European and the US However, these EFs shown in Table 1 are thought to be reasonable for application in China until additional updated data are available.

3 Results and discussion

3.1 Non-agricultural NH_3 emission inventory in 2010

Using the methodology described above and the data collected, I evaluated the source-based non-agricultural NH_3 emissions and their corresponding contributions (Fig. 2). Taking the 113 cities as a whole, the overall non-agricultural NH_3 emissions in 2010 is estimated at 210 478 Mg, which accounts for about 5 % of the annual agricultural NH_3 emissions in China. A nationwide inventory on the non-agricultural NH_3 emission for all sources/cities is beyond the scope of this paper, therefore, this share is only one-third as much as the UK (15 %), and also lower than that in the EU27 (7 %) (Reis et al., 2009) and the USA (9 %) (Aneja et al., 2001). In the case of emission sources, traffic sources were the highest-profile contributor, accounting for about 32.2 % (67 671 Mg). Fuel combustion, waste disposal, pets, green land, human and household products were responsible for 26.7 % (56 275 Mg), 21 % (44 289 Mg), 11.1 % (23 355 Mg), 3.6 % (7509 Mg), 3.5 % (7312 Mg) and 1.9 % (4069 Mg) of the total non-agricultural NH_3 emissions, respectively (Fig. 2a). It is evident that nearly 60 % of NH_3 emissions originated from traffic and fuel combustion, which can be explained by the large population of vehicles equipped with catalytic converters as well as high energy consumption density in urban China. A more obvious example might be the USA, where on-road traffic is second to livestock manure management and application of chemical fertilizers, comprising approximately 7 % of the national NH_3 emission inventory (Burgard et al., 2006; Reis et al., 2009). Without involving NH_3 in China’s auto emission standards, it is expected that the sharing of vehicular NH_3 emissions in the nation’s NH_3 emission inventory will be increased steadily. In addition, as the first initial assessment of pets and green land in China, the result indicates that ignoring their contribution of

NH_3 emissions may lead to around a 15 % underestimate of the total non-agricultural NH_3 emissions. Totalling approximately 9.8 Tg on the base year of 2006, in which non-agricultural sources (chemical industry, waste disposal and traffic) jointly contributed over 4 %, Huang et al. (2012) provided an unparalleled high-resolution (1 km × 1 km) NH_3 emission inventory in China. However, Huang et al. (2012) excluded the contribution of fuel combustion, waste disposal, pets, humans, green land and household products, which are collectively responsible for 46.9 % of the total urban NH_3 emissions in current study.

A full and detailed inter-comparison of the non-agricultural NH_3 emission inventory on sectoral level in different cities is presented in Fig. 2b and Table 2. As illustrated in Fig. 2, the overall amount and the contributing proportion of the seven categories for each city show quite different patterns, which reflect their own unique socioeconomic profile. Despite the large variations of the contributing proportion from one city to another, traffic, fuel combustion and waste disposal are consistently the majority of the 47.3 % to 93.1 % of the non-agricultural NH_3 emissions in most of these cities. It is estimated that the average NH_3 footprint, i.e. the NH_3 emission intensity, from all the municipal districts of the 113 cities reached 0.84 $Mg\,km^{-2}\,yr^{-1}$. This indicator to date has also been regarded in the previous studies, and is summarized in Table 1. In 2010, 67 and 8 out of the 113 cities exceeded the lower and upper limit of NH_3 emission intensity estimate (0.7–2.3 $Mg\,km^{-2}\,yr^{-1}$) from Manchester city centre, respectively (Table S2). Domestically, estimates of the non-agricultural NH_3 emission intensity in this study are in line with those reported in Beijing, Shanghai and Nanjing (the provincial capital of Jiangsu), but significantly higher than that from Guangzhou (the provincial capital of Guangdong). The main reason for the estimated gap is attributed to the use of a different study domain in the two emission inventories in addition to the different activity data, emission factors and base years used. Although the non-agricultural sources covered in this study are estimated to be a small contributor to the national annual inventories, Table 3 indicates that at a city scale, the percentage of the non-

agricultural NH_3 emissions to the agricultural NH_3 emissions in Beijing and Shanghai could reach 27 %, 43 %, respectively.

