A high-resolution ammonia emission inventory in China

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[1] The existence of gas-phase ammonia (NH_3) in the atmosphere and its interaction with other trace chemical species could have a substantial impact on tropospheric chemistry and global climate change. China is a large agricultural country with an enormous animal population, tremendous nitrogen fertilizer consumption and, consequently, a large emission of NH_3. Despite the importance of NH_3 in the global nitrogen (N) cycle, considerable inaccuracies and uncertainty exist regarding its emission in China. In this study, a comprehensive NH_3 emission inventory was compiled for China on a 1 km $\times$ 1 km grid, which is suitable for input to atmospheric models. We attempted to estimate NH_3 emissions accurately by taking into consideration as many native experiment results as possible and parameterizing the emission factors (EFs) by the ambient temperature, soil acidity and other factors. The total NH_3 emission in China was approximately 9.8 Tg in 2006. The emission sources considered included livestock excreta (5.3 Tg), fertilizer application (3.2 Tg), agricultural soil (0.2 Tg), nitrogen-fixing plants (0.05 Tg), crop residue compost (0.3 Tg), biomass burning (0.1 Tg), urine from rural populations (0.2 Tg), chemical industry (0.2 Tg), waste disposal (0.1 Tg) and traffic (0.1 Tg). The regions with the highest emission rates are located in Central and Southwest China. Seasonally, the peak ammonia emissions occur in spring and summer.

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1. Introduction

[2] Ammonia (NH_3) is a very important alkaline constituent in the atmosphere, and it has a wide variety of impacts. A large part of atmospheric aerosols consist of sulfate neutralized to various extents by NH_3. NH_3 is a key precursor to the neutralization of HNO_3 and H_2SO_4 in the atmosphere to form the secondary inorganic aerosols $(NH_4)_2SO_4$, NH_4HSO_4 and NH_4NO_3, which contribute to ambient particulate matter [*Seinfeld and Pandis*, 2006], as shown in following chemical reaction equations:

$$2NH_{3(g)} + H_2SO_{4(g)} \rightleftharpoons (NH_4)_2SO_{4(s)} \tag{1}$$

$$NH_{3(g)} + H_2SO_{4(g)} \rightleftharpoons NH_4HSO_{4(s)} \tag{2}$$

$$NH_{3(g)} + HNO_{3(g)} \rightleftharpoons NH_4NO_{3(s)} \tag{3}$$

Measurements have indicated that the mass of secondary sulfate, nitrate and ammonium could account for 25–60% of the total $PM_{2.5}$ (fine particles in the ambient air, 2.5 μm or less in aerodynamic diameter) mass in China [*He et al.*, 2001; *Niu et al.*, 2006; *Fang et al.*, 2009]. Results from recent studies show that these fine particles can be deposited deep in the lungs, which may lead to increased morbidity [*Dockery et al.*, 1993]. Fine particles also can result in regional visibility degradation by light scattering [*Ye et al.*, 2011]. Global climate change due to ambient particulate matter has gained increasing attention in recent years. These aerosol particles have a significant effect on radiative forcing by both direct scattering and the absorption of solar radiation and modification of the shortwave reflective properties of clouds [*Charlson et al.*, 1992; *Martin et al.*, 2004], thereby exerting a cooling effect on the planet. In addition, NH_3, as an important alkaline gas in the atmosphere, can react readily with acidic substances and neutralize the acidity of precipitation. Gaseous ammonia and ammonium compounds in particles are eventually deposited to the soil or water bodies. NH_4^+ deposited to the soil may either be taken up by plant roots or be subjected to nitrification yielding NO_3^-, as shown in following equations. Both processes result in proton release into the soil [*Krupa*, 2003]. Consequently, ammonia emission is also known as one of the causes of soil acidification, eutrophication and even the perturbation of ecosystems [*Van Breemen et al.*, 1983; *Pearson and Stewart*, 1993].

$$NH_4{}^+ + 2O_2 \rightarrow 2H^+ + NO_3{}^- + H_2O \tag{4}$$

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Table 1. Activity Data for Various Sources

Activity Data Set and Its Description	Amount
Synthetic fertilizer application (synthetic fertilizer consumption)	22.4 Tg
Agriculture soil (cultivated areas)	1.2×10^8 hectare
Nitrogen-fixing crop (cultivated areas)	2.1×10^7 hectare
Compost of crop residues (mass composted)	235 Tg
Livestock wastes (animal population)	1.2×10^{10}
Biomass burning (burned biomass or burned area)	411 Tg burned biomass; 2118 km^2 burned area
Excrement of rural population (rural population using tatty latrine)	2.6×10^8
Chemical industry (production)	49 Tg synthetic ammonia; 39 Tg N nitrogen fertilizers
Waste disposal (waste amount)	2.0×10^{10} m^3 wastewater; 78 Tg solid waste
Traffic sources (vehicle population)	1.6×10^8

The most important ammonia emitters are livestock waste and fertilizer, which jointly contribute over 57% globally and over 80% in Asia [*Zhao and Wang*, 1994; *Bouwman et al.*, 1997; *Streets et al.*, 2003]. China is a country with dense rural populations, and it has the world's top-ranking agricultural production. Both of these factors contribute significantly to the global ammonia emissions budget. Previous researchers have demonstrated that approximately 14 Tg NH_3 is released in China annually, making it one of the most important emitters, with a contribution of 20% to the global budget and 55% of Asian emissions. What is worth mentioning, global ammonia column was mapped based on the observation of IASI sensor (Infrared Atmospheric Sounding Interferometer) onboard EUMETSAT's Meteorological Operational Satellite (MetOp-A), which also demonstrated that China is an important emitter [*Clarisse et al.*, 2009].

[3] There are several ammonia inventories for China [*Zhao and Wang*, 1994; *Olivier et al.*, 1998; *Xing and Zhu*, 2000; *Streets et al.*, 2003; *Ohara et al.*, 2007]. However, these inventories have weaknesses. First, they often use uniform and European-based EFs for the entire country. Ammonia volatilization, especially from nitrogen fertilizer application and animal waste management systems, depend strongly on soil acidity and ambient temperature [*Kirk and Nye*, 1991; *European Environment Agency* (*EEA*), 2009]. The EFs should be parameterized by such factors. Second, not all ammonia emission sources are included during estimation, such as vehicle exhaust and wastewater treatment. This omission could introduce inaccuracies in the estimation of total emissions. Third, the prevailing inventories often have coarse temporal and spatial resolutions. Agricultural activities such as fertilization and crop burning exhibit seasonal variations due to planting and harvesting practices. Ammonia volatilization also strongly depends on the temperature, which fluctuates seasonally. Hence, a temporal resolution of one year tends toward overestimation in winter and underestimation in summer. Spatially, the existing inventories are mostly compiled at coarse spatial resolutions, such as at the provincial level [*Wang et al.*, 1997], at $1° \times 1°$ [*Bouwman et al.*, 1997] and at a 30 min $\times$ 30 min grid [*Streets et al.*, 2003]. For a modeling study of the NH_3 cycle and air quality simulation, a more detailed inventory is preferred.

[4] A comparison of measured and predicted NH_4NO_3 concentrations also indicates that the existing inventories might be overestimated by approximately 20–75% in East Asia [*Kim et al.*, 2006]. Therefore, an accurate ammonia emission inventory is needed to reflect various sources with a fine spatial and temporal resolution for China, especially for air environmental quality simulation. In this study, a comprehensive ammonia inventory for the year 2006 is developed. The sources of ammonia considered include the following: (1) farmland ecosystem (soil and nitrogen-fixing plant emission, fertilizer application and crop residue compost), (2) livestock wastes (free-range, intensive and grazing rearing systems), (3) biomass burning (forest and grass fires, crop residue burning and fuel wood combustion), (4) excrement of rural populations; 5) chemical industry (ammonia synthesis and nitrogen fertilizer production), (6) waste disposal (wastewater and solid waste treatment), and (7) traffic sources. The activity data are from province-specific statistical data sets, with the exception of a MODIS burned area product (MCD45A1), which was used in the assessment of biomass burning emissions. The EFs are characterized by seasons and locations, in which the ammonia volatilization from nitrogen fertilizer and animal agriculture is parameterized by the ambient temperature, soil acidity and other crucial influential factors.

2. Data and Methods

[5] Ammonia emission is calculated as a product of activity data and corresponding EFs. The important sources and its activity data are listed in Table 1, more detailed information could be found in Table S1 in the auxiliary material.[1]

2.1. Farmland Ecosystem

[6] Farmland ecosystems are an important source of ammonia emission. These emitters include synthetic fertilizer application, agricultural soil, nitrogen-fixing plants and the compost of crop residues.

2.1.1. Synthetic Fertilizer Application

[7] In China, a large agricultural country with intensive agricultural areas and conventional cultivation practices, N fertilizers are applied extensively, with more than 20 Tg of N fertilizers applied every year [*National Bureau of Statistics of China* (*NBSC*), 2008a]. Recent studies have indicated that 10–30% of applied N is lost though NH_3 volatilization [*Ju et al.*, 2009]. NH_3 volatilization depends substantially upon fertilizer types. In China, urea and ammonium bicarbonate (ABC) are the two dominant N fertilizers, followed by ammonium nitrate (AN) and ammonium sulfate (AS).

[1]Auxiliary materials are available in the HTML. doi:10.1029/2011GB004161.

Table 2. EFs, Expressed as Percentage of Volatilized NH_3-N From Applied Fertilizer-N, and Correction Coefficients for Different N Fertilizer Categories

Fertilizer Categories	Measured EFs		Correction Coefficients		
	Acid Soil	Alkaline Soil	CF_{rate}[a]	CF_T[b]	CF_{method}[c]
Urea	8.8[d]	30.1[d]	1.18	0.35	0.32
ABC	18.2[e]	39.1[e]	1.18	0.44	0.32
AN	0.8[f]	0.8[f]	1.18	0.01	0.32
AS	2.3[g]	4.6[g]	1.18	0.06	0.32
Others	0.8[f]	0.8[f]	1.18	0.01	0.32

[a]Values are derived from *Li et al.* [2002], *Song et al.* [2004], and *Fan et al.* [2006].
[b]Values are derived from *Lu et al.* [1980] and *EEA* [2009].
[c]Values are derived from *Lu et al.* [1980], *Qu* [1980], *Fillery et al.* [1986], *Zhang and Zhu* [1992], *Li and Ma* [1993], and *Cai et al.* [2002].
[d]Measurement results of *Zhu et al.* [1989].
[e]Measurement results of *Cai et al.* [1986].
[f]Values recommended by *EEA* [2009].
[g]Measurement results of *Qu* [1980].

Urea and ABC accounted for approximately 69% and 26%, respectively, of the total chemical N fertilizer consumption in 2006 [*NBSC*, 2007c, 2007e]. Hence, the N fertilizers considered in our study are classified into five categories: urea, ABC, AN, AS and other (including calcium ammonium nitrate and ammonium phosphates). The loss rate strongly depends on numerous factors, including soil properties (pH, calcium content, water content, buffer capacity and porosity), meteorological conditions (temperature, wind speed and precipitation), time of application in relation to a crop canopy and the method of application [*Bouwman et al.*, 1997].

