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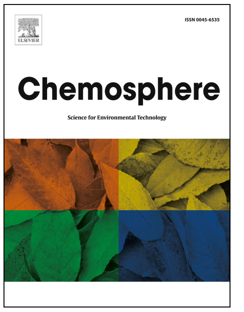

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High-resolution anthropogenic ammonia emission inventory for the Yangtze River Delta, China

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Abstract: The Yangtze River Delta (YRD) is one of the regions with air pollution and high ammonia (NH_3) emission in China. A high-resolution ammonia emission inventory for the YRD region was developed based on the updated source-specific emission factor (EFs) and the county-level activity data. The 1×1 km gridded emissions were allocated by using the appropriate spatial surrogate. The total NH_3 emissions changed insignificantly from 2006 to 2014 and varied in the range of 981.65 kt - 1014.30 kt. The fertilizer application and livestock were the major contributors of total emission. Humans, biomass burning and vehicles were the top three contributors of non-agricultural sources, accounting for 37.24%, 31.02% and 10.85%, respectively. Vehicles were calculated to be the non-agricultural source with the fastest annual growth rate. NH_3 emissions from the nitrogen fertilizer application generally peaked in summer, corresponding to the planting schedule and relatively high temperature. High NH_3 emissions occurred in the north as opposed to low emissions in the south of the YRD. The cities of Xuzhou, Yancheng and Nantong with more agricultural activities were demonstrated to have relatively high NH_3 emissions, contributing 10.0%, 9.0 and 7.1% of total emissions, respectively. The validity of the emission estimates was further evaluated based on the uncertainty analysis by Monte Carlo simulation, comparison with previous studies, and correlation analysis between NH_3 emission density and observed ground NH_3 concentration. A detailed NH_3 emission inventory is the basis of regional-scale air quality model simulation and can provide valuable information for understanding the formation mechanism of pollutants.

Keywords: Ammonia (NH_3); Emission inventory; Spatial distribution; Evaluation

1. Introduction

Ammonia (NH_3) is one of the most important trace gases in the atmosphere and has an important impact on the atmospheric environment and ecosystem. As the most abundant alkaline gas in the atmosphere, NH_3 can easily react with NO_x and SO_2 in the atmosphere and produce ammonium forms, such as $(NH_4)_2SO_4$, NH_4HSO_4, NH_4NO_3 and NH_4Cl. These forms of ammonium are important components of fine aerosol particles (Wang et al., 2011; Wu et al., 2015), which can cause aerosol pollution and can affect atmospheric visibility. NH_3 can also accelerate the nucleation process of sulphate particles, thus contributing to the formation of cloud condensation nuclei. In addition, most gaseous ammonia and particulate ammonium can be returned to soil or water by wet and dry deposition, which may lead to the acidification of soil, eutrophication of water and even a decrease in biological diversity (Anderson et al., 2003; Krupa, 2003; Matson et al., 2002). Although China has attached great importance to the treatment of air pollution, NH_3 was not a monitoring project stipulated in the National Environmental Air Quality Standard (http://www.mee.gov.cn/). There are various pollution sources of NH_3 due to the lack of corresponding reduction targets, causing sources such as industry and agriculture to discharge NH_3 without restraint.

The previous study indicated that the ammonium contributed approximately 23% and 17% of the total ion concentration in Nanjing and Shanghai, while this ratio reached 29% during the haze event in Suzhou, which significantly drove the formation of $PM_{2.5}$ during air pollution episodes in the YRD (Chen, 2017; Tian et al., 2016; Yu et al., 2019). For deposition of nitrogenous species, the study showed that nitrogen deposition was dominated by deposition of reduced nitrogen (NH_3+NH_4^+), which contributed approximately 67% to the total flux (Pan et al., 2012). The NH_3

dry deposition flux in Nanjing was 855.03 μg m^{-2} h^{-1} in 2017 and 1273.20 μg m^{-2} h^{-1} in 2018, respectively (Zhao, 2019). Because NH_3 plays a key role in the formation of fine particle and nitrogen deposition, a detailed and accurate inventory of NH_3 are crucial to the control strategies of particulate pollution, and the guidance of NH_3 limitation measures.

Since the early 1990s, many scholars have performed considerable research on ammonia emission inventories from anthropogenic sources (Battye et al., 2003; Olivier et al., 1998) and natural sources (Sarwar et al., 2005; Van et al., 1998). According to the EEA's (European Economic Area) NH_3 assessment report in 2013, the total NH_3 emissions of EEA member countries were calculated to be 428 wt in 2011, and agricultural sources accounted for 93.7% of the total emission. The NH_3 emission inventories indicated that nitrogen fertilizer application and livestock contributed more than 57% of global emissions (Bouwman et al., 1997) and more than 80% of the total NH_3 emissions in China (Kang et al., 2016; Zhou et al., 2015). Compared to agricultural sources, ammonia emissions from non-agricultural sources (including human excrement, traffic, waste treatment, industries and fossil burning) were calculated to be 739.6 kt, accounting for approximately 7.5% of the total ammonia emissions (Huang et al., 2012). NH_3 emission inventories have been established on various levels, such as national scale (Kang et al., 2016; Huang et al., 2012; Zhang et al., 2018), and regional scale, including the North China Plain (Zhang et al., 2010), the Beijing-Tianjin-Hebei (Zhou et al., 2015), the Pearl River Delta (Zheng et al.,2012), and the YRD (Dong et al., 2009), and also some provincial scale including Henan, Fujian and Sichan (Feng et al., 2015; Wang et al., 2018; Wu et al., 2017). However, previous emission inventories do not entirely reflect the local emission characteristics. The reason can be attributed to low-resolution activity data, the incomplete NH_3 emission sources and different emission factors.

With the rapid development of industrialization and urban agglomeration, the regional secondary pollution is becoming more serious in the YRD region. The prior study of ammonia emission cannot meet the needs of the current study on regional air quality and acid deposition. Therefore, a comprehensive NH_3 emission inventory in the YRD (25 cities) with high resolution and a 1 km × 1 km gridded emission allocation were developed in 2014 based on the activity data and emission factors of various ammonia emission sources.

2. Method and data

In this study, fertilizer application and livestock were regarded as agricultural sources. The non-agricultural sources included humans, the chemical industry, fossil burning, biomass burning, garbage treatment, sewage treatment, vehicles, pets and urban grassland.

NH_3 emissions of each source were calculated as a product of the activity data and specific EFs, according to the following equation (1):

$$E = \sum_i \sum_j (A_{i,j} \times EF_{i,j}) \quad (1)$$

where E is total NH_3 emissions, i , j represent the source type and prefecture-level city respectively, $A_{i,j}$ is the activity data, and $EF_{i,j}$ is the corresponding EF . The activity data of each source are derived from the local statistical yearbooks. The emissions factors are mainly adopted from the Technical Guidelines for Preparation of Atmospheric Ammonia Emissions Inventory from Ministry of Ecology and Environment of the People's Republic of China (2014) as well as domestic and foreign research results, and the former is preferred.

2.1 Agricultural source

2.1.1 Synthetic fertilizer application

Nitrogen fertilizer contains elemental nitrogen that is applied to crops. NH_3 is discharged into the atmosphere through microbial action or decomposition, resulting in significant volatilization of NH_3 in farmland areas. In general, NH_3 emissions from fertilizer are a function of the fertilizer type, soil properties (pH values, water content, calcium content, etc.), meteorological conditions (temperature, precipitation, and wind speed), application rate and method of application (injection or surface). In this study, we classified the synthetic fertilizers used in the YRD region as urea, ammonium bicarbonate (ABC), ammonium nitrate (AN), ammonium sulphate (AS), and other fertilizers (compound fertilizer related to NH_3 emissions were also considered, such as three-nutrient compound fertilizer and ammonium phosphate). In the statistical yearbooks, the first four types are classified as N fertilizer, while the other fertilizers are classified as phosphate fertilizer and compound fertilizer, respectively. The ratios of different fertilizer applications are listed in Table S1.