3.2 Geographical distribution

Figure 3 displays the spatial distributions of the 113 cities' non-agricultural NH_3 emissions for different emissions sources (Fig. 3a–g) and the total emissions (Fig. 3h). The spatial patterns of NH_3 emission intensity can be found in Fig. 4. Generally, the spatial variability for all sources is in agreement with China's socioeconomic landscape, i.e., owing to a higher urbanization and larger population density in eastern and southern China, NH_3 emissions from non-agricultural sources such as traffic, fuel combustion and others tend to be higher than those in the nation's remote northwest and southwest. Besides, Fig. 3e reveals that the bulk of urban green land emissions are contributed from the south, particularly in Guangzhou, Shanghai, Shenzhen and Chongqing, where there is a monsoon-influenced humid subtropical climate with abundant rainfall throughout the year. The favourable climate and fertile soil make south China known for its high plant diversity. In contrast, the fuel combustion from the long-lived heating systems and energy-intensive industry amplifies the NH_3 emissions in north China (Fig. 3b).

The atmospheric behaviour of NH_3 characterized by short lifetime, near-source deposition, highly sensitive to meteorology and fast gas-to-particle conversion rate, highlights the need to improve NH_3 emission estimate with fine temporal and spatial resolution (Behera and Sharma, 2010). Unlike the agricultural NH_3 sources, the non-agricultural NH_3 emissions originate from a variety of stationary sources (industrial coal/oil/gas combustion, wastewater, landfill, compost and incineration), mobile sources and area sources (e.g., humans, green land, domestic fuel combustion). Data such as the location/capacity/number of stationary sources are far from well documented in China. Therefore, a gridded emissions inventory of non-agricultural NH_3 sources was not introduced in current work. The problem of data scarcity can be easily resolved for a city case study; however, it is nearly an impossible task for me to collect all allocation-needed data for the 113 cities. Besides, given the city scale this work used, the spatial distribution of non-agricultural NH_3 sources in any city cannot be distinguished on the map of China. Nevertheless, those results based on city-scale datasets are still valuable for chemical transport modellers who focus on a specific Chinese city.

3.3 Uncertainties of the inventory

Ammonia emission inventories have inherent uncertainties because they associated with both the activity data and the emission factors used. For this inventory, activity data were mostly from statistical information. Although some question census data in Chinese statistical yearbooks are manipulated, with a tendency for officials to overstate economic outcome. However, compared with GDP data, the officials has less intention to manipulate the items that current work used. Therefore, the manipulation was not a serious issue during the study period and the data used in this study was credible. Nevertheless, the detailed pets and vehicle activity data (number of vehicles in each category, mileage, and oil usage of vehicles) was rarely collected and reported in China's statistics, which may introduce some uncertainties.

The EFs in current inventory are lack of the real physical and chemical examination. Moreover, the EFs show a substantial variation among different studies, and China has not established an adequate non-agricultural ammonia emission factors database yet. Therefore, the major uncertainties of this inventory stem from the EFs this study used. Figure 5 illustrates the lower, the best and the upper estimates for the seven non-agricultural ammonia sources of the 113 key cities, respectively. The lower and the upper estimates are derived from the lowest and the highest EFs available in the published literature (Table S3). The overall estimated emission range was 140 421–336 811 $Mg\,yr^{-1}$, corresponding to the uncertainties of −33–60 %. In terms of the potential change in mass of ammonia emissions over the entire domain, the uncertainty for traffic and fuel combustion represents 38 475 and 38 438 $Mg\,NH_3\,yr^{-1}$. From a standpoint of variability, the humans and domestic activity category represents one of the most important sources of uncertainty.

A better characterization of the timing of climate-dependence non-agricultural NH_3 sources, such as humans, pets, and urban land cover could be expected to reduce the uncertainty of seasonal NH_3 emissions estimates. However, these three sources contribute less than 10 % of the total non-agricultural NH_3 emissions. Moreover, it is easy to understand that the range of variation for dominant non-agricultural NH_3 sources, such as traffic and fuel combustion, is much lower than agricultural NH_3 sources (e.g., fertilizer application, livestock operations). Therefore, the seasonal differences of non-agricultural NH_3 emission inventory are not been given in current work.

3.4 Future recommendation and outlook

China is now facing the world's worst air quality in terms of $PM_{2.5}$ and its precursors (e.g., SO_2, NO_x and NH_3) pollution. Some control measures such as upgrading motor vehicle emission standards to the China National Standard IV or higher, switching vehicle fuel to lower sulfur levels of gasoline in urban areas had been implemented in order to achieve a significant reduction in NO_x and SO_2 emissions. However, in Europe, based on a source-attribution simulation work, Megaritis et al. (2013) revealed that the reduction of NH_3 emissions was the most cost-effective control strategy for reducing $PM_{2.5}$ in both winter and summer. Besides, in the eastern United States, Pinder et al. (2007) conducted a series of PMCAMx simulations and found many currently available NH_3 control technologies were cost-effective compared to SO_2 and NO_x. In China, using response surface modeling technique, Wang et al. (2011) suggested that the 90 % increase of NH_3 emissions during 1990–2005 resulted in nearly 50–60 % increases of NO_3^- and SO_4^{2-} aerosol concentrations in East China. Furthermore, Wang et al. (2013) used GEOS-Chem to investigate the influence of precursor emission changes on SNA aerosols concentration changes from 2000 to 2015, finding that the benefit of SO_2 reduction would be completely offset if NH_3 emissions were allowed to keep their recent growth rate over China. China's national annual total NH_3