[8] In this study, ammonia emissions were estimated by multiplying consumption and gridded (1 km × 1 km) condition-specific EFs for five kinds of fertilizer as well as 16 major plants. The rates of fertilizer consumption were obtained as the product of cultivated area and the application rate of plants. The plants to which considerable amounts of N fertilizers are applied include early rice, semi-late rice, late rice, non-glutinous rice, wheat, maize, bean, potato, peanut, oil crop, cotton, beet, sugarcane, tobacco, vegetables and fruits [*NBSC*, 2007c, 2007e].

[9] The type of fertilizer, the soil properties, the fertilization method, the application rate and temperature were introduced as parameters to develop EFs for specific conditions. For each kind of fertilizer, a reference emission factor (EF_0), referring to existing native measurements, was adjusted by soil pH, application rate, temperature and fertilization method to develop a specific EF for a given scenario, as shown in following equation:

$$EF_i = EF_{0i} \times CF_{pH} \times CF_{rate} \times CF_T \times CF_{method} \tag{5}$$

where EF_i is the emission factor for a specific condition; EF_{0i} is the reference emission factor for a type i N fertilizer; CF_{pH} is the correction factor for different soil acidity; CF_{rate} is the correction factor for different application rates; CF_T is the correction factor for diverse temperatures; and CF_{method} is the correction factor for the fertilization method, including basal dressing and top dressing. The monthly and gridded EFs were developed considering all the factors. Ammonia emissions could then be calculated as the product of monthly fertilizer consumption and corresponding condition-specific EFs.

2.1.1.1. Fertilizer Type

[10] The potential ammonia emission from each fertilizer type often varies widely. ABC is a highly volatile compound that is extensively used in China. The ammonia loss from the direct use of ABC could be up to 30–40% of the applied N [*Zhu et al.*, 1989]. Urea, which is less volatile than ABC, is first converted to ammonium bicarbonate by the enzyme urease, which takes approximately 2–3 days in agricultural soil. Other N fertilizers, such as AN and AS, have a much lower ammonia emission potential (<10% of applied N) [*Sutton et al.*, 1995; *Van der Hoek*, 1998]. The EF_0 for urea and ABC were based on experiments carried out in Henan and Jiangsu Province through the micrometeorological method [*Cai et al.*, 1986; *Zhu et al.*, 1989]. The EF_0 for other less prevalent fertilizers refers to the up-to-date and reliable EFs provided by the European Environment Agency [*EEA*, 2009], as shown in Table 2.

2.1.1.2. Soil Property

[11] The volatilization of ammonia greatly depends on the soil properties, including bulk density, volumetric water content, CEC, soil acidity and urease activity. The results from the analysis of covariance demonstrate that the volatilization is most strongly correlated with pH [*Li and Ma*, 1993; *Corstanje et al.*, 2008]. Several publications have shown the disparity of ammonia volatilization from urea and ABC in different soils [*Cai et al.*, 1986; *Zhu et al.*, 1989]. For fertilizers such as AS, which form sparingly soluble salts with calcium and thus increase the dissolution of calcium carbonate, the emission rate will be greater in the alkaline soil, whereas for AN, the emission rate is nearly unaffected by the soil pH. Several studies support that ammonia loss progressively increases with an increase in soil pH, and linear regression was used to yield specific EFs in some studies [*Whitehead and Raistrick*, 1991; *Bouwman et al.*, 2002; *Fan et al.*, 2005; *Jones et al.*, 2007]. In our study, linear interpolation method was used to specify EFs by assuming that a linear relationship between ammonia volatilization and soil pH. A soil pH with a spatial resolution of 1 km in China was provided by the Harmonized World Soil Database (available from http://www.iiasa.ac.at/Research/LUC/External-World-soil-database/HTML/).

2.1.1.3. Application Rate

[12] The application rate was used as a modifier for EFs for various types of fertilizers. *Ju et al.* [2009] reported an increase in ammonia emission with increasing application rate. Experiments on the relationship between application rate and ammonia volatilization are limited. Correction was done only when application rate is relatively high. A value of 200 kg N ha^{-1} was considered the high rate, and a value of 1.18 for CF_{rate} was used for a province with a high application rate [*Li et al.*, 2002; *Song et al.*, 2004; *Fan et al.*, 2006]. For regions where the rate was lower than 200 kg N ha^{-1}, no correction was made.

2.1.1.4. Temperature

[13] As the partial pressure of NH_3 in solution increases exponentially with temperature, we would expect a strong relationship between temperature and ammonia emission. Several studies have noted an increase in emission from N fertilizers with increasing air temperature. *EEA* [2009] have summarized empirical volatilization rates as functions of

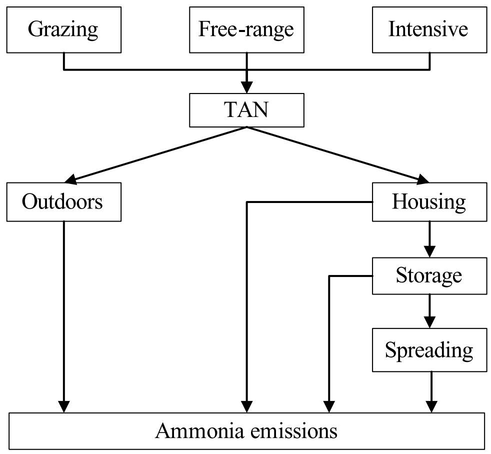

Figure 1. N flows in the manure management system and ammonia volatilization (TAN: ammoniacal nitrogen).

spring air temperature for various fertilizers except ABC. We derived a relationship of emission rate and temperature for ABC based on the available native experimental data [*Lu et al.*, 1980], as listed in Table 2. Monthly average temperatures for a 1 km grid were developed on the basis of NCEP (National Centers for Environmental Prediction) final analysis data set.

2.1.1.5. Fertilization Method

[14] The method of fertilization is another important factor affecting the volatilization of ammonia. We considered two common fertilization methods: basal dressing and top dressing. For the same fertilizer, the EF for basal dressing is approximately 32% of that for top dressing [*Lu et al.*, 1980; *Qu*, 1980; *Fillery et al.*, 1986; *Zhang and Zhu*, 1992; *Li and Ma*, 1993; *Cai et al.*, 2002]. The application time and amount of fertilizer used as basal dressing and top dressing are referenced from the Chinese planting information network (http://www.zzys.gov.cn/) and census data and investigation results [*NBSC*, 2007a, 2007e; *Wang et al.*, 2008].

2.1.2. Agriculture Soil

[15] Many organisms in soils are involved in the decomposition of organic matter excrete NH_3 directly or N compounds which could be readily hydrolyzed to NH_3. The NH_3 fluxes from soil are related closely to the biological activity in the soil and other meteorological factors [*Bouwman et al.*, 1997]. There is limited information on ammonia emissions from cultures without fertilizers in China. Background emission levels of ammonia from agricultural soil were reviewed by *Yan et al.* [2003], and an EF value of 1–2 kg NH_3-N ha^{-1} yr^{-1} was recommended as the loss of ammonia in temperate regions. The province-level area of arable land refers to the census data [*NBSC*, 2007e]. The provincial ammonia emission from agricultural soil was calculated as a product of arable land and EF.

2.1.3. Nitrogen-Fixing Crop

[16] Nitrogen-fixing crops, even if not given fertilizer-N, have been estimated to fix as much N as fertilized crops. Thus, emissions of NH_3 may be expected to be similar to those from fertilized agricultural crops. The most widely cultivated nitrogen-fixing plants in China are beans (mostly soybean, accounting 77% cultivated area), peanuts and green manure. The ammonia emissions may be approximately estimated as follows:

$$E_{NH_3} = \sum (A_i \times NF_i \times EF_{NH_3}) \tag{6}$$

where E_{NH3} is the emitted NH_3 from N-fixing crops; A_i is the area covered by N-fixing crops *i*; NF_i is the nitrogen fixation rate of type *i* N-fixing crops; and EF_{NH3} is the ammonia EF for crops *i.*

[17] The province-level cultivated areas of these three nitrogen-fixing crop groups were provided by the National Bureau of Statistics of China [*NBSC*, 2006, 2007e]. The N fixation rates of these crops are 105, 120 and 130 kg N/ha/yr, respectively [*Kuenzler and Craig*, 1986; *Yan et al.*, 2003]. Data on NH_3 fluxes over legume crops are sparse, but the 0.01 kg NH_3/kg N suggested by *EEA* was used in our estimation [*EEA*, 2006].

2.1.4. Compost of Crop Residues

[18] Composting refers to the use of both aerobic and anaerobic microbial processes to degrade waste materials for beneficial reuse. In the rural China, crop straws and other agricultural residues are often composted to add soil nutrients. A large amount of ammonia is released during this biological process. The ammonia emissions from this source are assessed by multiplying the amount and the corresponding EFs. Provincial crop residue production can be estimated based on crop production and production-to-residue ratios [*Lal*, 2005; *NBSC*, 2007e]. The portion of crop residues for composting refers to an investigation covering most of the provinces in China [*Gao and Ma*, 2002]. The ammonia EF for the composting operation is 0.32 kg/ton [*Stephen et al.*, 2004].

2.2. Livestock Wastes

[19] Nitrogen in animal excrement in the form of urea can rapidly hydrolyze to form ammonium carbonate and then volatilize as gaseous ammonia. Early inventories of ammonia emission from livestock wastes were calculated as the product of livestock numbers and annual EFs per animal [*Buijsman et al.*, 1987]. More recent inventories have improved the single EFs per animal with specific EFs for the various phases of manure management, including animal housing, manure storage, manure spreading and the grazing stage [*Pain et al.*, 1998; *Zhang et al.*, 2010]. Recently, the mass-flow approach has been widely used at the national and global scale [*Webb et al.*, 2006; *Beusen et al.*, 2008]. Ammonia is emitted from a pool of ammoniacal nitrogen (TAN), which is not increased during manure management, but it is depleted by the losses though ammonia volatilization, leaching and other pathways during different stages. This approach enables the rapid estimation of the consequences of abatement at the upstream stage on the ammonia losses at the downstream stage, which is more accurate than the previous methods [*Webb and Misselbrook*, 2004].