According to the method used in the National Ammonia Strategy Evaluation System (NARSES) N fertilizer module (DEFRA, 2001), the emission factor for fertilizer type is modified by functions relating to soil pH, land use, application rate, rainfall and temperature. The *EF* for a given scenario is calculated according to formula (2):

$$EF = EF_{MAX} \times RF_{PH} \times RF_{T} \times RF_{rain} \times RF_{rate} \times RF_{landuse} \quad (2)$$

where $EF_{\max}$ is the highest emission associated with each fertilizer type, RFs represents the reduction factors expressed as a proportion.

The $EF_{\max}$ for urea AS and ABC were considered to be 45%, while 4% for AN and other nitrogen fertilizers (Zhang and Luan, 2009). When ammonium nitrate was applied to lime soil and other soils, the RF_{PH} was considered to be 1 and 0.0889, respectively (Wang et al., 2018). The

RF_{PH} was also considered to be 1 when other types of nitrogen fertilizer were applied to all soils.

Because the partial pressure of NH_3 in solution increases exponentially with temperature, temperature is likely to be the most important meteorological factor for synthetic fertilizer application. According to the characteristics of temperature distribution in China, the RF_T of the original NARSES model was not suitable for the YRD region. Therefore, we used the modified functions for the estimation of ammonia volatilization associated with nitrogen fertilizer (Zhang et al., 2011), and calculated the RF_T for non-calcareous soil and calcareous soil using equations (3) and (4), respectively.

$$RF_T = EXP(0.2197225 \times (T_{mouth} - T_{annual})/3) \big/ 2 \quad (3)$$

$$RF_T = EXP(0.1386 \times (T_{mouth} - T_{year})/3) \big/ 2 \quad (4)$$

where T_{month} and T_{annual} are the local mean values of monthly and annual temperature, respectively, and T_{year} is the mean annual temperature in China. The meteorological parameters were derived from MICAPS (Meteorological Information Combine Analysis and Process System) ground observation data.

When urea is applied to soil, it hydrolyses to produce ammonium bicarbonate, an unstable compound that can quickly decompose to release gaseous NH_3. In the NARSES model, the influence of land use on NH_3 emissions was considered due to the reducing wind speed and temperature at the soil surface. In this study, a value of 0.7 was used for $RF_{landuse}$ for fertilizer applications in maize fields (Zhang and Luan, 2009). The higher fertilizer application rate and the pH value of soil surface causes a greater loss of NH_3.

2.1.2 Livestock and poultry

In this study, the ammonia emissions of livestock and poultry were estimated for each livestock category using headcount information at the county level and specific ammonia EFs. Livestock and poultry covered 9 subcategories: duck, chicken, rabbit, sheep, pig, buffalo, cattle, cow and beef cattle. The activity data of each subcategory was obtained from the China Rural Statistical Yearbook and local Statistical Yearbook, and detailed EFs for each animal listed in Table S7. We chose the number of stocks stored at the end of the year when the breeding period was longer than one year, while the number of slaughters was chosen for the livestock with a breeding period of less than one year.

2.2 Non-agricultural sources

NH_3 emissions from humans increase with the increase of the population. Generally, NH_3 emissions from humans include normal metabolic processes (respiration, perspiration and excretion). Population and sanitation are the key determinants of human NH_3 emissions. In this study, due to the different sanitary conditions of the toilet systems in rural and urban areas, NH_3 emissions in rural and urban areas were considered separately. NH_3 emissions from human were estimated by multiplying the year-end resident population in rural and urban areas by the corresponding emission factors (Shen, 2014).

Chemical industry sources include synthetic ammonia and nitrogen fertilizer production. Ammonia emissions were estimated using the city-level production output of industrial process, which was obtained from the local Statistical Yearbook and the corresponding EFs. The sources of ammonia in the combustion of fossil fuels mainly include natural gas, coal, gasoline, diesel, and kerosene. NH_3 emissions from fuel combustion were calculated as the product of industrial and domestic consumption and specific EFs. Biomass burning sources include waste straw burning, household straw burning, household firewood burning, and forest and grassland burning. Considering the topography and vegetation of the YRD region, the NH_3 emissions from forest and grassland fires were not considered. Based on the local cropping structure, a total of 7 crop types were considered and divided into domestic burning and in-field burning in this study. The related parameters of crop straw incineration were mainly selected from results related to the whole country and the adjacent areas of the YRD region. The annual yield data of the main crops in each city were obtained from the local Statistical Yearbook, and the total amount of pollutants discharged from the open-air combustion of waste was calculated according to equation (5):

$$E_i = \sum (P_{i,j} \times N_j \times F \times EF_j \times B_i) \quad (5)$$

where E is the ammonia emission, kt/a; i and j represent the area and crop type, respectively; P is the annual yield of crops, t/a; N is the ratio of the crop straw amount to crop yield; B is the drying ratio of straw; F is the burning rate of straw burning in open air; and EF is the ammonia emission factor of crops. Because the recommended EFs for these crop residues are significantly different, specific EFs are selected for each type of crop. The values of N, B, D and F are shown in Table S2.

NH_3 emissions from sewage treatment mainly come from three processes: microbial absorption in activated sludge of sewage treatment plants, nutrient treatment of digested sewage and sludge spreading. Referring to Gu et al. (2012), we calculated the NH_3 emissions based on the sewage discharge. Chinese Urban Construction Statistical Yearbook shows that waste disposal in the YRD region includes incineration and landfill. We estimated the NH_3 emissions according to the amount of domestic garbage disposal in each city. The NH_3 emissions from the waste incineration process were calculated based on equation (1). Sutton et al. (2000) showed that NH_3 emissions produced by landfills was considered to be 0.0073 times of methane emissions. The methane emissions can be calculated according to equation (6):

$$E = MSW \times MCF \times DOC \times DOC_F \times F \times 16/12 \quad (6)$$

where E represents the methane emissions, t/a; MSW is the landfill amount of domestic garbage, t/a; MCF is a methane correction factor, i.e., 1.0; DOC and DOC_F represent the content of degradable organic carbon in waste and the decomposition percentage of degradable organic carbon; the recommended values from the IPCC are 9% and 77%, respectively; *F* is the content of methane in landfill gas with a value of 0.5; and $16/12$ is the coefficient of carbon conversion to methane.

The exhaust emission of vehicle is an important NH_3 source in urban areas. Considering the rapid increase in the vehicle population from 2006-2014 in the YRD, the NH_3 emissions caused by on-road vehicles may change more significantly than that other non-agricultural sources. Activity data of vehicles are obtained by the Statistical Yearbook of Jiangsu, Shanghai and Zhejiang provinces. Referring to the classification by Zhang et al. (2013), a total of 9 vehicle types are considered in this study, and the definition of each vehicle type has been listed in Table S3. According to the technical guideline (Ministry of Ecology and Environment of the People's Republic of China, 2014), light-duty passenger vehicles, mini-duty passenger vehicles, light-duty truck and mini-duty truck are grouped into light-duty vehicles, and medium-duty passenger vehicles, heavy-duty passenger vehicles, medium-duty truck and heavy-duty truck are grouped into heavy-duty vehicles. According to vehicle classification and fuel type, motor vehicles can be divided into light gasoline vehicle, light gasoline truck, heavy gasoline vehicle, heavy gasoline truck, light diesel vehicle, light diesel truck, heavy diesel vehicle, heavy diesel truck and motorcycle. Because the fuel type data for vehicle classification in past years are rather limited, we referred to the fuel type share of various vehicle fleets observed by Che (2010). The NH_3 emissions of on-road vehicles were the product of the number of vehicles, the total annual mileage and the specific EFs. The annual average mileage of vehicles and specific EFs were adopted by Che (2010) and Ministry of Ecology and Environment of the People's Republic of China (2014) (See Table S4). In this study, we did not consider the impact of vehicle driving cycle on specific EFs due to the lack of fleet average speed.