emissions account for around 30 % and, 16 % of total Asia, and global NH_3 emissions, respectively. Obviously, in addition to current SO_2 and NO_x controls, China needs to put more scientific, technical, and legal attention on controlling NH_3 emissions.

Most experimental studies and inventory-based assessment of NH_3 emissions has been and is being given to agricultural and natural sources (Liu et al., 2013; Paulot and Jacob, 2014). Compared to dominant agricultural sources of NH_3 emissions, the non-agricultural sources covered in this study are estimated to contribute small amounts to national annual total NH_3 emissions inventory. However, a diverse range of non-agricultural sources, such as on-road traffic and waste treatment could expect to dominate in an urban domain, where agricultural sector contributions are relatively small. Existing NH_3 emissions inventories do not include emission estimates for all non-agricultural sources included in this study, thus significantly underestimate the NH_3 emissions in urbanized areas. Here I suggest that China should improve the understanding of current NH_3 emission estimate and put more eyes on controlling non-agricultural sources of NH_3 emissions in the future.

The efficiency of NH_3 emissions in producing inorganic aerosol is highly non-linear and may vary from urban area to urban area. This work still falls short of providing an answer to how much of ammonium salts in the urban areas derives from non-agricultural vs. agricultural NH_3 emissions, and how sensitive urban PM levels is to changes in local emissions. Therefore, it could be useful to run some source-receptor matrices (data from many model simulations and each of which reduces a different source of emissions) incorporating with the improved emission estimates in the vicinity of different Chinese cities to test the efficiency factors of different ammonia sources on particulate formation.

The use of three-way catalytic converters to reduce emissions of NO is considered to be an important factor in attaining present and future vehicle emissions standards and air quality goals. However, an unwanted side effect has been an increase in NH_3 emissions. Until recently, motor vehicles were not recognized to be a significant source of NH_3. In this paper, it is clear that the traffic sectors, especially the light-duty gasoline

vehicles in urban China are a major source of NH_3 emissions compared to the other non-agricultural sources. Therefore, future NH_3 control efforts in urban China should prioritize the traffic sector, and this is especially true for China's top three city clusters, i.e., Beijing-Tianjin-Hebei region, the Yangtze Delta region, and the Pearl River Delta region. NH_3 generation from petrol-engined vehicles is primarily catalyzed by platinum. The capacity for NH_3 generation can be modified by changing the catalyst or combination of catalysts used, though there are inevitably compromises to be reached given that performance has to be measured against a range of different pollutants (Handley et al., 2001). For the other sources, it is assumed that there are no reasonable options for controlling ammonia emissions from pets, human sweat and breath, cigarette smoking and nappies.

Because NH_3 is not currently included as a criteria air pollutant in China and there are no government announced daily NH_3 monitoring data, which limited the ability to validate the accuracy of emission inventory. The lack of monitoring data also emphasizes the need for publicly available comprehensive information systems in order to support the environmental evaluation of non-agricultural NH_3 in urban China. Besides, some EFs derived from foreign studies would imply a significant uncertainty between different cities across China, which is challenging to validate and interpret. A recommendation is that the emission inventory for non-agricultural NH_3 in China should be reevaluated as more local emission factors from China becomes available.

4 Summary

In this work, I presented a comprehensive non-agricultural NH_3 emission inventory for 113 Chinese cities. This work illustrates the possible under-estimation of NH_3 in some inventories, and can be expected to initiate wider discussion on methodologies for estimation of non-agricultural emissions. The source-based NH_3 emissions details will help guide development strategies that are economically and environmentally optimal, providing the basis for policy makers to determine how to prioritize future control efforts among different non-agricultural sources of NH_3 emissions in urban China.

Supplementary material related to this article is available online at http://www.atmos-chem-phys-discuss.net/14/8495/2014/acpd-14-8495-2014-supplement.pdf.

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Table 1. Representative ammonia EFs of non-agricultural sources.