[20] Based on the mass-flow methodology in the EEA's inventory guidebook [*EEA*, 2009], we analyzed the transformation and migration of nitrogen in the animal husbandry (Figure 1). The ammonia emissions from each stage of livestock manure management are affected by many factors, such as species, gender, age, bodyweight, nitrogen content of the feed, housing structure, manure storage system,

Table 3. Parameters Used in Estimates of Annual TAN Excretion per Animal for Each Livestock Class

Livestock Class	Period[a] (day)	Excrement[b] (kg/day/cap)		Nitrogen Contents[c] (%)		TAN Content[d] (%)
		Urine	Faece	Urine	Faece	
Beef <1 year	365	5.00	7.0	0.90	0.38	60
Beef >1 year	365	10.00	20.0	0.90	0.38	60
Dairy cows <1 year	365	5.00	7.0	0.90	0.38	60
Dairy cows >1 year	365	19.00	40.0	0.90	0.38	60
Goat <1 year	365	0.66	1.5	1.35	0.75	60
Goat >1 year	365	0.75	2.6	1.35	0.75	50
Sheep <1 year	365	0.66	1.5	1.35	0.75	60
Sheep >1 year	365	0.75	2.6	1.35	0.75	50
Sow	365	5.70	2.1	0.40	0.34	70
Weaner	75	1.20	0.5	0.40	0.34	70
Fattening pig	75	3.20	1.5	0.40	0.34	70
Horse	365	6.50	15.0	1.40	0.20	60
Donkey	365	6.50	15.0	1.40	0.20	60
Mule	365	6.50	15.0	1.40	0.20	60
Camel	365	6.50	15.0	1.40	0.20	60
Laying hen	365	-	0.12	-	1.63	70
Laying duck	365	-	0.13	-	1.10	70
Laying goose	365	-	0.13	-	0.55	70
Broilers	50	-	0.09	-	1.63	70
Meat duck	55	-	0.10	-	1.10	70
Meat goose	70	-	0.10	-	0.55	70

[a]Values are derived from *CAU* [1997] and *Zhou et al.* [2010].
[b]Values are derived from *CAU* [1997], *MEPPRC* [2001], and *K. Wang et al.* [2009].
[c]Values are derived from *CAU* [1978].
[d]Values are derived from *Webb and Misselbrook* [2004] and *EEA* [2009].

spreading technique, time spent outside or inside and meteorological conditions [*Zhang et al.*, 2010]. Although numerous experiments have been performed on ammonia emission during livestock production, few measurements have been recorded in China. Consequently, China-specific EFs must be recalculated on the basis of the native practice of livestock production and environmental conditions.

[21] In China, animals are raised in several completely different systems, including free-range, intensive and grazing system. Ammonia emissions were calculated separately for these three systems. The typical animal categories in China are listed in Table 3. Gender and age are also taken into account in the definition of livestock subcategories. The available statistical data, official surveys (http://www.caaa.cn/) and industry reports for husbandry were collected to obtain the number of each livestock class [*Editorial Office of China Animal Industry Yearbook* (*EOCAIY*), 2007]. The annual excretions of TAN by each livestock class were derived based on the rearing period [*China Agricultural University* (*CAU*), 1997; *Zhou et al.*, 2010], the daily amount of excrement [*CAU*, 1997; *Ministry of Environmental Protection of People's Republic of China (MEPPRC)*, 2001; *K. Wang et al.*, 2009], the nitrogen content and the percent of TAN [*CAU*, 1978; *Webb and Misselbrook*, 2004; *EEA*, 2009] (Table 3). In the following stepwise procedure of manure management, slurry and solid manures were calculated separately. Monthly emissions estimation was conducted based on temperature-depended EFs (Table S2). We assumed that the animal population is same during every month of the whole year since the monthly fluctuation of meat productions is very small (http://www.caaa.cn/). Monthly specific EFs were developed using provincial monthly average temperature from NCEP final analysis data set.

2.2.1. Housing

[22] Housing emission is related to the portion of excreta deposited inside buildings. Various livestock classes have different fractions of time spent in buildings (Table S2). Emissions from different locations were calculated by multiplying the amount of TAN and EFs. Ammonia volatilization from the housing stage depends on factors such as housing conditions, humidity, temperature and animal waste handling practices [*Anderson et al.*, 2003]. In our study, we characterized the EFs by flooring type, waste handling practice and temperature variations. Deep litter flooring is commonly used in free-range and grazing systems, but solid flooring is widespread in intensive large-scale farming. The differences between these methods are summarized by *Hutchings et al.* [2001]. The handling practice of waste in the housing stage has been surveyed in some provinces in China [*Liu et al.*, 2008], and the EFs for slurry and solid were derived from previous studies [*Webb and Misselbrook*, 2004; *Department for Environment, Food and Rural Affairs*, 2005; *EEA*, 2009]. The temperature adjustment of the volatilization rate was accomplished though the diverse EFs at different temperature intervals [*Mannebeck and Oldenburg*, 1991; *Koerkamp et al.*, 1998].

2.2.2. Storage

[23] According to the actual treatment of housing waste in China, usually no treatment is given for the excrement deposited outside buildings, and those excreted inside are usually settled in two ways: biogas generation and composting. The handling practices of manure storage are different in different parts of China. According to Liu *et al.*'s investigation, approximately 10% more manure is used to produce biogas in the Southern provinces in the free-range rearing system [*Liu et al.*, 2008]. Unfortunately, there is limited data on the fraction of different storage methods in

Table 4. Ammonia EFs of Miscellaneous Anthropogenic Sources

Sources	EFs	Unit	Reference
Biomass burning			
Forest fires	2.9	g NH_3/kg	*Song et al.* [2010]
Grassland fires	0.7	g NH_3/kg	*Song et al.* [2010]
Crop residues burning	0.37(wheat) 0.68(maize) 0.52(others)	g NH_3/kg	*Li et al.* [2007]
Fuelwood combustion	1.3	g NH_3/kg	*Andreae and Merlet* [2001]
Human excrement	0.787	kg NH_3/yr/cap	*Buijsman et al.* [1987], *Möller and Schieferdecker* [1989], *Yin et al.* [2010]
Chemical industry			
Synthetic ammonia	0.01	kg NH_3/ton	*EEA* [2009]
Nitrogen fertilizers production	5.0	kg NH_3/ton	*Asman* [1992]
Waste disposal			
Wastewater	0.003	g NH_3/m^3	*Yin et al.* [2010]
Landfill	0.560	kg NH_3/ton	*Yin et al.* [2010]
Compost	1.275	kg NH_3/ton	*Stephen et al.* [2004]
Incineration	0.210	kg NH_3/ton	*Yin et al.* [2010]
Traffic			
Light-duty gasoline vehicles	0.026	g NH_3/km	*Stephen et al.* [2004]
Heavy-duty gasoline vehicles	0.028	g NH_3/km	*Stephen et al.* [2004]
Light-duty diesel vehicles	0.004	g NH_3/km	*Stephen et al.* [2004]
Heavy-duty diesel vehicles	0.017	g NH_3/km	*Stephen et al.* [2004]
Motorcycles	0.007	g NH_3/km	*Stephen et al.* [2004]

the intensive rearing system, for which half of the manure was assumed to be handled as slurry. According to the EEA's guidelines [*EEA*, 2009], the immobilization of TAN, the emission of other nitrogenous gases (N_2O, NO and N_2) and the leaching loss of nitrogen are other notable pathways of nitrogen transformation. The parameters involved in these processes are all listed in Table S2.

2.2.3. Spreading

[24] Manure is extensively applied to cropland as basal fertilizer in China [*Liu et al.*, 2008; *Li et al.*, 2009]. The estimate refers to the approach recommended by EEA but is improved by adding the parameter Xfeed, which is defined as the portion of manure used for feed production. The reason for this addition is that large-scale henneries and hoggeries often use the manure as feed after treatments. The EFs for different livestock classes were derived from existing experiments and expert judgments (Table S2) [*Webb and Misselbrook*, 2004; *EEA*, 2009].

2.3. Miscellaneous Sources

2.3.1. Biomass Burning

[25] Biomass burning could release significant amounts of ammonia [*Andreae and Merlet*, 2001]. The types of biomass burning in this study include forest and grass fires, crop residue in-field burning, domestic combustion and fuelwood combustion. The emissions from crop residues and fuelwood were estimated based on the statistical data [*NBSC*, 2007e, 2008b]. The emissions from forest and grassland fires were estimated by combining a newly available MODIS (Moderate Emission Resolution Imaging Spectroradiometer) burned area product (MCD45A1) with the combustion completeness considering the impact of vegetation moisture content [*Song et al.*, 2010]. The EFs for each type of biomass are summarized in Table 4 [*Li et al.*, 2007].

2.3.2. Excrement of Rural Populations

[26] China's rural areas are widely distributed with tatty latrines (roughly constructed latrines without any flushing treatment). Feces in these tatty latrines accumulate in the open air and are subsequently used as fertilizer, which is another important ammonia emitter. The amount of human waste used for spreading was obtained based on the daily excrement of children and adults, the rural population and the fraction of tatty latrines in each province [*NBSC*, 2007a, 2007d]. The EFs were the average of several publications [*Buijsman et al.*, 1987; *Möller and Schieferdecker*, 1989; *Yin et al.*, 2010], which are listed in Table 4.

2.3.3. Chemical Industry

[27] The chemical industry is also a gaseous ammonia source, particularly the production of synthetic ammonia and nitrogen fertilizers. These emissions were estimated by multiplying the production and corresponding EFs. The available census data for the chemical industry were provided by the national statistical bureau [*NBSC*, 2007b]. The EFs were provided by EEA and EPA reports [*Stephen et al.*, 2004; *EEA*, 2009].

2.3.4. Waste Disposal

[28] Ammonia emissions from wastewater treatment plants and refuse processing plants were also included; these emissions should not be neglected, especially in urban areas [*Anderson et al.*, 2003]. The waste disposal types considered were wastewater treatment, landfill, compost and incineration of solid waste. These data were obtained from the available yearbook [*Statistics and Census Service of Macao SAR Government*, 2007; *NBSC*, 2007d] (http://www.dgbas.gov.tw), and the corresponding EFs were derived from existing publications [*Stephen et al.*, 2004; *Yin et al.*, 2010].

2.3.5. Traffic Sources

[29] There is evidence that traffic may also be an important source of ammonia in urban areas [*Ianniello et al.*, 2010; *Vega et al.*, 2010; *Meng et al.*, 2011]. The vehicles taken into consideration include light-duty and heavy-duty gasoline vehicles, light-duty and heavy-duty diesel vehicles and motorcycles. The emissions were calculated as the product of the number of operating vehicles [*China's Association of Automobile Manufacturers*, 2007], mileage [*Cai and Xie*, 2007] and ammonia emissions per kilometer for each vehicle category [*Stephen et al.*, 2004].