NH_3 emissions from pets and urban grasslands were also calculated according to the method reported by Wu et al. (2017). According to the report by Wu et al. (2015), an average of one in 13 people keeps a pet in China, accounting for approximately 62% for dogs and 19% for cats, respectively. Homeless pets were not considered in this research. For urban grasslands, the annual NH_3 EF was 2.5% of the applied N fertilizer, and the EF was calculated to be 5 kg ha^{-1} yr^{-1} (Chinkin and Ryan, 2002). In this study, the emission data and factors of each source were summarized in Table S5, Table S6 and Table S7.

2.3 Spatial distribution of NH_3 emission

The 1 km×1 km gridded NH_3 emissions were established by the various sources based on ArcGIS software version 10.2 and different distribution modes (Wang et al., 2017a; Wang et al., 2017b). Based on the results of emission inventory, the gridded ammonia emission can be obtained by appropriate allocation factors. The spatial distribution area of anthropogenic ammonia was selected between the latitude of 27.14°N and 35.12°N, and between the longitude of 116.35°E and 122.95°E. The 1 km×1 km spatial resolution was set, covering Shanghai, Jiangsu Province and Zhejiang Province. Model domain information and central point coordinates are listed in S8 for the mapping of the several different emission sources and for estimating the total emissions. The form of spatial distribution includes point, line and area sources distribution based on the emission characteristics, spatial distribution and geographic information of emission sources. The point sources are regarded as stationary emission sources such as nitrogen fertilizer production, power plants and industrial boilers, while vehicle emission as the line source. The area sources include nitrogen fertilizer application, livestock, fuel and biomass combustion, and urban green land. The spatial allocation of point sources is to locate the emission to the corresponding grid according to the longitude and latitude coordinates of each enterprise. The spatial distribution of line source is based on the information of traffic road (including the national road, provincial road, county road, and urban expressways). Firstly, all kinds of road information are combined to calculate the total length of roads within the administrative area of each city. Then, the total length of roads in grid cell is calculated according to the defined grid. The ratio of the length of roads within the grid to the administrative area is taken as the distribution factor, and the ammonia emission of traffic sources in 2014 is taken as the distribution target to calculate the gridded ammonia emission. The allocation method from area sources is similar to that of line sources. For livestock, emissions from free-range and intensive livestock production were redistributed based on rural population density. Emissions from fertilization application and urban greenland were allocated by type of land use. Emissions from fuel combustion, pets, sewage treatment, garbage treatment and human waste were also regridded on the basis of population distribution. The data of land use types, population grids and rural residential area/land used in spatial distribution are from the National Earth System Science Data Centre (http://www.geodata.cn/index.html).

2.3 Uncertainty analysis

Quantitative analysis has great importance for emission inventories and can help to identify sources with high uncertainty to help make pointed references for the inventory in future work (Streets et al., 2003). The uncertainty parameters and sources of probability distribution are as follows: 5% uncertainty for statistical data from the yearbook, 10% uncertainty for department survey data and 15% uncertainty for data estimated by experts. The input data can generally be considered to be a normal distribution when the uncertainty range is less than 60%. Therefore, we assumed that the activity data and emission factors followed a normal distribution. The NH_3 emission range was estimated using a Monte Carlo simulation, and we ran 10000 Monte Carlo simulations to estimate the NH_3 emissions for each source. In this article, the lower limit range and upper limit range of the 95% confidence interval and the emission amount of each source were selected to calculate the uncertainty range.

2.4 Observed NH_3 concentration and analysis

Ammonia can easily be converted into ammonium (NH_4^+) by chemical reactions with acidic gas in the atmosphere. The concentration of ammonium observed in the environment can be used as the main chemical index for ammonia emission inventory assessment. Assuming that the emission of NH_3 is separated simply and rapidly between gaseous NH_3 and particulate NH_4^+ or between gaseous NH_3 and wet NH_4^+, the concentration of environmental NH_3 on the ground can be directly related to the emission density of NH_3 (Wang et al., 2018). Ambient NH_3 was observed in Nanjing from September 2015 to August 2016 using ANALYST passive samplers. The samplers were installed on the rooftop of the Meteorology Building of Nanjing University of Information Science and Technology (NUIST, lat: 32.2°N, long: 118.7°E, 30 m). The ANALYST sampler for NH_3 uses a treated glass microfibre filter as an absorbent. Each sampler was exposed for one month. Samples were retained in their airtight vials and remained frozen in the laboratory refrigerator until they were analysed within one month. The samples were extracted and analysed according to the analysis method recommended by the manufacturer. NH_3 was absorbed as ammonium by the samplers and was determined by ion chromatography using 5 ml of deionized water to extract the exposed filters. The filters were placed in glass vials containing 5 ml deionized water and were extracted for 1 hour with shaking every 5 or 6 minutes. The samples were then ready for ammonium analysis by ion chromatography.

3. Results and discussion

3.1 Annual NH_3 emissions

Fig. 2 illustrates the trends of total NH_3 emissions from 2006 to 2014 in the Yangtze River Delta. The total ammonia emissions showed an unobvious variation from 2006 to 2014 with the range from 982.95 kt to 1014.30 kt. From 2006 to 2014, nitrogen fertilizer application was the most important source of NH_3 emission in the YRD, and the average emission from this source accounted for approximately 55% of the total NH_3 emissions. This ratio was much higher than that reported in Fujian (39.4% in 2015, Wu et al., 2017) and the Beijing-Tianjin-Hebei region (28.6% in 2010, Zhou et al., 2015). Similar ratios have been observed in the YRD (49.3% in 2004, Dong et al., 2009) and all of China (54.0% in 2006, Dong et al., 2010). Livestock was also an important source and contributed 26-33% of the total NH_3 emissions. This result was mainly attributed to the breeding of a large number of livestock. On average, nitrogen fertilizer application and livestock combined accounted for 85%-90% of the total NH_3 emissions. However, the non-agricultural sources accounted for 10%-15%, which is consistent with the results of Kang et al. (2016) and Xu et al. (2016).

3.2 Contributions by different sources

3.2.1 Agricultural sources

Figure 3 shows the estimations of NH_3 emissions from nitrogen fertilizer application for the period 2006-2014. Annual NH_3 emissions from nitrogen fertilizer application commonly showed a decreasing trend, with a range of 532 kt to 600 kt during 2006-2014. From Figure 3, urea application was the largest source of nitrogen fertilizer application, accounting for approximately 60% of the emissions. NH_3 emissions from ammonium acid carbonate (ABC) ranged from 159 kt to 202 kt and contributed to approximately 30% of the emissions from nitrogen fertilizer application. NH_3 emissions from urea and ABC decreased by approximately 45 kt and 23 kt, respectively, from 2006 to 2014. It is worth noting that the emissions of NH_3 from other nitrogenous fertilizers increased by approximately 17 kt from 2006 to 2014. In general, the annual variation in NH_3 emissions reflects the changes in agricultural activities in the YRD. A distinct seasonal variation in NH_3 emissions from nitrogen fertilizer application in the YRD in 2014 is shown in Figure 4. Obviously, the maximum NH_3 emissions occurred in summer, and minimum emissions occurred in winter. The monthly NH_3 emissions were highest in July, with a value of 310 kt. This seasonal variation in NH_3 emissions was mainly related to temperature and agricultural activities. The winter wheat-summer rice rotation or two season rice system is common farming characteristic in the YRD. NH_3 volatilizations begin to rise with increasing temperatures, the new spring seeding and basal dressing fertilizer application in spring. The higher emissions occurred in summer were mostly attributed to the common contribution of higher temperature, tillering stage dressing of early rice and basal dressing of late rice. In fall, the majority of crops began to be harvested, and then the winter wheat was seeded with basal dressing. Despite a large amount of fertilizer used in fall, the lower NH_3 emissions were mainly related to the low temperature.