Category	Subcategory	Emission factor	Unit (as NH_3)	Reference
Traffic	Light-duty gasoline vehicles	63.20	mg km^{-1}	Roe et al. (2004)
	Heavy-duty gasoline vehicles	28.00	mg km^{-1}	Roe et al. (2004)
	Light-duty diesel vehicles	4.20	mg km^{-1}	Roe et al. (2004)
	Heavy-duty diesel vehicles	16.80	mg km^{-1}	Roe et al. (2004)
	Motorcycles	7.00	mg km^{-1}	Roe et al. (2004)
Waste treatment	Wastewater	0.28	g m^{-3}	Y. Gu et al. (2012)
	Landfill	0.56	kg t^{-1}	Yin et al. (2012)
	Compost	1.28	kg t^{-1}	Roe et al. (2004)
	Incineration	0.21	kg t^{-1}	Yin et al. (2012)
Humans	Human breath	3.64	g person^{-1} yr^{-1}	Sutton et al. (2000)
	Human sweat	17.00	g person^{-1} yr^{-1}	Sutton et al. (2000)
	Infants (0–3 yr)	16.64	g infant^{-1} yr^{-1}	Sutton et al. (2000)
	Smoking	21.61	g smoker^{-1} yr^{-1}	Sutton et al. (2000)
Fuel combustion	Industrial coal combustion	0.02	kg t^{-1}	Roe et al. (2004)
	Domestic coal combustion	0.90	kg t^{-1}	Roe et al. (2004)
	Industrial oil combustion	0.10	kg (10^3 L)$^{-1}$	Roe et al. (2004)
	Domestic oil combustion	0.12	kg (10^3 L)$^{-1}$	Roe et al. (2004)
	Industrial gas combustion	51.30	kg (10^6 m^3)$^{-1}$	Roe et al. (2004)
	Domestic gas combustion	320.51	kg (10^6 m^3)$^{-1}$	Sutton et al. (2000)
Urban landcover	Green land	6.10	kg ha^{-1} yr^{-1}	Sutton et al. (2000), Zhang (2002)
Pets	Dogs	0.74	kg animal^{-1} yr^{-1}	Sutton et al. (2000)
	Cats	0.13	kg animal^{-1} yr^{-1}	Sutton et al. (2000)
Domestic activity	Household products	17.10	g person^{-1} yr^{-1}	Roe et al. (2004)
	Non-agricultural fertilizers	36.90	g person^{-1} yr^{-1}	Roe et al. (2004)

Table 2. Summary of non-agricultural ammonia estimates (in Mg) by city in 2010*.

City	TR	WD	HM	PE	FC	GL	HH	Total
Beijing	4909.7	4013.6	313.5	1152.9	2257.1	376.3	200.9	13 224.0
Tianjin	1817.7	966.8	214.4	788.1	1264.6	106.0	137.3	5294.7
Shijiazhuang	790.3	368.4	65.4	238.3	1117.5	50.2	41.5	2671.6
Tangshan	1038.1	483.9	82.7	301.3	2012.5	53.3	52.5	4024.2
Qinhuangdao	216.1	100.7	26.7	81.1	482.7	25.2	14.1	946.6
Handan	548.7	255.7	47.7	144.7	539.6	37.2	25.2	1598.8
Baoding	476.2	221.9	34.4	104.3	500.4	27.2	18.2	1382.6
Taiyuan	714.0	272.6	89.5	279.9	2026.8	47.0	48.8	3478.6
Datong	105.0	40.9	48.6	151.8	300.3	24.0	26.5	697.0
Yangquan	115.7	45.1	21.6	67.5	375.6	11.0	11.8	648.3
Changzhi	108.4	42.3	21.9	68.5	317.9	12.3	11.9	583.3
Linfen	85.2	33.2	26.1	81.8	288.8	7.3	14.2	536.6
Hohhot	436.7	292.7	36.6	116.6	2002.4	16.1	20.3	2921.4
Baotou	576.1	386.2	43.5	138.9	3093.4	43.0	24.2	4305.2
Chifeng	254.4	170.6	37.4	119.2	636.9	15.5	20.8	1254.8
Shenyang	1127.0	1057.8	149.2	502.8	1446.9	158.6	87.6	4529.9
Dalian	1158.5	1087.4	88.0	296.4	1236.2	74.4	51.6	3992.5
Anshan	469.3	447.8	42.9	144.5	439.5	34.1	25.2	1603.3
Fushun	197.7	188.7	40.5	136.5	229.0	27.3	23.8	843.3
Benxi	190.0	181.3	27.8	93.8	207.4	29.1	16.3	745.8
Jinzhou	201.5	192.2	27.2	91.7	160.7	15.8	16.0	705.1
Changchun	853.1	469.6	106.6	355.6	993.2	70.6	62.0	2910.7
Jilin	461.3	253.9	54.5	181.7	399.5	39.7	31.6	1422.1
Harbin	964.2	620.8	141.4	465.9	1300.6	74.3	81.2	3648.4
Qiqihar	231.6	149.1	42.3	139.4	200.4	29.8	24.3	816.8
Mudanjiang	201.3	129.6	23.8	78.4	115.1	30.9	13.7	592.8
Shanghai	1581.4	3369.3	404.0	1307.1	3139.6	713.3	227.7	10 742.4
Nanjing	1056.7	545.1	168.0	535.9	1344.7	471.1	93.4	4214.9
Wuxi	1192.3	615.1	73.3	233.7	317.3	101.0	40.7	2573.4
Xuzhou	605.5	312.4	57.3	182.8	137.9	63.6	31.8	1391.3
Changzhou	116.2	247.6	69.7	222.5	243.4	41.3	38.8	979.6
Suzhou	1900.2	980.3	73.9	235.8	354.3	79.3	41.1	3664.8