Table 5. Regional-Specific Ammonia Emissions (Gg) From Different Sources in 2006

	Fertilizer	Agricultural Soil	N-Fixing Crop	Compost	Livestock	Biomass Burning	Human Excrement	Chemical Industry	Waste Disposal	Traffic
Beijing	9.3	0.6	23.9	2.2	24.6	0.3	0.6	0.1	5.9	3.7
Tianjin	17.0	0.9	0.0	0.3	26.5	0.4	0.2	0.9	1.9	1.3
Hebei	230.2	12.5	0.0	14.5	505.5	6.9	15.5	11.8	4.4	4.2
Shanxi	73.9	8.4	1.1	4.2	113.4	2.9	6.6	18.4	1.6	2.1
Inner Mongolia	79.2	14.9	0.6	5.4	236.3	4.9	5.7	4.1	1.7	1.5
Liaoning	68.3	7.6	1.8	10.5	234.6	5.1	5.9	4.8	6.1	2.6
Jilin	72.5	10.2	0.5	7.9	184.1	8.6	3.1	1.1	1.2	1.3
Heilongjiang	102.4	21.4	0.7	8.8	155.9	8.4	5.1	3.1	2.6	1.6
Shanghai	13.3	0.6	5.4	0.2	8.4	0.2	0.1	0.1	7.5	1.8
Jiangsu	242.7	9.2	0.0	6.5	150.1	5.3	9.2	13.1	10.3	4.5
Zhejiang	49.1	3.9	0.7	4.1	43.8	1.4	2.8	3.1	5.9	4.4
Anhui	149.2	10.9	0.2	8.5	146.2	5.2	11.4	9.6	3.0	1.8
Fujian	45.2	2.6	1.6	3.3	48.8	0.9	4.4	3.7	2.8	1.8
Jiangxi	61.7	5.5	0.3	12.8	95.7	3.2	5.4	2.7	2.0	1.2
Shandong	335.9	14.0	0.5	12.2	467.9	10.1	8.8	39.7	8.4	5.8
Henan	566.8	14.8	1.5	17.2	587.4	10.4	13.6	24.8	4.2	3.4
Hubei	137.1	9.0	2.1	13.7	127.3	4.5	7.0	15.8	5.3	1.9
Hunan	130.6	7.2	0.6	15.2	195.5	4.8	10.5	13.6	3.8	1.7
Guangdong	107.9	6.0	0.4	17.9	143.5	2.0	4.7	0.5	11.9	7.8
Guangxi	110.4	8.0	0.6	23.3	183.4	2.6	10.1	3.4	2.4	1.6
Hainan	15.2	1.4	0.5	2.1	33.7	0.2	1.4	3.9	0.7	0.4
Chongqing	45.4	0.0	0.1	6.9	68.3	1.5	5.4	4.3	1.6	1.0
Sichuan	159.0	16.7	0.4	19.0	394.8	5.3	20.0	16.8	3.7	3.0
Guizhou	45.2	8.9	1.1	23.1	183.7	2.3	10.9	9.0	0.9	0.9
Yunnan	57.0	11.7	0.6	12.8	228.1	3.1	9.4	8.3	1.5	2.1
Tibet	1.6	0.7	0.9	0.3	77.5	0.1	0.7	0.0	0.1	0.2
Shaanxi	101.4	9.4	0.0	8.1	112.9	2.4	9.5	5.5	1.7	1.4
Gansu	57.3	9.2	0.5	4.6	157.6	1.4	5.1	3.6	0.9	0.6
Qinghai	5.2	1.3	0.5	0.5	94.2	0.2	1.2	0.0	0.4	0.2
Ningxia	38.1	2.3	0.1	0.6	36.2	0.7	1.3	4.4	0.7	0.3
Xinjiang	82.2	7.3	0.1	6.2	226.7	4.1	4.8	7.5	1.3	1.1
Hong Kong	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	4.1	0.5
Macao	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.1
Taiwan	4.3	1.5	0.3	0.2	19.2	0.3	0.0	1.0	4.1	8.2
Total	3214.4	238.3	47.7	272.9	5311.6	109.6	200.4	238.9	114.9	75.8

2.4. Gridded and Monthly Emissions

[30] The emissions were compiled at a 1 km × 1 km spatial resolution by the various types. The fertilization and wild fire emissions were already calculated based on gridded activity data and EFs, so no allocation was needed. The other emissions were presented at the provincial level first and then allocated using land cover, rural population, and other proxies. More specifically, emissions from farmland ecosystems, with the exception of fertilization, were allocated by land cover class. Emissions from free-range and intensive livestock production were redistributed based on rural population density, and the grassland land cover class was used to allocate grazing emission. Emission from the field burning of crop residue was gridded by using the MODIS thermal anomalies/fire products (MOD14A2 and MYD14A2). Emissions from biofuel combustion and human waste were also regridded on the basis of rural population distribution. The remaining sources were equally distributed in each grid. After the spatial allocation of all emissions was completed, an aggregation was conducted to sum up all emissions.

[31] The monthly emission estimation for fertilizer application and livestock waste were elaborated in detail in sections 2.1 and 2.2. Efforts were also devoted to temporally allocate ammonia emission from biomass burning. MCD45A1 (monthly burned area product), MOD14A2 and MYD14A2 products (8-day thermal anomalies/fire products) were utilized to ascertain the timing of different kind of biomes, more details could be found in two existing studies [*Song et al.*, 2010; *Huang et al.*, 2012]. Emissions from other minor sources were equally divided into 12 months.

3. Result and Discussion

3.1. Ammonia Source Apportionment

[32] The ammonia emissions at the individual province level in 2006 from different sources are presented in Table 5. As shown, the total ammonia emission was 9.8 Tg, contributing approximately 15% and 35% to global and Asian emissions, respectively, based on the previous assessment at the global and Asian scale [*Bouwman et al.*, 1997; *Streets et al.*, 2003]. The ammonia emission from China is much higher than elsewhere, exceeding Europe (5 Tg) and the USA (4 Tg) by approximately 96% and 145%, respectively [*Bouwman et al.*, 1997; *Pain et al.*, 1998; *Anderson et al.*, 2003].

[33] The most important contributor is livestock manure management (5.3 Tg), accounting for approximately 54% of the total budget. Next is the fertilizer application (3.2 Tg), which is responsible for 33% of emissions. The less important sources at the national scale are the following: soil 0.2 Tg (2%), N-fixing crops 0.05 Tg (1%), compost of crop residues 0.3 Tg (3%), biomass burning 0.1 Tg (1%), human

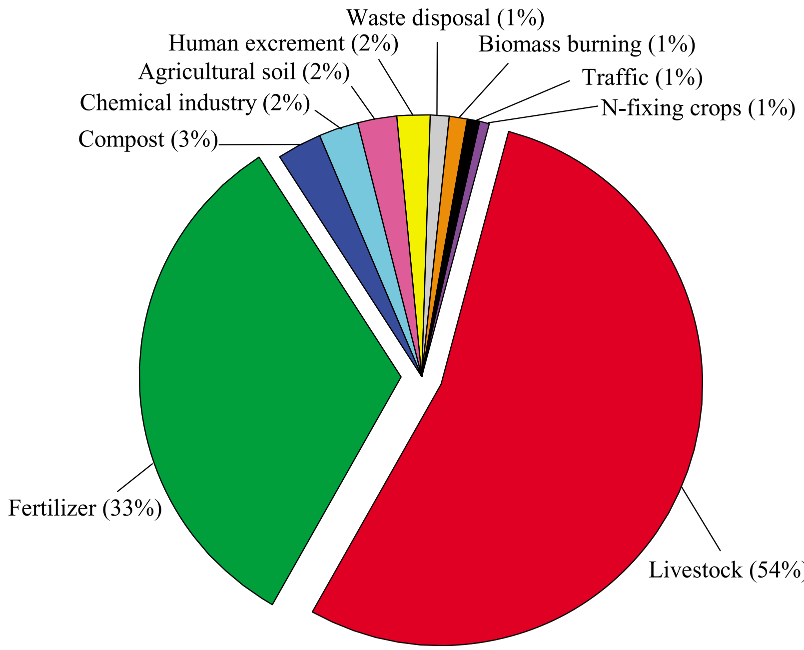

Figure 2. Source contributions (%) to ammonia emissions in China.

excrement 0.2 Tg (2%), the chemical industry 0.2 Tg (2%), waste disposal 0.1 Tg (1%) and traffic 0.1 Tg (1%).

[34] The emission characteristics in China reflect its unique agricultural structure and farming practice. For Europe and the USA, livestock overwhelmingly dominate the inventory. In China, however, both fertilizer and livestock play significant roles. The types of fertilizer used in China are quite different from those used in developed countries. Urea and ABC, which are highly volatile compounds, are the two most widely used N fertilizers in China. In contrast, less volatile fertilizers such as anhydrous ammonia and ammonium nitrate are much more popular in North America and Europe (http://www.fertilizer.org/ifa/ifadata). The incredibly large consumption and high volatility of urea and ABC make the ammonia emission from fertilizer application as important as that from the livestock in China. At the same time, the feature of husbandry in China is also distinct, being characterized by multiple rearing systems. The most important is the free-range system, accounting for 65% of the total. In most rural areas, livestock are raised in a traditional way; that is, a small number of animals are fed by individual families and are allowed to roam freely instead of being contained in any manner. Next is intensive rearing (approximately 30%), which refers to the process of raising livestock in confinement at high stocking density; in this process, the farm operates as a factory. This type of farming is extensive in broiler (73%), dairy cow (57%) and pig (41%) production [*EOCAIY*, 2007]. Grazing, a relatively less important system, only dominates in the northern and western parts of China. Consequently, the free-range system in rural areas is the greatest contributor, accounting for 67% of the emissions of livestock. The largest NH_3 emission livestock category is cattle, which emitted 1.9 Tg NH_3 per year, followed by laying hens and pigs, which both emitted 0.7 Tg NH_3 per year.

[35] Other sources were not as notable for the whole country, as shown in Figure 2. However, some of these sources could become significant at the provincial scale. For instance, the contribution of biomass burning is a bit higher in Heilongjiang and Jilin provinces (3%), where forest fires frequently occur. Human waste is responsible for greater portions in Guizhou and Shaanxi provinces because of the larger rural populations and the ubiquitous tatty latrines (sanitary latrines only account for only 30% and 35%, respectively). In Shanxi and Hainan provinces, 8% and 7% of ammonia emissions are caused by chemical industry production, respectively, which is higher than the average of 2% for the whole country. Both waste disposal and traffic source are far more prevalent in urban areas such as Beijing and Shanghai, with, on average, 12 and 7 times greater contributions than elsewhere, respectively.

3.2. Spatial and Temporal Distribution

[36] The spatial pattern of ammonia emission is illustrated in Figure 3. It is easy to see the high emission rates in Henan, Shandong, Hebei, and Jiangsu provinces and in Eastern Sichuan. The emission contributions from Henan, Shandong, Hebei and Sichuan provinces, all of which have high agriculture production, accounted for 12.7%, 9.2%, 8.2% and 6.5%, respectively. The highest emission rates of NH_3 were mainly caused by fertilizer application and livestock, which jointly contributed 93%, 88%, 91% and 87% for these four major emitters, respectively.