Ammonia emissions from livestock waste accounted for approximately 27% of the total emissions between 2006 and 2014. The livestock included beef, cattle, sheep, rabbit, buffalo, cattle, cow and poultry. As shown in Figure 5, pigs were the largest single source of livestock NH_3 emissions (approximately 80% of the emissions), and these emissions were significantly higher than those from other livestock sources. The second largest source was chickens, accounting for approximately 5% of livestock NH_3 emissions. These results are related to people's large demand for pigs and chickens. The other livestock sources contributed approximately 15% of the NH_3 emissions and varied from 38.47 kt to 57.54 kt. There were relatively higher NH_3 emissions from cattle and buffalo, which were due to the large amount of discharge of faecal material and urine during the breeding process. On average, the annual emissions from livestock from 2006 to 2014 ranged between 255.99 kt and 328.11 kt, with the highest emissions in 2012.

3.2.2 Non-agricultural sources

NH_3 emissions from the non-agricultural sources in the YRD between 2006 and 2014 are listed in Table 1. NH_3 emissions from non-agricultural sources contributed little to the ammonia emissions over the YRD region, accounting for approximately 10%-15% of the total emission and varying from 124.64 kt to 137.23 kt. This ratio was similar to a study in Fujian (17.5% in 2015, Wu et al., 2017) and China (19% in 2012, Kang et al., 2016). Humans were the largest non-agricultural sources for NH_3 emissions and contributed 37.2%-43.0% from 2006 to 2014. NH_3 emissions showed a declining trend for humans from 2006 to 2014, which was due to the decrease in rural depopulation under sanitary conditions and demography transition. Biomass burning was the second largest non-agricultural source of NH_3, contributing from 31.02% to 34.88% of the NH_3 emissions from non-agricultural sources. NH_3 emissions commonly showed a declining trend for humans, the chemical industry and biomass burning from 2006 to 2014, while NH3 emissions increased from vehicles, garbage treatment, grassland, sewage treatment, pets and fossil burning. It is worth noting that vehicles, garbage treatment and urban grassland were the fastest growing sources of NH_3 emissions, with emissions in 2014 being 2.9, 2.7 and 2.2 times higher, respectively, than those in 2006. These were mainly due to the increase in the number of vehicles, the production of more garbage with the improvement of living standards and beautifying the environment in urban construction planning. The NH_3 emissions from fossil burning, sewage treatment and pets showed a slight upward trend with the city expansion. In general, the NH_3 emissions from non-agricultural sources increased gradually, with the emissions in 2014 being 1.1 times those in 2006.

3.3 Contributions by city

The NH_3 emissions from various sources and the relative contributions from 25 cities over the YRD are summarized in Figure 6. Agricultural sources were the major sources of cities in the YRD, with a total contribution of 70%~90%. Nitrogen fertilizer application was the largest source in most cities in the YRD. However, livestock contributed the most in Shanghai (40.73%), Wenzhou (43.51%) and Zhoushan (41.63%) in 2014. The average NH_3 emissions of cities in the YRD reached 39.46 kt, which was much higher than that in PRD (21.74 kt in 2006, Zheng et al., 2012) and in Fujian (25.36 kt in 2015, Wu et al., 2017), but lower than that in the BTH region (121.05 kt in 2010, Zhou et al., 2015). NH_3 emissions from Shanghai, Jiangsu and Zhejiang provinces accounted for approximately 67.41%, 28.94% and 3.66%, respectively, of the YRD in 2014. Because the extensive breeding and large planting areas, high NH_3 emissions occurred in Jiangsu Province. Xuzhou had the highest NH_3 emissions, followed by Yancheng and Nantong, which exceeded the average NH_3 emissions by 59.84 kt, 49.30 kt and 30.35 kt, respectively. This was mainly due to the high production of livestock and poultry and the large area of cultivated agricultural land. Zhoushan had the minimum NH_3 emissions and accounted for only 0.36% of the total emissions. This can be attributed to the more islands and lesser planting areas in Zhoushan.

3.4 Spatial distribution

The 1 km × 1 km gridded NH_3 emissions in 2014 in the YRD region are shown in Figure 7. As shown in Figure 7a, the nitrogen fertilizer application source showed a large emission area and mainly concentrated in Jiangsu Province. It is worth noting that high NH_3 emissions were found in northern areas in Jiangsu Province, which was mainly related to the large planting areas. The NH_3 emission was estimated to be 4 t/km^2 in northern areas of Jiangsu Province, while the emission was only 2 t/km^2 in most areas of Zhejiang Province and Shanghai. The NH_3 emissions from livestock showed a similar trend as that observed for nitrogen fertilizer application (Figure 7b). The gridded value was below 40 t/km^2 in most areas, while the gridded ammonia emission in Nantong exceeded 80 t/km^2 due to the large amount of livestock. It is worth noting that there were sporadic high gridded emissions in Lishui with values over 80 t/km^2, which can be explained by the concentrated distribution of rural residential areas in Lishui.

The spatial distribution of ammonia emissions from humans, pets, garbage treatment, sewage treatment and domestic fuel sources was determined based on the population density to avoid unmanned areas such as oceans and forests. The gridded data of the population distribution in

China were obtained from the National Earth System Science Data Sharing Infrastructure. The ammonia emissions from population-related sources were concentrated in Shanghai and Jiangsu Province, while the emissions were relatively scattered in Zhejiang Province (Figure 7c). The high emission areas were mainly concentrated in Shanghai, Suzhou, Hangzhou and coastal areas of Wenzhou. Figure 7d shows that the high NH_3 emissions of urban grasslands were mainly concentrated in Shanghai, Hangzhou, Suzhou and Nanjing, and were dispersed in northern areas of Jiangsu Province and southern areas of Zhejiang Province, which were related to the areas of the urban scale and grassland areas. Obviously, higher NH_3 emissions from industry were concentrated in the central part of the YRD, such as Shanghai, Nanjing, Wuxi, and Suzhou (Figure 7e). These cities have many industrial enterprises and relatively high energy consumption, which led to a higher level of ammonia emissions. As shown in Figure 7f, high NH_3 emissions from vehicles mainly occurred in urban areas with dense populations, heavy traffic flows and crowded road networks, especially in Shanghai, Hangzhou, Wuxi and Suzhou.

The distribution of the agricultural sources showed a trend of high emissions in the northern area and low emissions in the southern area of the YRD, which was related to the amount of livestock and planting areas. In contrast, the high value of NH_3 emissions from non-agricultural sources in the YRD was concentrated in Shanghai with a gridded value higher than 8 t/km^2, while the gridded value was less than 2 t/km^2 in most areas of the YRD. Obviously, this result was related to the emissions from humans and vehicles, which accounted for 37.27% and 21.99%, respectively, of NH_3 emissions from the non-agricultural sources in Shanghai.

3.5 Comparison with previous estimates

The comparison of ammonia emissions in different regions is listed in Table 2. Consistent with the results of most studies, nitrogen fertilizer application was the largest source of NH_3 emissions in the YRD. The total emission in this study was calculated to be 1002.52 kt in 2006, which is less than that calculated by Dong et al. (2010) and higher than that calculated by Huang et al. (2012) in the same base year. This was mainly due to the differences in the emission sources and emission factors. Dong et al. (2010) used uniform emission factors for the same source over China, and these factors were derived mainly from European experiments rather than domestic measurements in China. The 5 ammonia emission sources (nitrogen fertilizer application, livestock, humans, vehicles and the industrial process of nitrogen fertilizer production) were considered in the studies by Wang et al. (1997), Dong et al. (2009) and Dong et al. (2010). In this study, we considered more NH_3 sources (domestic fuel, grassland and pets). In addition, some localized revision for nitrogen fertilizer application sources was made in the calculation of emission factors based on the local soil pH, temperature and some agricultural practices. The revised emission factors can better reflect the actual NH_3 emission of the nitrogen fertilizer application in the YRD.