Table 2. Continued.

City	TR	WD	HM	PE	FC	GL	HH	Total
Nantong	713.8	368.3	65.1	207.6	134.8	23.2	36.2	1549.0
Lianyungang	245.6	126.7	27.3	87.1	46.8	22.0	15.2	570.6
Yangzhou	458.8	236.7	37.5	119.7	95.3	20.9	20.9	989.8
Zhenjiang	398.7	211.2	31.8	101.5	96.7	39.0	17.7	896.7
Hangzhou	1784.2	899.8	133.5	421.5	506.7	95.7	73.4	3914.7
Ningbo	1548.0	780.7	68.9	217.7	330.5	52.6	37.9	3036.5
Wenzhou	877.1	442.3	45.0	142.1	145.6	20.8	24.8	1697.7
Huzhou	390.8	197.1	33.7	106.6	62.7	20.5	18.6	829.9
Shaoxing	838.0	422.6	20.2	63.7	74.1	21.1	11.1	1450.9
Hefei	665.0	360.3	67.6	204.7	818.3	64.3	35.7	2216.0
Wuhu	272.9	147.9	34.0	103.0	342.6	30.4	17.9	948.7
Maanshan	199.6	108.1	20.6	62.4	268.0	29.6	10.9	699.3
Fuzhou	676.3	393.8	60.0	183.9	231.8	46.5	32.0	1624.2
Xiamen	446.2	259.8	56.7	173.7	283.0	87.3	30.3	1336.9
Quanzhou	772.0	449.5	33.0	101.0	118.7	19.7	17.6	1511.6
Nanchang	467.4	354.3	73.0	218.4	311.2	45.3	38.0	1507.6
Jiujiang	218.6	165.7	20.9	62.6	92.2	24.4	10.9	595.4
Ji’nan	1090.0	508.6	110.7	341.8	771.3	66.9	59.5	2948.9
Qingdao	1580.5	737.4	87.5	270.4	860.8	97.6	47.1	3681.3
Zibo	799.5	373.0	88.6	273.6	610.4	89.8	47.7	2282.6
Zaozhuang	380.1	177.3	69.8	215.5	202.9	22.5	37.5	1105.7
Yantai	1234.2	575.9	57.0	175.9	468.2	55.1	30.7	2597.0
Weifang	861.8	402.1	57.6	177.9	224.3	47.0	31.0	1801.7
Jining	708.9	330.7	38.0	117.4	164.3	27.1	20.5	1406.9
Taian	572.3	267.0	50.6	156.3	178.2	23.8	27.2	1275.5
Rizhao	286.2	133.5	39.0	120.6	200.0	18.4	21.0	818.7
Zhengzhou	1037.8	592.5	90.2	279.7	637.2	62.8	48.7	2749.0
Kaifeng	238.4	136.1	27.0	83.8	86.0	17.1	14.6	603.1
Luoyang	596.0	340.3	50.7	157.1	287.6	30.0	27.4	1489.0
Pingdingshan	336.8	192.3	32.2	100.0	176.9	13.1	17.4	868.8
Anyang	338.0	193.0	34.0	105.5	167.5	14.7	18.4	871.2
Jiaozuo	320.2	182.8	26.4	82.0	103.1	16.9	14.3	745.8
Sanmenxia	30.3	17.3	9.2	28.7	39.5	7.3	5.0	137.3

Table 2. Continued.