[37] Table 5 lists the emission amounts from fertilizer application in each province (the pattern is shown in Figure S1 in 1 km grid cells). The intensity of NH_3 volatilization from synthetic N fertilizers was concentrated in Henan, Shandong, Hebei and Jiangsu province, where the crop cultivation is most developed over the whole country. The crop production in these four provinces is the greatest in China, with a total contribution of 33% of China's crop production, and they consumed approximately 30% of the N

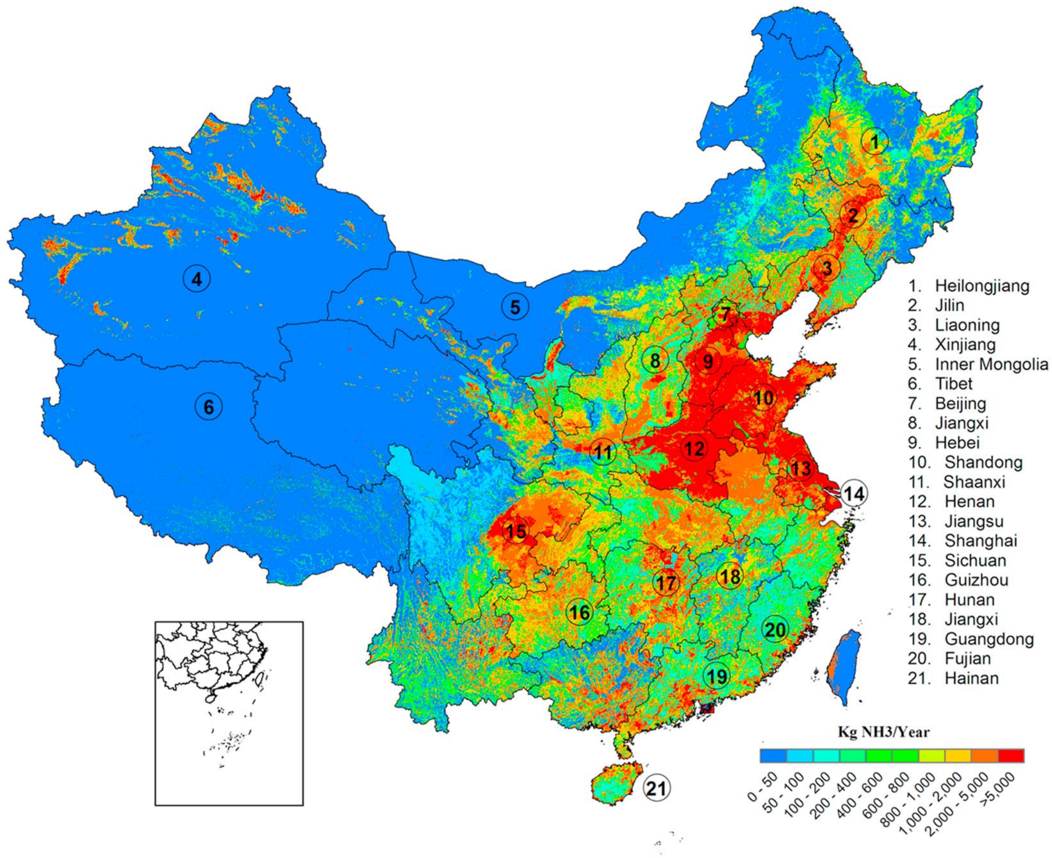

Figure 3. Spatial distribution of ammonia emissions in 1 km grid cell (kg yr^{-1}).

fertilizer for the whole country in 2006 [*NBSC*, 2007e]. For example, the ammonia emission rates in Henan province reached a maximum value of 7.2 kg per square kilometer of arable land. The farmers here usually adhere to fertilization during plant cultivation. The application rate in this region was incredibly high, and consequently, the nitrogen loss was tremendous [*Ju et al.*, 2009]. According to an investigation carried out by the government [*NBSC*, 2007c], the application rate of N fertilizer in Henan province is 1.1–3.4 times higher than the national average for staple crops. For ABC, the gap was even greater (1.1–9.3 times). Among the various kinds of crops, sugarcane and vegetable have the highest fertilization rates (335 and 264 kg N per hectare, respectively), which are double the rates of rice and wheat and are approximately tenfold higher than those of beans and peanuts. In addition, vegetables are cultivated extensively in China, with a sowing area that is just behind cereal crops. Over 20% of ammonia loss originates from vegetable fertilization. The North China Plain, with a 25,624,000 and 5,754,000 ha area under cultivation of cereal crops and vegetables, respectively, is responsible for 43% of the NH_3 emissions from fertilization in China. The cultivated land in Guangdong, Fujian, Hunan and Jiangxi provinces also shows large NH_3 volatilization. Paddy fields are common in this region, with rice being the dominant crop and contributing the most emissions (approximately 40% for Jiangxi province). The smaller emitters are located mostly in western China, with a minimal amount of arable land or low use of synthetic nitrogenous fertilizers.

[38] The ammonia emissions from livestock during 2006 are also presented in Table 5 and Fig. S2. The high NH_3 emission areas, with rates over 2000 kg NH_3 per square kilometer, were concentrated in Henan, Shandong, Hebei and East Sichuan province. Approximately 5.3 Tg NH_3 was released in China due to livestock rearing, 0.6 Tg of which was emitted from Henan province, followed by Hebei (0.5 Tg), Shandong (0.5Tg) and Sichuan (0.4 Tg). These four provinces are well-known for their large animal population, providing approximately 36% of China's husbandry production [*NBSC*, 2007e]. Many kinds of animals are extensively bred in Henan, Hebei and Shandong provinces, including those raised for beef, dairy, pork and poultry. Among them, the greatest contributors are laying hens and beef, which contribute 26% and 24%, respectively, to the total emissions of these four provinces. Inner Mongolia, Tibet and Xinjiang province, where sheep are widely raised, also emit remarkable ammonia emissions related to sheep manure management, accounting for over 40% of emissions of these provinces. Cattle are widely raised in Northeast China and are responsible for 53% and 51% of the ammonia emissions in Liaoning and Heilongjiang provinces, respectively. Sichuan province is another large emitter, with an emission rate of 0.4 Tg NH_3 per year. Cattle and pig production released 54% and 20% NH_3 among all livestock emission in Sichuan province, respectively.

[39] The seasonal distribution of ammonia emissions was in general agreement with that of temperature and agricultural timing (Figure 4). Generally, the maximum emission

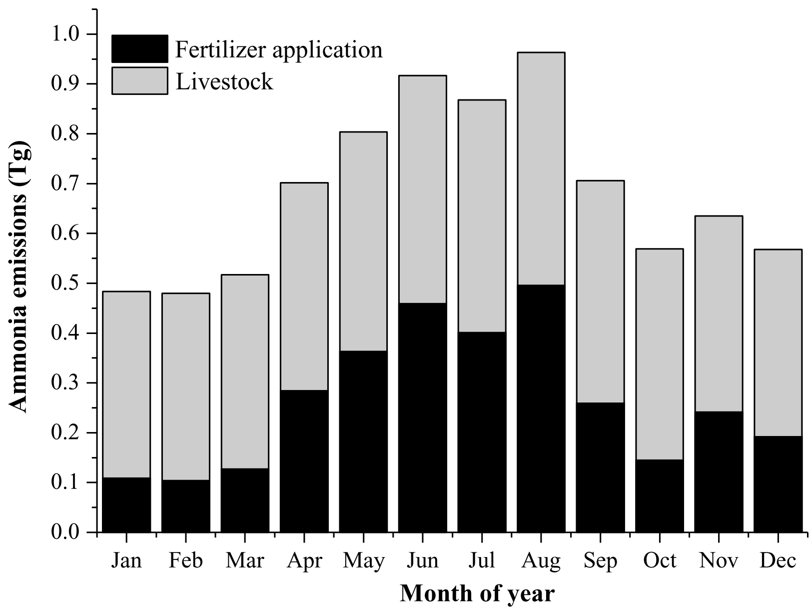

Figure 4. Monthly ammonia emissions in 2006.

was in summer, and nearly 42% and 28% of the fertilization and livestock emissions, respectively, occurred during this season because of the high temperature and dense fertilization. *Streets et al.* [2003] also showed a roughly similar seasonal pattern of ammonia emission, except for fluctuations from June to August and September to November. The disparity was attributed to the timing of different plants. Various crops were planted on the same farmland in rotation over large areas in China, particularly south part, such as the winter wheat-summer maize rotation system practiced in the North China Plain. Summer plants such as maize are usually seeded in June with the application of base fertilizer, and the topdressing fertilizer is applied two months later, and the peak emissions correspondingly occur during June and August (http://www.zzys.gov.cn/). Moreover, fertilizer is densely applied for semi-late rice in June with emissions of 0.16 Tg NH_3 (42% of total emissions in June), and cotton is intensively fertilized in August (0.14 Tg, 35% of total emissions in August), which also leads to peak volatilization in June and August. The high emission rates in September and November could be attributed to the basal dressing and top dressing of wheat, with 0.11 Tg and 0.15 Tg NH_3 emissions respectively. The high ammonia volatilization rate from livestock also occurs in summer. There is little variation of animal population among the different seasons. The increasing emissions during the time period from June to August were caused by larger EFs associated with the substantial increase of temperature. This tendency could be mostly attributed to emissions from laying hens, which were responsible mostly for the monthly variation because their EFs change more rapidly with temperature than other species [*Mannebeck and Oldenburg*, 1991]. Although other emission sources such as biomass burning also have seasonal variations, temporal disparities were not considered in our estimate due to their relatively small contributions.

3.3. Comparison With Existing Result

[40] Several inventories of ammonia emissions have been established in China for different base years in support of various activity data set and EFs. Among them, the ammonia emission inventory developed by *Streets et al.* [2003] (available at http://www.cgrer.uiowa.edu/EMISSION_DATA_new/) has been widely used in air quality modeling [*Zhang et al.*, 2007; *Lin et al.*, 2008].

[41] Our estimation was compared with the previous emission inventories listed in Table 6. As presented, the results are generally 40–50% higher than ours. These disparities are caused mainly by the emission estimations from the fertilizer volatilization. Our estimate has smaller emissions and larger spatial disparities. The fertilizer consumptions are comparable, and the discrepancy is mostly caused by the distinct EFs employed. *Zhao and Wang* [1994],

Table 6. Comparison of the Ammonia Emissions (Tg yr^{-1}) in Our Study With Other Published Results

	Base Year	Total	Fertilizer	Husbandry	Biomass Burning	Others
This study	2006	9.8	3.2	5.3	0.2	1.1
Zhao and Wang [1994]	1990	13.6	6.4	4.2	-	3.0
Yan et al. [2003]	1995	-	4.6	-	-	-
Streets et al. [2003]	2000	13.6	6.7	5.0	0.8	1.1
Ohara et al. [2007]	2000	-	-	5.5	0.5	-
S. Wang et al. [2009]	2005	13.4	5.1	7.0	-	1.3
Zhang et al. [2011]	2005	-	4.3	-	-	-

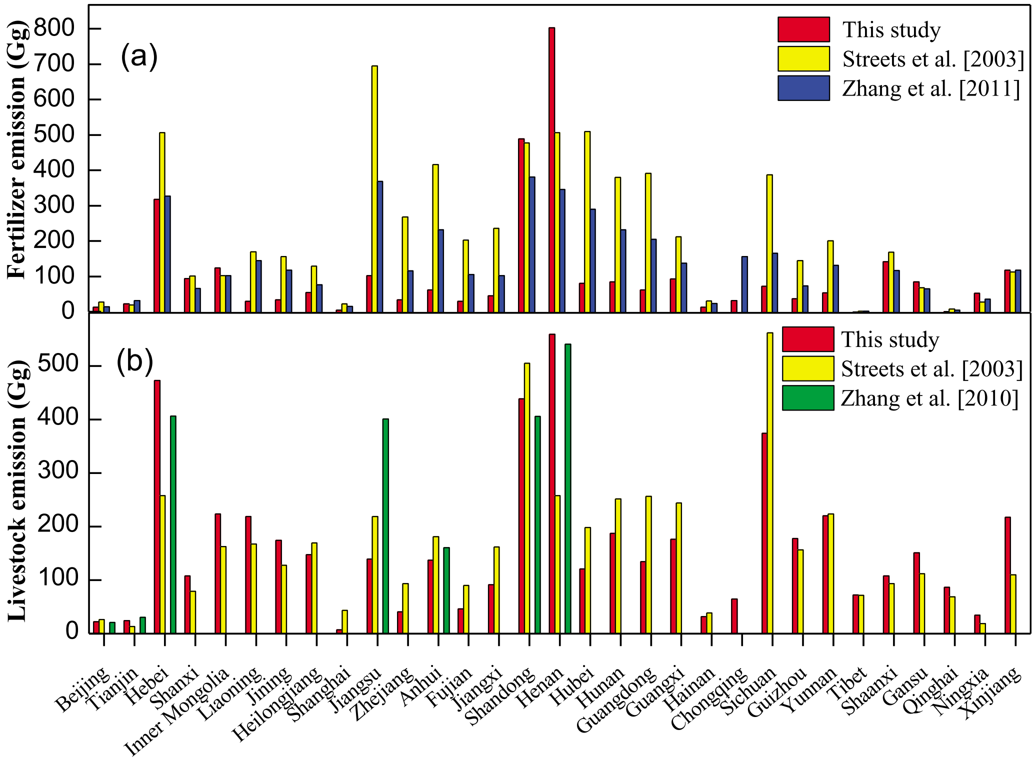

Figure 5. Comparison of provincial-level results on (a) fertilizer and (b) livestock emissions.