3.6 Uncertainty analysis of NH_3 emissions

The uncertainties of the emission inventory were due to the lack of some activity levels and localized emission factors. The uncertainties of NH_3 emissions from various sources are listed in Table 3. Relatively high uncertainties were associated with livestock (-58%, 114%) and garbage treatment (-52%, 72%) sources. The higher uncertainty among these sources is mainly due to the following: (1) the emission factors were mainly from other research results and showed differences in different studies, especially for livestock. The emission factors were selected as the converted mean value, but there were differences in emission factors through different breeding methods. (2) In this study, the data related to cattle, buffalo and poultry data in some cities were calculated using provincial data because of the incomplete statistics in the statistical yearbook. The inaccuracy of source-level classification was also another reason for the uncertainty. More detailed parameters cannot be obtained from the statistical yearbook, such as the proportion of chickens and ducks. (3) In addition, there was great uncertainty in garbage treatment due to the emissions from the amount of CH_4 released from the landfill process. Compared with other studies, the reliability of ammonia emissions from nitrogen sources is higher because of the use of localized emission factors. As numerous parameters, including temperature, pH, precipitation, application rate and landuse, were considered in nitrogen fertilizer application, the uncertainty was relatively small. The overall estimated range of NH_3 emission was 496.29 kt to 1784.33 kt, corresponding to uncertainties of -55% to 60%.

3.7 Characteristics of ambient NH_3 concentration

The NH_3 emissions in Nanjing in 2014 were compared with the observed data. As shown in Figure S1, the concentration of observed NH_3 showed obvious seasonal variation, and the ranking of the seasonal average concentration was summer (19.25 $\mu g \cdot m^{-3}$) > autumn (15.33 $\mu g \cdot m^{-3}$) > spring (12.54 $\mu g \cdot m^{-3}$) > winter (9.89 $\mu g \cdot m^{-3}$). The observed NH_3 concentration in summer was twice as high as that in winter. The NH_3 concentration distribution peaked in the warm season and decreased to the lowest value in the coldest season. To compare and analyse the time variation in ammonia emissions, we analysed the molar ratios of $(2SO_2 + NO_2)/NH_3$ from September 2015 to August 2016 in Nanjing. The molar ratio was used to determine the state of NH_3; for example, NH_3 is suggested to be deficient when the value is greater than 1. The molar ratios from June to August were lower than that form other months, indicating that ammonia emissions in summer are higher than in other months. This change was consistent with the trend of the emission inventory in this study.

In addition, linear regression analysis was performed to investigate the relationship of the observed NH_3 concentrations with NH_3 emission density and temperature (Figure S2). The results showed that the observed NH_3 concentration increased with temperature (R^2 =0.79) and transformed NH_3 emission density (R^2 =0.57). The relationship suggests that the observed NH_3 concentrations displayed a moderate correlation with temperature and NH_3 emission density. This may be explained by the fact that the observed NH_3 concentration can be affected by the temperature, rainfall and position of the passive sampler. Further multi-site observations are required to quantify the contributions of local sources to the ambient NH_3 concentrations and spatial-temporal variations.

4. Conclusions

A comprehensive NH_3 emission inventory from 2006 to 2014 was developed based on activity data and specific EFs for agricultural and non-agricultural sources. Generally, the total ammonia emissions changed insignificantly between 2006 and 2014. The annual variations in emissions were mainly attributed to the type of fertilizer and the management of livestock. The NH_3 from nitrogen fertilizer application showed a decreasing trend, while the NH_3 from livestock showed an increasing trend. NH_3 emissions generally peaked in summer, corresponding to the planting schedule and relatively high temperature. Vehicles were calculated to be the fastest growing non-agricultural source in NH_3 emission inventory. In addition, the spatial grid allocation of the ammonia emission inventory was also carried out from point, area and line sources. The spatial pattern of the total emissions has been basically the same in recent years, with a trend of higher emissions in the northern area and lower emissions in the southern area of the YRD region.

The uncertainty range of emissions in the YRD was -55% to 60% using Monte Carlo simulation with 95% confidence. The assessments of uncertainty demonstrated that high uncertainty occurred in the emissions of livestock and waste treatment. Moreover, the seasonal averaged NH_3 concentration in Nanjing ranked in the order of summer > autumn > spring > winter. The molar ratio of $(2SO_2 + NO_2)/NH_3$ was consistent with the emission density of our study. A moderate correlation was observed between NH_3 concentration and temperature and NH_3 emission density.

Our results were different from previous published studies in this region, probably due to the selection of EFs and emission sources. There were still gaps in the results of emission inventories due to the lack of some activity data and emission factors. Therefore, our inventories should be further improved, especially for the estimation of ammonia emissions from agricultural sources.

Acknowledgments

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References

Anderson, N., Strader, R., Davidson, C., 2003. Airborne reduced nitrogen: ammonia emissions from agricultural and other sources. Environ. Int., 19, 3873-3883.

Andrea, R., Corry de Keizer., Jeroen P. Van der Sluijs., et al., 2008. Monte carlo analysis of uncertainties in the netherlands greenhouse gas emission inventory for 1990-2004. Atmos. Environ., 42, 8263-8272.

Bouwman, A.F., Lee, D.S., Asman, W.A.H., et al., 1997. A global high-resolution emission inventory for ammonia. Global Biogeochem. Cy., 11, 561-587.

Battye, W., Aneja, V.P., Roelle, P.A., 2003. Evaluation and improvement of ammonia emissions inventories. Atmos. Environ., 37, 3873-3883.

Che, W.W., 2010. A highly resolved mobile source emission inventory in the Pearl River Delta and assessment of motor vehicle. South China University of Technology, Guangzhou, China. (in Chinese)

Chen, Y., 2017. Ionic characteristic of atmospheric particles and precipitation in Shanghai. Shanghai Normal University, Shanghai, China. (in Chinese)

DEFRA (Department for Environment, Food and Rural A□airs), 2001. National Ammonia Reduction and Strategy Evaluation System (NARSES). Project AM0101.

Dong, Y.Q., Chen, C.H., Huang, C., et al., 2009. Anthropogenic emission and distribution of ammonia over the Yangtze River Delta. Acta Sci. Circumstantiae, 29(8), 1611-1671. (in Chinese)

Dong, W.X., Xing, J., Wang, S.X., 2010. Temporal and Spatial Distribution of Anthropogenic Ammonia Emissions in China: 1994-2006. Environ. Sci., 31, 1457-1463. (in Chinese)

EEA (European Environment Agency), 2013. EMEP/EEA Air Pollutant Emission Inventory Guidebook 2013, Tech. Rep. 12/2013, Copenhagen.

Feng, X.Q., Wang, X.R., He, M., et al., 2015. A 2012-based anthropogenic ammonia emission inventory and its spatial distribution in Sichuan Province. Acta Sci. Circumstantiae, 35, 394-401 (in Chinese).

Gu, Y,Y., Wang, B.G., Yang, J., et al., 2012. Ammonia source and emission factor in municipal wastewater treatment plant. Environ. Chem., 31, 146-151. (in Chinese)

Huang, X., Qiu, R., Chan, C.K., et al., 2011. Evidence of high $PM_{2.5}$ strong acidity in ammonia-rich atmosphere of Guangzhou, China. Transition in pathways of ambient ammonia to from aerosol ammonia at $[NH_4^+]/[SO_4^{2-}]=1.5$. Atmos. Res., 99,488-495.

Huang, X., Song, Y., Li, M.M., et al., 2012. A high-resolution ammonia emission inventory in China. Global Biogeochem. Cy., 26, GB1030,

Huang, R.J., Zhang, Y., Bozzetti, C., et al., 2014. High secondary aerosol contribution to particulate pollution during haze events in China. Nature, 514, 218-222.

Krupa, S.V., 2003. Effects of atmospheric ammonia (NH_3) on terrestrial vegetation: a review. Environ. Pollut., 124, 179-221.

Kang, Y.N., Liu, M.X., Song, Y., et al., 2016. High-resolution ammonia emissions inventories in China from 1980 to 2012. Atmos. Chem. Phys., 16, 2043-2058.

Matson, P., Lohse, K.A., Hall, S.J., 2002. The globalization of nitrogen deposition: consequences for terrestrial ecosystems. Ambio, 31,113-119.