City	TR	WD	HM	PE	FC	GL	HH	Total
Wuhan	1052.5	915.3	158.3	505.5	2536.6	93.1	88.1	5349.2
Yichang	293.4	255.2	38.4	122.5	416.3	18.6	21.3	1165.6
Jingzhou	63.4	55.1	35.9	114.7	164.6	11.3	20.0	465.0
Changsha	912.9	705.8	78.1	236.5	613.2	49.6	41.2	2637.3
Zhuzhou	256.0	197.9	32.5	98.4	266.0	21.1	17.1	889.0
Xiangtan	179.4	138.7	28.4	86.1	268.6	21.3	15.0	737.5
Yueyang	309.1	239.0	28.4	85.9	278.6	23.9	15.0	979.8
Changde	299.5	231.5	45.6	138.0	279.0	17.4	24.0	1035.0
Zhangjiajie	48.6	37.6	16.2	49.0	51.7	7.9	8.5	219.4
Guangzhou	2840.3	1771.5	208.3	642.6	1204.9	759.0	112.0	7538.4
Shaoguan	179.9	112.2	29.3	90.4	45.8	18.9	15.7	492.3
Shenzhen	2531.5	1578.9	78.2	241.4	1177.6	587.9	42.1	6237.5
Zhuhai	319.8	199.4	32.7	100.8	170.3	31.8	17.6	872.3
Shantou	319.8	199.4	160.2	494.1	231.8	41.5	86.1	1532.8
Zhanjiang	370.8	231.3	48.3	149.0	92.6	67.9	26.0	985.8
Nanning	478.3	269.2	88.0	262.2	245.9	213.1	45.7	1602.4
Liuzhou	349.4	196.7	34.2	101.9	165.4	35.7	17.8	901.0
Guilin	293.2	165.0	25.0	74.4	68.9	13.9	13.0	653.4
Beihai	106.6	60.0	19.9	59.3	45.6	12.3	10.3	314.0
Haikou	184.8	118.2	52.5	155.3	120.8	22.0	27.1	680.7
Chongqing	662.9	1454.1	477.2	1514.3	298.8	564.0	263.8	5235.0
Chengdu	1696.4	1009.8	159.3	511.2	278.7	98.4	89.1	3842.9
Zigong	198.0	117.9	46.0	147.8	103.3	12.5	25.7	651.3
Panzhihua	160.2	95.4	21.2	67.9	147.9	12.3	11.8	516.7
Luzhou	218.5	130.1	44.5	142.8	93.0	17.8	24.9	671.6
Deyang	281.5	167.6	20.2	64.7	70.3	11.6	11.3	627.0
Mianyang	293.6	174.8	37.4	120.1	95.3	21.6	20.9	763.6
Nanchong	253.2	150.7	59.1	189.8	54.8	18.3	33.1	758.9
Yibin	266.3	158.5	24.5	78.6	56.3	10.7	13.7	608.5
Guiyang	390.3	283.2	71.5	214.8	1176.4	35.1	37.4	2208.7
Zunyi	316.2	229.4	27.9	83.7	375.3	14.5	14.6	1061.6
Kunming	1025.1	356.4	80.5	245.6	687.7	61.2	42.8	2499.3
Qujing	486.2	157.6	22.3	68.2	128.4	19.9	11.9	894.5

Table 2. Continued.

City	TR	WD	HM	PE	FC	GL	HH	Total
Yuxi	285.7	92.6	13.5	41.3	173.8	6.1	7.2	620.1
Lhasa	73.3	0.0	7.3	21.3	223.8	12.6	3.7	341.8
Xi'an	904.8	547.0	174.6	551.2	1121.9	73.6	96.0	3469.1
Tongchuan	52.3	31.6	23.5	74.3	68.1	9.6	12.9	272.4
Baoji	272.4	164.7	44.0	138.8	217.5	20.2	24.2	881.7
Xianyang	306.6	185.4	27.9	88.0	156.1	12.9	15.3	792.1
Weinan	223.8	135.3	30.1	95.0	67.8	8.3	16.6	576.8
Yan'an	247.2	149.5	14.0	44.0	62.6	5.5	7.7	530.4
Lanzhou	307.9	176.3	68.5	206.6	577.5	26.7	36.0	1399.3
Jinchang	58.9	33.7	7.0	21.1	102.5	4.0	3.7	230.9
Xining	192.4	158.7	37.8	112.0	229.3	8.3	19.5	758.0
Yinchuan	263.8	243.6	30.2	89.7	283.1	31.6	15.6	957.6
Shizuishan	99.4	91.8	15.0	44.6	570.7	37.2	7.8	866.4
Urumqi	431.5	318.1	78.4	227.6	1406.2	93.8	39.7	2595.2
Karamay	229.2	169.0	13.3	38.6	384.5	13.4	6.7	854.7
113 cities	67 670.8	44 287.8	7312.3	23 354.6	56 275.0	7508.8	4068.7	210 478.0

[*] TR, traffic; WD, waste disposal; HM, humans; PE, pets; FC, fuel combustion; GL, green land; HH, household.