Streets et al. [2003], *Yan et al.* [2003] and *S. Wang et al.* [2009] used uniform EFs for the whole country, and these values were based on European experiments or foreign expert judgments rather than native measurements in China. *Zhang et al.* [2011] spatially and temporally characterized the EFs at the county level using the National Ammonia Reduction Strategy Evaluation System (NARSES) model developed for agriculture emissions in the UK. Similarly, many parameters have been introduced based on measurements in Europe. As mentioned above, the ammonia volatilization from chemical fertilizer strongly depends on numerous environmental conditions. The soil pH values, temperatures and agricultural practices are completely different between Europe and China. Our estimation made full use of local experiments, and it adjusted the EFs for different crops under local conditions. For instance, the EFs for Urea and ABC are identical, at 15% and 30%, respectively, in *Streets et al.*'s [2003] paper, while in our calculation, the EFs can reach up to 35–40% for non-glutinous rice, which is top-dressed during June to July, and then decrease to lower than 5% for early rice that is basal-dressed during February to March. Consequently, the disparity between their results and the estimates made in this study are to be expected. The comparisons of province-level emissions are demonstrated in Figure 5a. The largest emitters are concentrated in the Hebei, Henan, Shandong, Jiangsu and Sichuan provinces, in accordance with previous results [*Streets et al.*, 2003; *Zhang et al.*, 2011]. Henan and Shandong were given more weight in our budget, with 18% and 10% of the total fertilizer emissions, while the corresponding contributions in the inventory of Streets *et al.* are 10% and 7%, respectively. Simultaneously, the contribution of Jiangsu Province declines from 10% in Streets *et al.*'s result to 8% in our estimate. This difference could be attributed to the involvement of the soil acidity and the application rate. More emissions occurred in Henan and Shandong provinces because of alkaline soil and higher application rates, which were not considered in previous studies and might cause underestimation in this area. Jiangsu has fewer emissions in our results due to the lower soil pH and the smaller application rate.

[42] Comparisons of emissions related to animal manure management at the national scale and the province level are indicated in Table 6 and Figure 5b. Our estimation is comparable to the results of *Zhao and Wang* [1994], *Streets et al.* [2003] and *Ohara et al.* [2007], at approximately 5.0 Tg/yr. The value given by *S. Wang et al.* [2009] was 7.0 Tg/yr. The differences among these inventories mainly resulted from the selection of EFs. For previous livestock ammonia inventories, the EFs were either presented in loss rates per capita and then multiplied by population [*Zhao and Wang*, 1994; *Streets et al.*, 2003; *Ohara et al.*, 2007] or were recalculated for distinct stages as the portion of ammonia [*S. Wang et al.*, 2009; *Zhang et al.*, 2010]. However, most of the previous inventories used Europe-based EFs, introducing significant inaccuracies [*Streets et al.*, 2003]. Our research considered three different rearing systems with Chinese characteristics. The EFs were also disaggregated both spatially and temporally according to the climate conditions and local manure treatment practices. Region-specific EFs made it possible to accurately represent the emission distribution. Spatially, Henan and Hebei provinces, two biggest emitters, released 0.6 and 0.5 Tg NH_3 in our estimate, respectively, comparable with the results given by *Zhang et al.* [2010]; the corresponding value in Streets et al.'s paper is 0.3 and 0.3 Tg, as shown in Figure 5b. Higher emissions in our

study were caused by larger animal population in these two provinces in 2006 than 2000 (base year of Streets et al.'s inventory) [*NBSC*, 2007a]. In contrast, Sichuan province plays a much more important role in Streets et al.'s inventory due to the high assessment of N excrement from pig production. N excrement is age-dependent, especially for pigs [*Liu et al.*, 2008]. Sichuan, with a large amount of pig production, might be overemphasized in Streets *et al.*'s budget due to the neglect of disparities among excrements in different growth phases.

[43] Compared with the previous ammonia inventories, many more sources were considered, including biomass burning and waste disposal. Although the total emissions of these miscellaneous sources were relatively small, some play an important role at the regional scale. For instance, waste disposal contributed almost 32% of the ammonia emission in Shanghai. Therefore, it is not acceptable to neglect these sources during calculation. Most existing inventories ignored these small sources, thus resulting in inaccurate estimates of some areas.

[44] Our results are also compared with the satellite data revealing global NH_3 column distribution [*Clarisse et al.*, 2009]. Elevated columns are observed in Northeast China and the North China Plain through satellite monitoring, similar to our inventory. However, little ammonia is found in South China and Sichuan province according to satellite observation. The difference between our estimation and satellite observed distributions is first attributed to the fact that satellite monitoring suffers signal impairments caused by meteorological effects such as water vapor, clouds [*Garcia et al.*, 2008]. In South China and Sichuan province, higher amount of rain and cloud cover might introduce some uncertainties. Second, concentration distribution could not be always consistent with emission pattern. As mentioned in section 1, ammonia is very reactive and consequently short-lived in the atmosphere. It could react with many acid materials like SO_2. Sichuan and Guizhou province are highly polluted areas with large amounts of SO_2 emission [*Zhang et al.*, 2009]. Simultaneously, both temperature and humidity are high there, which further accelerate NH_3 depletion. The disparities between our estimation and satellite results are to be expected.

3.4. Uncertainty

[45] Emission uncertainty is associated with activity data and EFs. Activity data were obtained through various first-hand investigations and from statistical information. Local conditions and situations were also considered in the process of EF correction. Nevertheless, there are still large uncertainties, particularly for fertilization and livestock emissions, due to the extremely large values and the numerous parameters involved in the EF adjustment. Generally, for economic activities such as fossil fuel use and industrial and agricultural production, which are often reasonably accurate, the coefficient variation (CV) is usually 5–10%. Activities such as biofuel use, waste burning and landfilling are less well-documented [*Zhao et al.*, 2011]. The distribution and uncertainties for activity levels for various sources are listed in Table S3 based on existing publications or assumptions made for this study. The EFs show a substantial variation according to the type of process [*Olivier et al.*, 1998]. The EFs for NH_3 emissions from biofuel combustion and chemical industry production have larger fluctuations ($CV \geq 100\%$), and others have smaller uncertainties, such as livestock (CV: 50%) and waste burning (CV: 50%). Further details can be found in Table S3. We ran 20 000 Monte Carlo simulations to estimate the range of fire emissions with a 95% confidence interval. The estimated emission range was 6.5–12.5 Tg/yr.

4. Conclusion

[46] We developed a comprehensive and fine emission inventory of ammonia in China by combining the statistical data set with MODIS products. EFs specified temporally and spatially were used to enhance the accuracy of this estimation. The total emissions for ammonia in China was 9.8 Tg, of which nitrogen fertilizer application and livestock manure management were the largest contributors, at 3.2 Tg and 5.3 Tg, respectively. Other sources accounted for 13% of the total, but some may play an important role in certain locations. This inventory covered the whole year of 2006, and it showed obvious seasonal variability. The lowest emissions occurred in winter, while the highest emissions were in summer. These emissions were significantly associated with temperature variations and the timing of farming practices. At the province level, Henan was the largest contributor, mostly because of the robust agricultural activity, with both crop production and animal husbandry. This work provides a detailed ammonia emission inventory for China on a 1 km × 1 km grid, and it can be used for global and regional air quality simulation. This inventory can be accessed through our website (http://phy-en.pku.edu.cn/pdf/inventory/NH3-2006.rar) or by contacting the corresponding author.

[47] **Acknowledgments.** The MCD45A1 burned area product was provided by the University of Maryland. The MODIS NDVI, VCF, and 8-day thermal anomalies/fire products were provided by the Land Process Distributed Active Archive Center (LPDAAC), USA. The China Land Cover product was provided by the Environmental and Ecological Science Data Center for West China, National Natural Science Foundation of China. The 1 km population distribution data set was developed by the Data Center for Resources and Environmental Sciences Chinese Academy of Sciences (RESDC). This study was funded by National Natural Science Foundation of China (40975088 and 70821140353), the Public Welfare Projects for Environmental Protection (200809018), and National Program on Key Basic Research Project of China (973) under grant 2010CB428501.

References

Anderson, N., R. Strader, and C. Davidson (2003), Airborne reduced nitrogen: Ammonia emissions from agriculture and other sources, *Environ. Int.*, *29*(2–3), 277–286, doi:10.1016/S0160-4120(02)00186-1.

Andreae, M. O., and P. Merlet (2001), Emission of trace gases and aerosols from biomass burning, *Global Biogeochem. Cycles*, *15*(4), 955–966, doi:10.1029/2000GB001382.

Asman, W. A. H. (1992), Ammonia emissions in Europe: Updated emission and emission variations, report, 97 pp., Natl. Inst. of Public Health and Environ. Prot., Bilthoven, Netherlands.

Beusen, A. H. W., A. F. Bouwman, P. S. C. Heuberger, G. Van Drecht, and K. W. Van Der Hoek (2008), Bottom-up uncertainty estimates of global ammonia emissions from global agricultural production systems, *Atmos. Environ.*, *42*(24), 6067–6077, doi:10.1016/j.atmosenv.2008.03.044.

Bouwman, A. F., D. S. Lee, W. A. H. Asman, F. J. Dentener, K. W. VanderHoek, and J. G. J. Olivier (1997), A global high-resolution emission inventory for ammonia, *Global Biogeochem. Cycles*, *11*(4), 561–587, doi:10.1029/97GB02266.

Bouwman, A. F., L. J. M. Boumans, and N. H. Batjes (2002), Estimation of global NH_3 volatilization loss from synthetic fertilizers and animal manure applied to arable lands and grasslands, *Global Biogeochem. Cycles*, *16*(2), 1024, doi:10.1029/2000GB001389.

Buijsman, E., H. F. M. Maas, and W. A. H. Asman (1987), Anthropogenic NH_3 emissions in Europe, *Atmos. Environ.*, *21*(5), 1009–1022, doi:10.1016/0004-6981(87)90230-7.

Cai, H., and S. D. Xie (2007), Estimation of vehicular emission inventories in China from 1980 to 2005, *Atmos. Environ.*, *41*(39), 8963–8979, doi:10.1016/j.atmosenv.2007.08.019.

Cai, G. X., Z. L. Zhu, A. C. F. Trevitt, J. R. Freney, and J. R. Simpson (1986), Nitrogen loss from ammonium bicarbonate and urea fertilizers applied to flooded rice, *Fert. Res.*, *10*(3), 203–215, doi:10.1007/BF01049350.

Cai, G. X., D. L. Chen, H. Ding, A. Pacholski, X. H. Fan, and Z. L. Zhu (2002), Nitrogen losses from fertilizers applied to maize, wheat and rice in the North China Plain, *Nutr. Cycl. Agroecosyst.*, *63*, 187–195, doi:10.1023/A:1021198724250.

Charlson, R. J., S. E. Schwartz, J. M. Hales, R. D. Cess, J. A. Coakley, J. E. Hansen, and D. J. Hofmann (1992), Climate forcing by anthropogenic aerosols, *Science*, *255*(5043), 423–430, doi:10.1126/science.255.5043.423.

China Agricultural University (CAU) (1978), *Fertilizer Manual* [in Chinese], 452 pp., China Agric. Press, Beijing.

China Agricultural University (CAU) (1997), *Coprology of Domestic Animals* [in Chinese], 328 pp., Shanghai Jiaotong Univ. Press, Shanghai, China.

China's Association of Automobile Manufacturers (2007), *China Automotive Industry Yearbook* [in Chinese], 672 pp., China Automot. Ind. Yearb. Press, Beijing.

Clarisse, L., C. Clerbaux, F. Dentener, D. Hurtmans, and P. F. Coheur (2009), Global ammonia distribution derived from infrared satellite observations, *Nat. Geosci.*, *2*(7), 479–483, doi:10.1038/ngeo551.

Corstanje, R., G. J. D. Kirk, M. Pawlett, R. Read, and R. M. Lark (2008), Spatial variation of ammonia volatilization from soil and its scale-dependent correlation with soil properties, *Eur. J. Soil Sci.*, *59*(6), 1260–1270, doi:10.1111/j.1365-2389.2008.01087.x.

Department for Environment, Food and Rural Affairs (DEFRA) (2005), Evaluation of targeted or additional straw use as a means of reducing NH_3 emissions from buildings housing cattle and pigs, *Rep. AM0103*, 20 pp., London.

Dockery, D. W., C. A. R. Pope, X. Xu, J. D. Spengler, J. H. Ware, M. E. Fay, B. G. Ferris Jr., and F. E. Speizer (1993), An association between air pollution and mortality in six U.S. cities, *N. Engl. J. Med.*, *329*(24), 1753–1759, doi:10.1056/NEJM199312093292401.

Editorial Office of China Animal Industry Yearbook (EOCAIY) (2007), *China Animal Industry Yearbook* [in Chinese], 400 pp., China Agric. Press, Beijing.

European Environment Agency (EEA) (2006), EMEP/CORINAIR emission inventory guidebook 2006, *Tech. Rep. 11/2006* , Copenhagen.

European Environment Agency (EEA) (2009), EMEP/EEA air pollutant emission inventory guidebook 2009, *Tech. Rep. 9/2009*, Copenhagen.

Fan, X. H., Y. S. Song, D. X. Lin, L. Z. Yang, and J. M. Zhou (2005), Ammonia volatilization losses from urea applied to wheat on a paddy soil in Taihu Region, China [in Chinese], *Pedosphere*, *15*(1), 59–65.

Fan, X. H., Y. S. Song, D. X. Lin, L. Z. Yang, and J. F. Luo (2006), Ammonia volatilization losses and N-15 balance from urea applied to rice on a paddy soil [in Chinese], *J. Environ. Sci.*, *18*(2), 299–303.

Fang, M., C. K. Chan, and X. H. Yao (2009), Managing air quality in a rapidly developing nation: China, *Atmos. Environ.*, *43*(1), 79–86, doi:10.1016/j.atmosenv.2008.09.064.

Fillery, I. R. P., J. R. Simpson, and S. K. Dedatta (1986), Contribution of ammonia volatilization to total nitrogen loss after applications of urea to wetland rice fields, *Fert. Res.*, *8*(3), 193–202, doi:10.1007/BF01048620.

Gao, X., and W. Ma (2002), Analysis on the current status of utilization of crop straw in China [in Chinese], *J. Huazhong Agric. Univ.*, *21*(3), 242–247.

Garcia, P., A. Benarroch, and J. M. Riera (2008), Spatial distribution of cloud cover, *Int. J. Satell. Comm. Networking*, *26*(2), 141–155, doi:10.1002/sat.899.

He, K. B., F. M. Yang, Y. L. Ma, Q. Zhang, X. H. Yao, C. K. Chan, S. Cadle, T. Chan, and P. Mulawa (2001), The characteristics of PM2.5 in Beijing, China, *Atmos. Environ.*, *35*(29), 4959–4970, doi:10.1016/S1352-2310(01)00301-6.

Huang, X., M. Li, J. Li, and Y. Song (2012), A high-resolution emission inventory of crop burning in field in China based on MODIS Thermal Anomalies/Fire products, *Atmos. Environ.*, *50*, 9–15, doi:10.1016/j.atmosenv.2012.01.017.

Hutchings, N. J., S. G. Sommer, J. M. Andersen, and W. A. H. Asman (2001), A detailed ammonia emission inventory for Denmark, *Atmos. Environ.*, *35*(11), 1959–1968, doi:10.1016/S1352-2310(00)00542-2.

Ianniello, A., F. Spataro, G. Esposito, I. Allegrini, E. Rantica, M. P. Ancora, M. Hu, and T. Zhu (2010), Occurrence of gas phase ammonia in the area of Beijing (China), *Atmos. Chem. Phys.*, *10*(19), 9487–9503, doi:10.5194/acp-10-9487-2010.

Jones, C. A., R. T. Koenig, J. W. Ellsworth, B. D. Brown, and G. D. Jackson (2007), Management of urea fertilizer to minimize volatilization, *Ext. Bull. 173*, Mont. State Univ., Bozeman.

Ju, X. T., et al. (2009), Reducing environmental risk by improving N management in intensive Chinese agricultural systems, *Proc. Natl. Acad. Sci. U. S. A.*, *106*(9), 3041–3046, doi:10.1073/pnas.0813417106.

Kim, J. Y., C. H. Song, Y. S. Ghim, J. G. Won, S. C. Yoon, G. R. Carmichael, and J. H. Woo (2006), An investigation on NH_3 emissions and particulate NH_4^+-NO_3^- formation in East Asia, *Atmos. Environ.*, *40*(12), 2139–2150, doi:10.1016/j.atmosenv.2005.11.048.

Kirk, G. J. D., and P. H. Nye (1991), A model of ammonia volatilization from applied urea. V. The effects of steady-state drainage and evaporation, *J. Soil Sci.*, *42*(1), 103–113, doi:10.1111/j.1365-2389.1991.tb00095.x.

Koerkamp, P. W. G. G., et al. (1998), Concentrations and emissions of ammonia in livestock buildings in Northern Europe, *J. Agric. Eng. Res.*, *70*(1), 79–95, doi:10.1006/jaer.1998.0275.

Krupa, S. V. (2003), Effects of atmospheric ammonia (NH_3) on terrestrial vegetation: A review, *Environ. Pollut.*, *124*(2), 179–221, doi:10.1016/S0269-7491(02)00434-7.

Kuenzler, E. J., and N. J. Craig (1986), Land use and nutrient yields of the Chowan River watershed, in *Watershed Research Perspectives*, edited by D. L. Correll, pp. 77–107, Smithsonian Inst., Washington, D. C.

Lal, R. (2005), World crop residues production and implications of its use as a biofuel, *Environ. Int.*, *31*(4), 575–584, doi:10.1016/j.envint.2004.09.005.

Li, G., B. Li, and D. Chen (2002), Ammonia volatilization from large field planted with winter wheat and summer maize [in Chinese], *Acta Agric. Boreali Sin.*, *17*(1), 76–81.

Li, J., F. Fang, B. Huang, X. Hu, X. Ju, and F. Zhang (2009), Ammonia volatilization from field-applied chicken manure in Beijing [in Chinese], *J. Agro-Environ. Sci.*, *28*, 1959–1964.

Li, S., and S. Ma (1993), Ammonia volatilization from calcareous soil II. The relationship between ammonia loss from N fertilizers and applied methods [in Chinese], *Agric. Res. Arid Areas*, *11*, 130–134.

Li, X. G., S. X. Wang, L. Duan, J. Hao, C. Li, Y. S. Chen, and L. Yang (2007), Particulate and trace gas emissions from open burning of wheat straw and corn stover in China, *Environ. Sci. Technol.*, *41*(17), 6052–6058, doi:10.1021/es0705137.

Lin, M. Y., T. Oki, T. Holloway, D. G. Streets, M. Bengtsson, and S. Kanae (2008), Long-range transport of acidifying substances in East Asia - Part I - Model evaluation and sensitivity studies, *Atmos. Environ.*, *42*(24), 5939–5955, doi:10.1016/j.atmosenv.2008.04.008.

Liu, D., F. Wang, L. Ma, W. Ma, and F. Zhang (2008), Estimation of NH_3 emission factor for pig manure in China [in Chinese], *Trans. Chin. Soc. Agric. Eng.*, *24*(4), 218–224.

Lu, D., X. Liu, and C. Wu (1980), The research on conditions affecting the volatilization of ammonium bicarbonate on the calcareous soil and prevention measures [in Chinese], *Shaanxi J. Agric. Sci.*, *6*, 7–10.

Mannebeck, H., and J. Oldenburg (1991), Comparison of the effect of different systems on ammonia emissions, in *Odour and Ammonia Emissions From Livestock Farming*, edited by V. C. Nielsen, J. H. Voorburg, and Pierre L'Hermite, pp. 42–49, Elsevier, London.

Martin, S. T., H. M. Hung, R. J. Park, D. J. Jacob, R. J. D. Spurr, K. V. Chance, and M. Chin (2004), Effects of the physical state of tropospheric ammonium-sulfate-nitrate particles on global aerosol direct radiative forcing, *Atmos. Chem. Phys.*, *4*, 183–214, doi:10.5194/acp-4-183-2004.

Meng, Z. Y., W. L. Lin, X. M. Jiang, P. Yan, Y. Wang, Y. M. Zhang, X. F. Jia, and X. L. Yu (2011), Characteristics of atmospheric ammonia over Beijing, China, *Atmos. Chem. Phys.*, *11*(12), 6139–6151, doi:10.5194/acp-11-6139-2011.

Ministry of Environmental Protection of People's Republic of China (MEPPRC) (2001), *The National Large-Scale Livestock Farming Systems Pollution Situation Investigation Technical Reports* [in Chinese], 59 pp., Beijing.

Möller, D., and H. Schieferdecker (1989), Ammonia emission and deposition of NHx in the G. D. R, *Atmos. Environ.*, *23*(6), 1187–1193, doi:10.1016/0004-6981(89)90145-5.

National Bureau of Statistics of China (NBSC) (2006), *China's Second National Agricultural Census Data Assembly* [in Chinese], 726 pp., China Stat. Press, Beijing.

National Bureau of Statistics of China (NBSC) (2007a), *China Statistical Yearbook 2007* [in Chinese], 1028 pp., China Stat. Press, Beijing.

National Bureau of Statistics of China (NBSC) (2007b), *China Industry Economy Statistical Yearbook* [in Chinese], 391 pp., China Stat. Press, Beijing.

National Bureau of Statistics of China (NBSC) (2007c), *Cost and income of Chinese farm produce 2007* [in Chinese], 703 pp., China Stat. Press, Beijing.

National Bureau of Statistics of China (NBSC) (2007d), *China Statistical Yearbook on Environment 2007* [in Chinese], 236 pp., China Stat. Press, Beijing.

National Bureau of Statistics of China (NBSC) (2007e), *China Rural Statistical Yearbook 2007* [in Chinese], 427 pp., China Stat. Press, Beijing.

National Bureau of Statistics of China (NBSC) (2008a), *China Rural Statistical Yearbook 2008* [in Chinese], 419 pp., China Stat. Press, Beijing.

National Bureau of Statistics of China (NBSC) (2008b), *China Statistical Yearbook on Environment 2008* [in Chinese], 248 pp., China Stat. Press, Beijing.

Niu, Y. W., L. Y. He, M. Hu, J. Zhang, and Y. L. Zhao (2006), Pollution characteristics of atmospheric fine particles and their secondary components in the atmosphere of Shenzhen in summer and in winter [in Chinese], *Sci. China Ser. B*, *49*(5), 466–474, doi:10.1007/s11426-006-2010-0.

Ohara, T., H. Akimoto, J. Kurokawa, N. Horii, K. Yamaji, X. Yan, and T. Hayasaka (2007), An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, *Atmos. Chem. Phys.*, *7*(16), 4419–4444, doi:10.5194/acp-7-4419-2007.

Olivier, J. G. J., A. F. Bouwman, K. W. Van der Hoek, and J. J. M. Berdowski (1998), Global air emission inventories for anthropogenic sources of NOx, NH_3 and N_2O in 1990, *Environ. Pollut.*, *102*, 135–148, doi:10.1016/S0269-7491(98)80026-2.

Pain, B. F., T. J. Van der Weerden, B. J. Chambers, V. R. Phillips, and S. C. Jarvis (1998), A new inventory for ammonia emissions from UK agriculture, *Atmos. Environ.*, *32*(3), 309–313, doi:10.1016/S1352-2310(96)00352-4.

Pearson, J., and G. R. Stewart (1993), The deposition of atmospheric ammonia and its effects on plants, *New Phytol.*, *125*(2), 283–305, doi:10.1111/j.1469-8137.1993.tb03882.x.

Qu, X. (1980), The research on the loss from ammonium nitrogen fertilizer on the calcareous soil (in Chinese), *Soils Fert.*, *3*, 31–35.

Seinfeld, J. H., and S. N. Pandis (2006), *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, 2nd ed., 1203 pp., John Wiley, Hoboken, N. J.

Song, Y., X. Fan, and D. Lin (2004), Ammonia volatilation from paddy fields in the Taihu lake region and its influencing factors [in Chinese], *Acta Pedol. Sin.*, *41*(2), 265–269.

Song, Y., D. Chang, B. Liu, W. J. Miao, L. Zhu, and Y. H. Zhang (2010), A new emission inventory for nonagricultural open fires in Asia from 2000 to 2009, *Environ. Res. Lett.*, *5*(1), 014014, doi:10.1088/1748-9326/5/1/014014.

Statistics and Census Service of Macao SAR Government (2007), *Yearbook of Statistics 2007* [in Chinese], 475 pp., Macau.

Stephen, M. R., D. S. Melissa, C. L. Holly, B. T. Kirstin, P. S. Randy, and P. E. H. Associates (2004), Estimating ammonia emissions from anthropogenic nonagricultural sources, final report, 87 pp., U.S. Environ. Prot. Agency, Washington, D. C.

Streets, D. G., et al. (2003), An inventory of gaseous and primary aerosol emissions in Asia in the year 2000, *J. Geophys. Res.*, *108*(D21), 8809, doi:10.1029/2002JD003093.

Sutton, M. A., C. J. Place, M. Eager, D. Fowler, and R. I. Smith (1995), Assessment of the magnitude of ammonia emissions in the United-Kingdom, *Atmos. Environ.*, *29*(12), 1393–1411, doi:10.1016/1352-2310(95)00035-W.

van Breemen, N., J. Mulder, and C. T. Driscoll (1983), Acidification and alkalinization of soils, *Plant Soil*, *75*(3), 283–308, doi:10.1007/BF02369968.

Van der Hoek, K. W. (1998), Estimating ammonia emission factors in Europe: Summary of the work of the UNECE ammonia expert panel, *Atmos. Environ.*, *32*(3), 315–316, doi:10.1016/S1352-2310(97)00168-4.

Vega, E., et al. (2010), Particulate air pollution in Mexico city: A detailed view, *Aerosol Air Qual. Res.*, *10*(3), 193–211.

Wang, J., W. Ma, and R. Jiang (2008), Analysis about amount and ratio of basal fertilizer and topdressing fertilizer on rice, wheat, maize in China [in Chinese], *Chin. J. Soil Sci.*, *39*(2), 329–333.

Wang, K., J. Liu, X. Chen, X. Dai, and S. Feng (2009), Pollutant production and discharge from livestock and poultry industries and land carrying capacity in Zhejiang Province [in Chinese], *Chin. J. Appl. Ecol.*, *20*(12), 3043–3048.

Wang, S., J. Liao, and Y. Hu (2009), A preliminary inventory of NH_3-N emission and its temporal and spatial distribution of China [in Chinese], *J. Agro-Environ. Sci.*, *28*(3), 616–629.

Wang, W., X. Lu, Y. Pang, and D. Tang (1997), Geographical distribution of NH_3 emission intensities in China [in Chinese], *Acta Sci. Circumstantiae*, *17*(1), 2–7.

Webb, J., and T. H. Misselbrook (2004), A mass-flow model of ammonia emissions from UK livestock production, *Atmos. Environ.*, *38*(14), 2163–2176, doi:10.1016/j.atmosenv.2004.01.023.

Webb, J., M. Ryan, S. G. Anthony, A. Brewer, J. Laws, M. F. Aller, and T. H. Misselbrook (2006), Cost-effective means of reducing ammonia emissions from UK agriculture using the NARSES model, *Atmos. Environ.*, *40*(37), 7222–7233, doi:10.1016/j.atmosenv.2006.06.029.

Whitehead, D. C., and N. Raistrick (1991), Effects of some environmental-factors on ammonia volatilization from simulated livestock urine applied to soil, *Biol. Fertil. Soils*, *11*(4), 279–284, doi:10.1007/BF00335848.

Xing, G. X., and Z. L. Zhu (2000), An assessment of N loss from agricultural fields to the environment in China, *Nutr. Cycl. Agroecosyst.*, *57*(1), 67–73, doi:10.1023/A:1009717603427.

Yan, X. Y., H. Akimoto, and T. Ohara (2003), Estimation of nitrous oxide, nitric oxide and ammonia emissions from croplands in East, Southeast and South Asia, *Global Change Biol.*, *9*(7), 1080–1096, doi:10.1046/j.1365-2486.2003.00649.x.

Ye, X. N., Z. Ma, J. C. Zhang, H. H. Du, J. M. Chen, H. Chen, X. Yang, W. Gao, and F. H. Geng (2011), Important role of ammonia on haze formation in Shanghai, *Environ. Res. Lett.*, *6*, 024019, doi:10.1088/1748-9326/6/2/024019.

Yin, S., J. Zheng, L. Zhang, and L. Zhong (2010), Anthropogenic ammonia emissions inventory and characteristics in the Pearl River Delta Region [in Chinese], *Environ. Sci.*, *31*(5), 1146–1151.

Zhang, M. G., L. J. Gao, C. Ge, and Y. P. Xu (2007), Simulation of nitrate aerosol concentrations over East Asia with the model system RAMS-CMAQ, *Tellus, Ser. B*, *59*(3), 372–380, doi:10.1111/j.1600-0889.2007.00255.x.

Zhang, Q., et al. (2009), Asian emissions in 2006 for the NASA INTEX-B mission, *Atmos. Chem. Phys.*, *9*(14), 5131–5153, doi:10.5194/acp-9-5131-2009.

Zhang, S., and Z. Zhu (1992), Losses of urea—Nitrogen applied to maize grown on a calcareous fluvo-aquic soil in North China Plain, *Pedosphere*, *2*(2), 171–178.

Zhang, Y., A. J. Dore, L. Ma, X. J. Liu, W. Q. Ma, J. N. Cape, and F. S. Zhang (2010), Agricultural ammonia emissions inventory and spatial distribution in the North China Plain, *Environ. Pollut.*, *158*(2), 490–501, doi:10.1016/j.envpol.2009.08.033.

Zhang, Y. S., S. J. Luan, L. L. Chen, and M. Shao (2011), Estimating the volatilization of ammonia from synthetic nitrogenous fertilizers used in China, *J. Environ. Manage.*, *92*(3), 480–493, doi:10.1016/j.jenvman.2010.09.018.

Zhao, D. W., and A. P. Wang (1994), Estimation of anthropogenic ammonia emissions in Asia, *Atmos. Environ.*, *28*(4), 689–694, doi:10.1016/1352-2310(94)90045-0.

Zhao, Y., C. P. Nielsen, Y. Lei, M. B. McElroy, and J. Hao (2011), Quantifying the uncertainties of a bottom-up emission inventory of anthropogenic atmospheric pollutants in China, *Atmos. Chem. Phys.*, *11*(5), 2295–2308, doi:10.5194/acp-11-2295-2011.

Zhou, K., Z. Lei, Z. Wang, J. Shi, and Z. Peng (2010), Estimation of annual total livestock/poultry excrement in Henan Province [in Chinese], *Chin. J. Eco-Agric.*, *18*(5), 1060–1065, doi:10.3724/SP.J.1011.2010.01060.

Zhu, Z. L., G. X. Cai, J. R. Simpson, S. L. Zhang, D. L. Chen, A. V. Jackson, and J. R. Freney (1989), Processes of nitrogen loss from fertilizers applied to flooded rice fields on a calcareous soil in North-Central China, *Fert. Res.*, *18*(2), 101–115, doi:10.1007/BF01049507.

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