Meng, Z.Y., Lin, W.L., Jiang, X.M., et al., 2011. Characteristics of atmospheric ammonia over Beijing, China. Atmos. Chem. Phys., 11, 6139-6151.

Ministry of Ecology and Environment of the People's Republic of China. Technical Guidelines for Preparation of Atmospheric Ammonia Emission Inventory (for Trial Implementation). 2014-8-19. http://www.mee.gov.cn/gkml/hbb/bgg/201408/W020140828351293771578.pdf

Olivier, J.G.J., Bouwman, A.F., Van Der Hoek, K.W., et al., 1998. Global air emission inventories for anthropogenic sources of NO_x, NH_3 and N_2O in 1990. Environ. Pollut., 102, 135-148.

Pan, Y.P., Wang, Y.S., Tang, G.Q., et al., 2012. Wet and dry deposition of atmospheric nitrogen at ten sites in Northern China. Atmos. Chem. Phys., 12, 6515-6535.

Shen, X.L., 2014. A highly resolved anthropogenic ammonia emission inventory in Guangdong Province and assessment of control strategies. South China University of Technology, Guangzhou, China (in Chinese).

Streets, D.G., Bond, T.C., Garmichael, G.R., et al., 2003. An inventory of gaseous and primary aerosol emissions in Asia in the year 2000. J. Geophys. Res., 108(21), 8809.

Sarwar, G., Corsi R.L., Kinney, K. A., et al., 2005. Measurements of ammonia emissions from oak and pine forests and development of a non-industrial ammonia emissions inventory in Texas. Atmos. Environ., 39, 7137-7153.

Sutton, M.A., Dragosits, U., Tang, Y.S., et al., 2000. Ammonia emissions from non-agricultural sources in the UK. Atmos. Environ., 34,856-863.

Tian, M.A., Wang, H.B., Chen, Y., et al., 2016. Characteristics of aerosol pollution during heavy haze events in Suzhou, China. Atmos. Chem. Phys., 16, 7357-7371.

Van Der Hoke, K.W., 1998. Estimating ammonia emission factors in Europe: summary of the Work of the UNECE Ammonia Expert Pane1. Atmos. Environ., 32, 315-316.

Wang, X.W., Lu, X.F., Pang, Y.B., et al., 1997. Geographical Distribution of NH_3 emission inventories in China, Acta Scientiae Circumstantiae, 17, 2-7. (in Chinese)

Wang, S.X., Xing, J., Jang, C., et al., 2011. Impact assessment of ammonia emissions on inorganic aerosols on inorganic aerosols in East China using response surface modeling technique. Environ. Sci. Tec., 45, 9293-9300.

Chinkin, L.R. and Ryan, P.A., 2002. Recommended Improvements to the CMU Ammonia Emission Inventory Model for Use by LADCO. (Prepared for Lake Michigan Air Directors Consortium).

Wu, S.P., Zhang, Y.J., Schwab, J.J., et al., 2017. High-resolution ammonia emissions inventories in Fujian, China, 2009-2015. Atmos. Environ., 162, 100-14.

Wang, K., Cao, J.J., Tian H.Z., et al., 2017. An emission inventory spatial allocate method based on POI data. China Environ. Sci., 37(6), 2377-2382. (in Chinese)

Wang, R.J., Wang, K., Zhang, F., et al., 2017. Emission Characteristics of Vehicles from National Roads and Province in China. Environ. Sci., 38(9), 3553-3560. (in Chinese)

Wang, R.Y., Ye, X.N., Liu, Y.X., et al., 2018. Characteristics of atmospheric ammonia and its relationship with vehicle emissions in a megacity in China. Atmos. Environ., 182, 97-104.

Wang, C., Yin, S.S., Bai, L., et al., 2018. High-resolution ammonia emission inventories with comprehensive analysis and evaluation in Henan, China, 2006-2016, Atmos. Environ., 193, 11-23.

Wang, C., 2015. Characteristics and prediction research of automobile emissions in Shandong province based on COPERT4 model. Ocean University of China, Qingdao, China. (in Chinese)

Xu, P., Liao, Y.J., Lin, Y.H., et al., 2016. High-resolution inventory of ammonia emissions from agricultural fertilizer in China from 1978 to 2008. Atmos. Chem. Phys., 16, 1207-1218.

Yu, X.N., Shen, L., Xiao, S.H., et al., 2019. Chemical and Optical Properties of Atmospheric Aerosols during the Polluted Periods in a Megacity in the Yangtze River Delta, China. Aerosol Air Qual. Res., 19, 103-117.

Zhang, M.S. and Luan, S.J., 2009. Application of NARSES in evaluation of ammonia emission from nitrogen fertilizer application in planting system in China. J. Anhui Agr. Sci., 37, 3583-3586. (in Chinese)

Zhang, Y.S., Luan, S.J., Chen, L.L., et al., 2011. Estimating the volatilization of ammonia from synthetic nitrogenous fertilizers used in China. J. Environ. Manage., 92, 480-493.

Zhang, L., Chen, Y., Zhao, Y., et al., 2018. Agricultural ammonia emissions in China: reconciling bottom-up and top-down estimates. Atmos. Chem. Phys., 18, 339-355.

Zhang, Y., Dore, A.J., Ma, L., et al., 2010. Agricultural ammonia emissions inventory and spatial distribution in the North China Plain. Environ. Pollut., 158, 490-501.

Zhang, S., Wu, Y., Liu, H., et al., 2013. Historical evaluation of vehicle emission control in Guangzhou based on a multi-year emission inventory. Atmos. Environ., 76, 32-42.

Zhao, X.F., 2019. Positioning observation and regional simulation of atmospheric nitrogen emission and deposition. Nanjing University, Nanjing, China. (in Chinese)

Zheng, J.Y., Yin, S.S., Kang, D.W., et al., 2012. Development and uncertainty analysis of a high-resolution NH_3 emissions inventory and its implications with precipitation over the Pearl River Delta region, China. Atmos. Chem. Phys., 12, 7041-7058.

Zhou, Y., Cheng, S.Y., Lang J.L., et al., 2015. A comprehensive ammonia emission inventory with high-resolution and its evaluation in the Beijing-Tianjin-Hebei (BTH) region, China. Atmos. Environ., 106, 305-317.

High-resolution anthropogenic ammonia emission inventory for the Yangtze River Delta, China

Xingna Yu[1,*], Li Shen[1,2], Xinhong Hou[1], Liang Yuan[3], Yuepeng Pan[4], Junlin An[1], Shuqi Yan[1]

Tables:

Table 1. Contributions to NH_3 emissions (kt) from non-agricultural sources in the YRD from 2006 to 2014

Table 2. Comparison of NH_3 emissions ($kt\cdot yr^{-1}$) with other published results in the YRD

Table 3. Uncertainty of ammonia emissions from various sources in the YRD in 2014

Table 1. Contributions to NH_3 emissions (kt) from non-agricultural sources in the YRD from 2006 to 2014.