Table 3. Comparison of current work with previous NH_3 emissions estimates.

City	Year	Non-agri emissions (Mg)	Agri emissions (Mg)	Non-agri emission intensity ($Mg\,km^{-2}\,year^{-1}$)	Source
Beijing, CN	2010	13 224		1.72	This study
Beijing, CN	2006	12 800	46 700	1.68	Huang et al. (2012)
Shanghai, CN	2010	10 742		2.08	This study
Shanghai, CN	2006	13 600	31 600	2.14	Fu (2009)
Guangzhou, CN	2010	7539		1.96	This study
Guangzhou, CN	2006	10 870		1.46	Zheng et al. (2012)
Nanjing, CN	2010	4215		0.89	This study
Nanjing, CN	2004	4390		0.93	Dong et al. (2009)
Manchester, UK	2004			0.70–2.30	Whitehead et al. (2007)

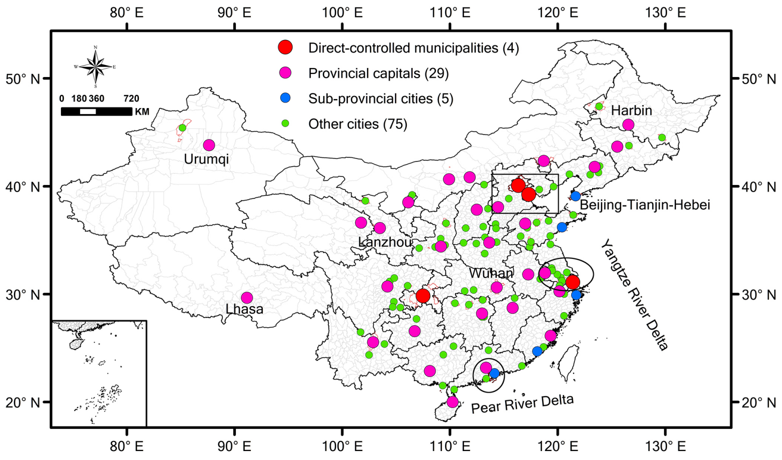

Fig. 1. Distribution of the 113 "key cities" in China and their classification.

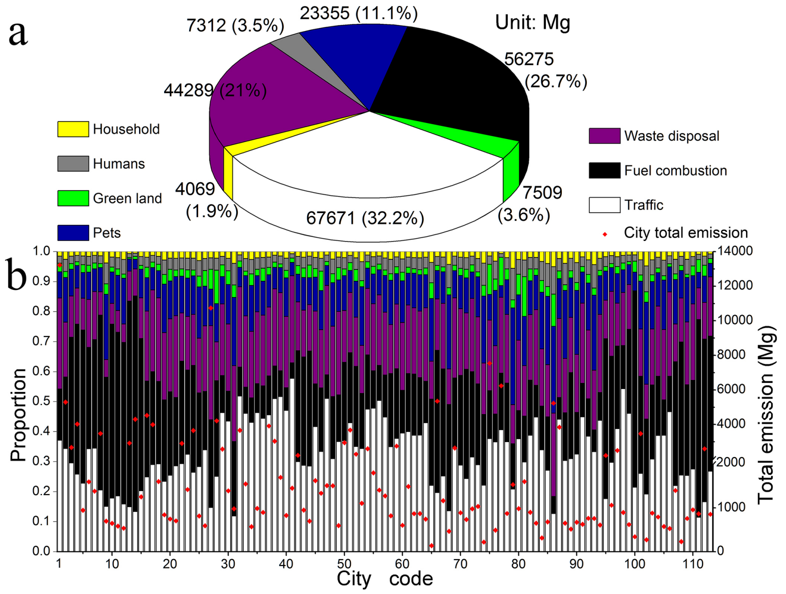

Fig. 2. Pie chart **(a)** and stack column **(b)** presenting the source-based non-agricultural NH_3 emission contributions of all the 113 cities and each specific city, respectively; x-axis **(b)**: every number representing a specific city (see Table S1), right y-axis **(b)**: red dot indicating the total non-agricultural NH_3 emissions of each city (set 2000 as the breakpoint), left y-axis **(b)**: stacked histograms showing the variations of contribution sources for each city.

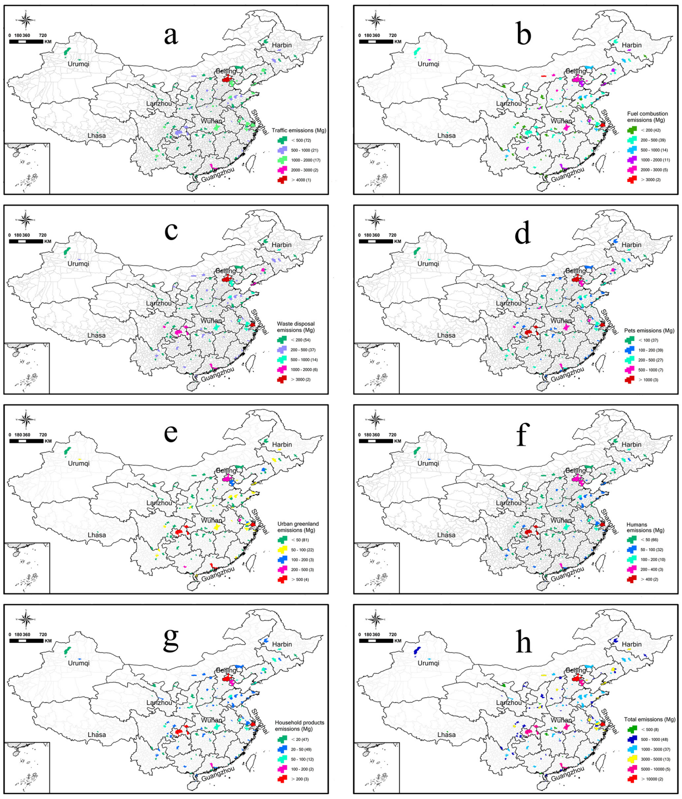

Fig. 3. Spatial distributions of non-agricultural NH_3 emissions by source category in China's 113 key cities (figures in brackets represent the number of cities).

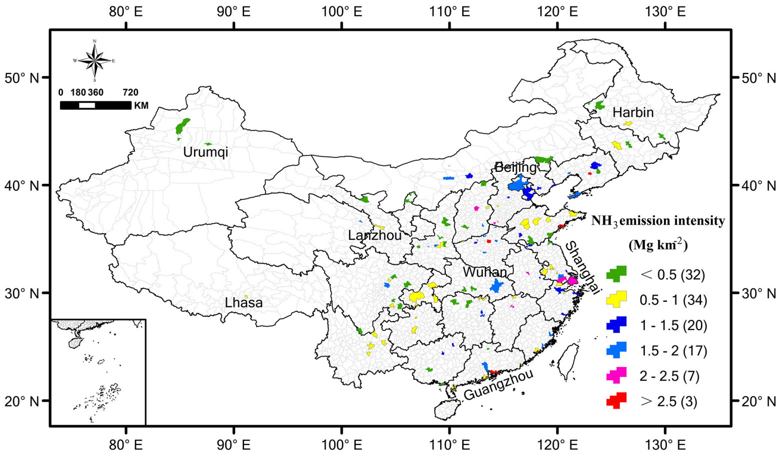

Fig. 4. Spatial patterns of NH_3 emission intensities in China's 113 key cities (figures in brackets represent the number of cities).

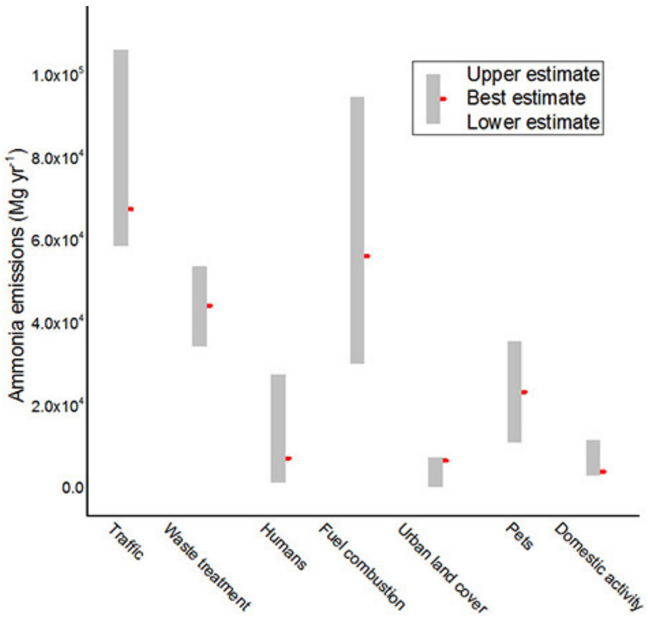

Fig. 5. Uncertainties estimates for the seven non-agricultural ammonia sources of the 113 key cities.