	2006	2007	2008	2009	2010	2011	2012	2013	2014
Humans	55.02	52.16	52.14	52.10	51.42	51.35	51.24	51.18	51.11
Chemical industry	9.94	10.42	9.74	11.72	8.92	8.81	9.86	9.59	8.99
Fossil burning	6.74	6.74	6.30	6.39	6.52	6.80	6.97	6.94	7.37
Biomass burning	44.71	43.76	42.80	40.33	42.56	41.95	42.74	42.73	42.57
Garbage treatment	1.66	1.87	1.99	2.51	2.75	2.89	3.44	3.61	4.51
Sewage treatment	1.36	1.48	1.71	1.81	1.91	2.01	2.14	2.17	2.17
Vehicles	5.15	5.22	5.95	7.10	8.52	10.04	11.48	13.26	14.89
Pets	2.39	2.47	2.53	2.60	2.78	2.84	2.89	2.94	2.99
Urban grassland	1.21	1.38	1.50	2.06	2.14	2.32	2.47	2.53	2.63
Total	128.18	125.5	124.66	126.62	127.52	129.01	133.23	134.95	137.23

Table 2. Comparison of NH_3 emissions ($kt{\cdot}yr^{-1}$) with other published results in the YRD

Base Year	Total	Nitrogen Fertilizer application	Livestock	Humans	Vehicles	others	
1991	711.41	226.85	314.65	160.22	—	9.69	Wang et al,
2004	460.68	227.33	203.28	4.11	—	25.96	Dong et al,. 2009
2006	1416.22	957.06	364.64	87.10	—	7.42	Dong et al., 2010
2006	607.70	305.10	202.30	12.10	10.70	84.90	Huang et al., 2016
2006	1002.52	584.23	290.10	55.02	5.15	68.02	This study
2014	986.73	532.82	316.68	51.11	14.89	71.24	This study

Table 3. Uncertainty of ammonia emissions from various sources in the YRD in 2014

Source	Emission (kt)	Mean (kt)	Uncertainty range
Nitrogen fertilizer application	532.82	586.71	[-47%, +68%]
Livestock	316.68	391.63	[-58%, +114%]
Industry	8.99	7.49	[-49%, +27%]
Fossil burning	7.37	7.39	[-42%, +60%]
Traffic	14.89	12.77	[-60%,+86%]
Garbage treatment	4.51	4.40	[-52%, +72%]
Sewage treatment	2.17	2.20	[-46%, +67%]
Human	51.11	56.44	[-34%, +71%]
Biomass	42.57	40.20	[-36%, +40%]
Grassland	2.63	2.25	[-39%, +42%]
Pets	2.99	2.91	[-36%, +47%]
Total	986.73	1114.39	[-55%, +60%]

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Figures:

Figure 1. Location and administrative divisions of the Yangtze River Delta region

Figure 2. Interannual variation in total NH_3 emissions in the YRD from 2006 to 2014

Figure 3. Annual variation in total NH_3 emissions from nitrogen fertilizer application in the YRD from 2006 to 2014

Figure 4. Monthly variation of ammonia emissions from nitrogen fertilizer application in the YRD in 2014

Figure 5. Interannual variation in total NH_3 emissions from livestock waste in the YRD region from 2006 to 2014

Figure 6. City-specific NH_3 emissions from different sources in the Yangtze River Delta in 2014

Figure 7. Spatial distribution of the major NH_3 sources and the total emission in a 1 km × 1 km grid cell in the Yangtze River Delta

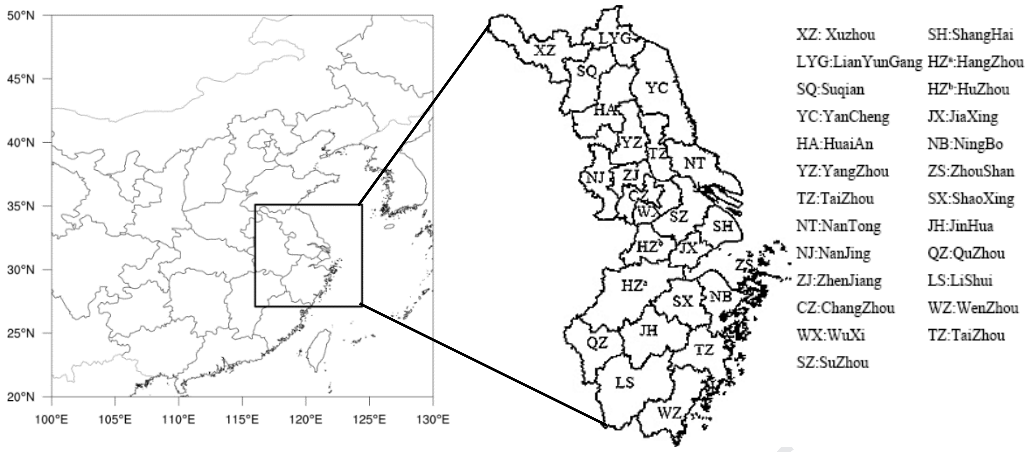

Figure 1. Location and administrative divisions of the Yangtze River Delta region

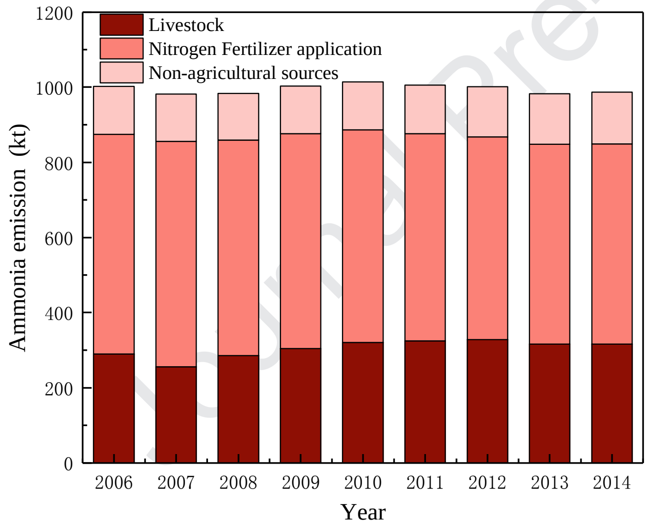

Figure 2. Interannual variation in total NH_3 emissions in the YRD from 2006 to 2014. The sources of emissions were divided into three categories: nitrogen fertilizer application, livestock, and non-agricultural sources (humans, chemical industry, fossil burning, biomass burning, garbage treatment, sewage treatment, vehicles, pets and urban grassland)

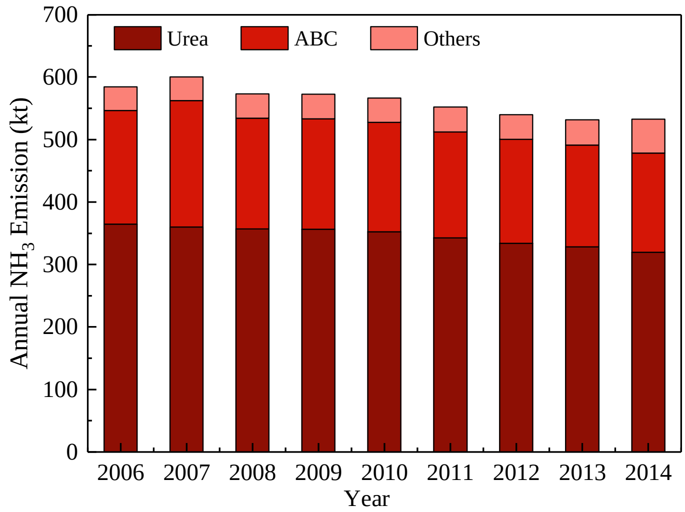

Figure 3. Annual variation in total NH_3 emissions from nitrogen fertilizer application in the YRD from 2006 to 2014. The types of sources included urea, ABC, and other nitrogenous fertilizers.

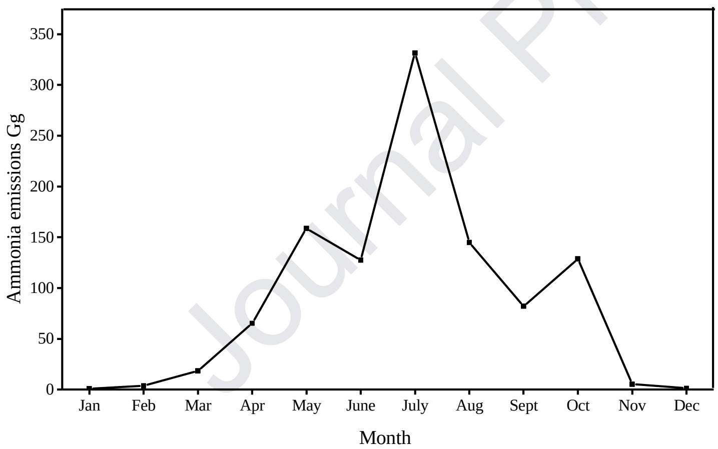

Figure 4. Monthly variation of ammonia emissions from nitrogen fertilizer application in the YRD in 2014.

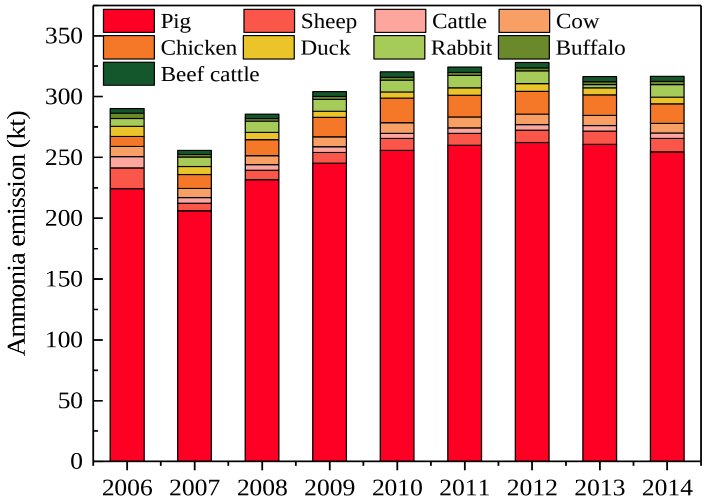

Figure 5. Interannual variation in total NH_3 emissions from livestock waste in the YRD region from 2006 to 2014.

The types of sources included duck, chicken, rabbit, sheep, pig, buffalo, cattle, cow and beef cattle.

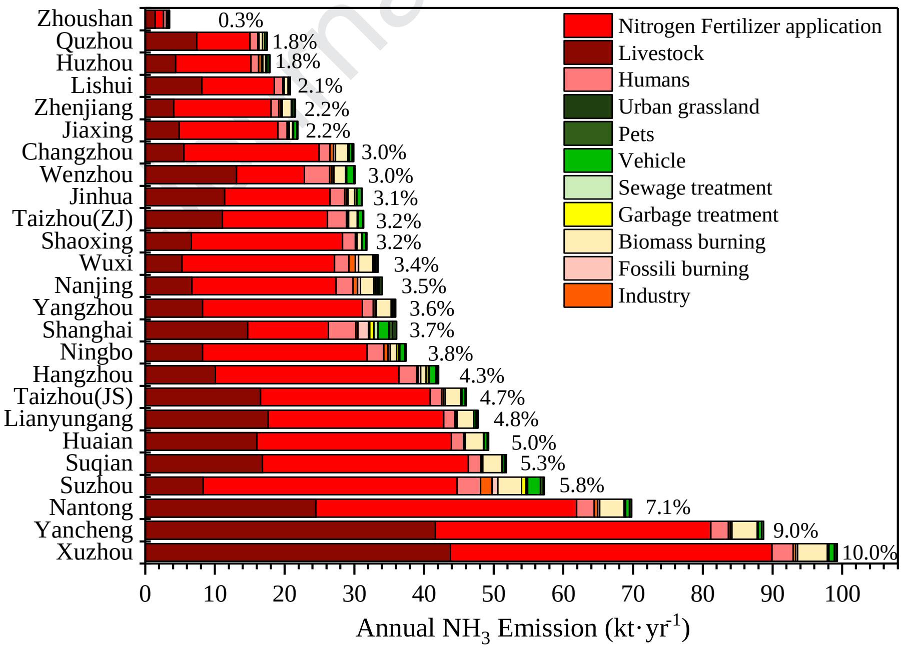

Figure 6. City-specific NH_3 emissions from different sources in the Yangtze River Delta in 2014.

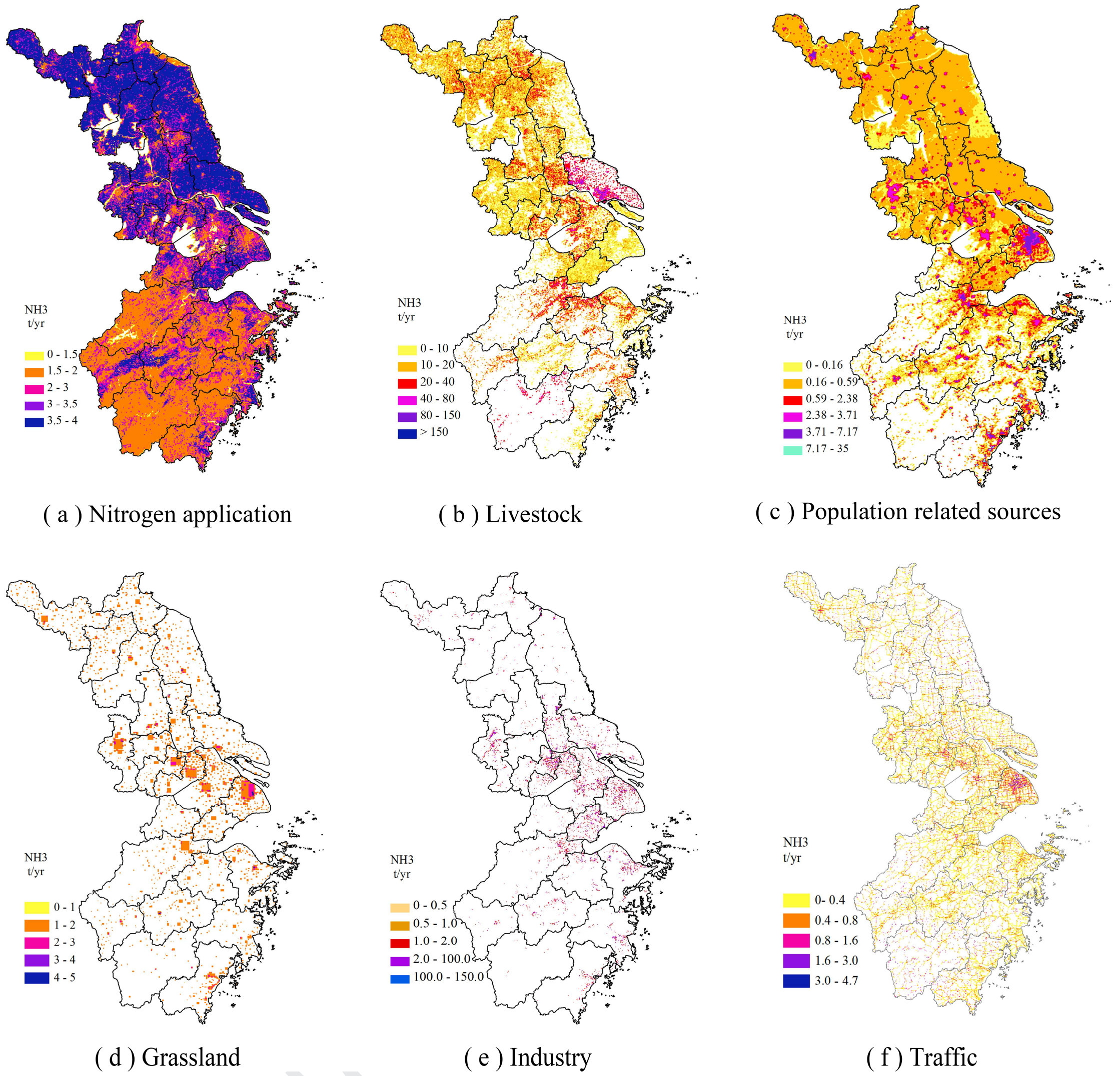

Figure 7. Spatial distribution of the major NH_3 sources and the total emission in a 1 km × 1 km grid cell in the Yangtze River Delta (2014). Non-point sources include humans, pets, garbage treatment, waste treatment and civilian fossil fuels.

Highlights

1. The 1km×1km gridded NH_3 emission inventory was developed based on county-level activity data.
2. The emission of ammonia in the YRD was 998.83 kt in 2014.
3. Uncertainty and correlation analysis were used to evaluate the inventory.

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: