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Agricultural ammonia emissions inventory and spatial distribution in the North China Plain

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Abstract

An agricultural ammonia (NH_3) emission inventory in the North China Plain (NCP) on a prefecture level for the year 2004, and a 5 × 5 km resolution spatial distribution map, have been calculated for the first time. The census database from China's statistics datasets, and emission factors re-calculated by the RAINS model supported total emissions of 3071 kt NH_3-N yr^{-1} for the NCP, accounting for 27% of the total emissions in China. NH_3 emission from mineral fertilizer application contributed 1620 kt NH_3-N yr^{-1}, 54% of the total emission, while livestock emissions accounted for the remaining 46% of the total emissions, including 7%, 27%, 7% and 5% from cattle, pigs, sheep and goats, and poultry, respectively. A high-resolution spatial NH_3 emissions map was developed based on 1 × 1 km land use database and aggregated to a 5 × 5 km grid resolution. The highest emission density value was up to 198 kg N ha^{-1} yr^{-1}.

Keywords: Ammonia; Emission inventory; Livestock emission; Mineral fertilizer application; Spatial distribution

The first high resolution spatial distribution of ammonia emissions for the North China Plain showed rates up to 200 kg NH_3-N ha^{-1} y^{-1} (Capsule).

1. Introduction

As a natural component of the atmosphere, ammonia (NH_3) plays an important role in atmospheric chemistry and ambient aerosol formation. Its emission rapidly increased during the 20th century due to the doubled or even tripled intensification of agricultural production (Galloway et al., 2004; 2008; Erisman et al., 2008). Ammonia is a major atmospheric pollutant, contributing to eutrophication, acidification and loss of biodiversity (Pearson and Stewart, 1993; Fangmeier et al., 1994; Krupa, 2003). Most emitted reactive nitrogen (N) will be deposited back on land (Goulding et al., 1998), even more so for NH_3 because of its short-distance transport (Asman et al., 1998).

This process, called ‘N deposition’, fertilizes ecosystems, influences the N cycle and introduces N saturation (Aber et al., 1989; Matson et al., 2002; Adams, 2003). Increased N availability in ecosystems can lead to rapid decline in species richness, even at the current N deposition level (Sala et al., 2000; Gotelli and Ellison, 2002; Stevens et al., 2004). In addition, NH_3 is a key precursor to neutralize H_2SO_4 and HNO_3 in the air and form $(NH_4)_2SO_4$, NH_4HSO_4 and NH_4NO_3 (Pinder and Adams, 2007; Erisman and Schaap, 2004; Walker et al., 2004; Olszyan et al., 2005), which contribute to reduced visibility, regional haze and health impacts associated with fine particular matter (PM).

Bouwman et al. (1997) estimated that the global NH_3 emission was about 54 Tg in 1990, half of which had been estimated to derive from Asia. In the total global NH_3 emission, 70% is estimated to be related to food production, and the other 30% is estimated to be related to natural sources, industrial processes, fossil fuels, etc. (Bouwman, et al., 1997; Olivier et al., 1998). A host of evidence shows that agricultural sources, i.e. volatilization from livestock manures and mineral fertilizer application, contribute to the major part of NH_3 emissions, approximately 80-90% of the total anthropogenic emission (Bouwman et al., 1997; Asman et al., 1998; Van Der Hoek, 1998; Battye et al., 2003).

Current researches on ammonia emissions inventories at large scales, such as global, Asian and national inventories, reveal that approximate 20% of global NH_3 emission comes from China (Zhao and Wang, 1994; Kilmont et al., 2001; Yamaji et al., 2004), especially from the intensive agricultural area (Yan et al., 2003; Wang et al., 2005). To feed 22% of the global population on 9% of the world’s arable land, China consumed more than 30% of the world’s total N fertilizer in the last decade (http://www.stats.gov.cn/; IFA). Moreover, intensive livestock husbandry has greatly developed in China due to the increasing requirement for livestock products. In 2006, China’s annual production of meat, eggs and milk were 80.5, 29.5 and 33.0 Mt, respectively, an increase of 4.2, 5.5 and 11.4 times those in the early 1980s, respectively (http://www.stats.gov.cn/) (Fig.1a). There was nearly no change for the expenditure on grain and eggs, while the expenditure on meat and milk were 1.3 and 4.8 times that in the previous decade, respectively (Fig. 1b). Meeting this demand was associated with a sharp increase in the use of mineral nitrogen fertilizers and intensive livestock production. This, in turn, has led to the growth of NH_3 emissions in China in the last decade.

The North China Plain (NCP) (Fig. 2a), which is called “China’s granary”, provides 40% and 25% of China’s wheat and corn production on 3.3% of the national area. Application rates of mineral nitrogen fertilizers in the NCP are up to 600 kg N ha^{-1} yr^{-1} (Zhao et al., 1997; Cui, 2005). Less than 30% efficiency in N application introduces about 40% N loss by various routes, including leaching of nitrate (NO_3^-) and emissions of NH_3, nitrous oxide (N_2O) and molecular nitrogen (N_2). In addition, about 30% of national animal products are also from this area, which further increases the ammonia emission. Significantly high N deposition has been found in this area: 27 kg N ha^{-1} yr^{-1} deposited in inorganic forms, with 67% in the form of ammonium (Zhang et al., 2008a) , and 9 kg N ha^{-1} yr^{-1} in organic forms (Zhang et al., 2008b) from bulk deposition; 55 kg N ha^{-1} yr^{-1} derived from dry deposition (Shen, in press); He et al. (2007) estimated the airborne N input up to 80-90 kg ha^{-1} yr^{-1} across the NCP using ^{15}N dilution method. All of this evidence supports the potential intensive NH_3 emission in the NCP.

In this study, estimated NH_3 emissions in the NCP were calculated at a 5 ×5 km grid resolution based on a Chinese national land use database (1 × 1 km grid resolution). Ammonia emissions from livestock farms were calculated using the agricultural model of RAINS (Klimont and Brink, 2004); emissions from mineral fertilizer application were estimated from earlier measurements. This work represents the first estimate of NH_3 emissions in China at high spatial resolution. In spite of some uncertainties, these data form the essential input for calculations with an atmospheric transport model (ATM) to estimate atmospheric concentrations of NH_3 and deposition of N, which can be applied to estimate exceedances of critical loads and levels and for testing the efficiency of future ammonia emissions abatement strategies.

2. Methodology

2.1 Database

Data for the year 2004 were used for both the fertilizer use and livestock populations. Primary census databases at prefecture level were obtained from the China Statistical Yearbook (http://www.stats.gov.cn/), the Beijing Statistical Yearbook (http://www.bjstats.gov.cn/), the Tianjin Statistical Yearbook (http://www.stats-tj.gov.cn/), the Hebei Statistical Yearbook (http://www.hetj.gov.cn/), the Henan Statistical Yearbook (http://www.ha.stats.gov.cn/), the Shandong Statistical Yearbook (http://www.stats-sd.gov.cn/), the Anhui Statistical Yearbook (http://www.ahtjj.gov.cn/) and the Jiangsu Statistical Yearbook (http://www.jssb.gov.cn/). The geographical database used in this study was published by the Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Science. It included the information of administration area and land use area, which covers arable land, forest, grass land, urban and industrial land etc., comprising 18 land types on a 1 ×

1 km latitude-longitude grid. Five provinces (Hebei, Henan, Shangdong, Jiangsu and Anhui) and two municipalities (Beijing and Tianjin) (Fig. 2a) (Table 1), including fifty-three prefectures (Fig. 2b) were covered in the study area. The average area of the prefectures is 8,452 km^2, with the minimum value of 2,153 km^2 for the Hebi prefecture in the Henan province and maximum value of 14,132 km^2 for the Cangzhou prefecture in the Hebei province (prefectures with only a part of their area in the NCP were not included).

2.2 Emission factors

Although a number of studies have been undertaken on NH_3 emission inventories as well as the emission factors (e.g. Klaassen, 1992, Bouwman et al., 1997, Misselbrook et al., 2000, Klimont, 2001, Doorn et al., 2002), measured emission factors for different sources specific to China do not yet exist. Emission factors for livestock (including poultry) used in European countries are summarized in Table 2. Many factors affect ammonia production, such as species, gender, age, body weight, N content of the feed, the conversion of N in feed to N in meat, milk and eggs, housing systems, litter/manure storage, spreading technique, proportion of time spent by livestock indoor and outside and climatic conditions (EEA, 2007). Bouwman et al. (1997) calculated NH_3 emission factors for livestock excretion for developing countries with the assumption of lower feeding levels, lower N content of the feed, a small portion of the livestock housed in stables and a higher temperature in the developing areas. Klimont (2001) adjusted the emission factors for Chinese specific production efficiency of milk or meat. Here, we re-calculated emission factors using the RAINS model methodology (Klimont and Brink, 2004) but considering specific Chinese factors in the calculation:

$$ef_1 = N_{x1} * v_1$$

$$ef_2 = N_{x1} * (1 - v_1) * v_2$$

$$ef_3 = N_{x1} * (1 - v_1 – (1 - v_1) * v_2) * v_3$$

$$ef_4 = N_{x4} * v_4$$

$$EF = ef_1 + ef_2 + ef_3 + ef_4$$

Where, $ef_{1,2,3,4}$ are NH_3-N loss at distinct emission stages, i.e., housing (1), storage (2), spreading (3), and grazing (4); $N_{x1,4}$ are N excretion during housing (1) and grazing (4); $v_{1,2,3,4}$ are NH_3-N volatilization rates at distinct emission stages; EF is the final emission factor. Livestock management specific to the Chinese situation was considered in the study, such as the household scale and industrial scale, housing system, manure storage, as well as the differences in different provinces.

Taking the emission factor for chicken as an example, all the parameters required in the RAINS model are listed in Table 3. The NH_3-N loss rates at different stages were combinations of the parameters from references (Asman, 1992, ECETOC, 1994, Hutchings et al., 2001, EEA, 2007a) and the specific Chinese livestock management systems. In the traditional households, chicken are free-range. NH_3-N loss rates are very high at the housing stage. The main storage method, composting of the manure, increases ammonia emission as well. However, in the intensive chicken farms, laying hens are caged, which gives a low NH_3-N loss rate in the housing stage. Anaerobic digestion is the main storage method for the manure of laying hens in intensive farms, which greatly reduces the ammonia emission. Broilers in intensive farms are kept with bedding material on the floor. For short production cycles, usually 55 days, manure is not treated during the whole period. Bedding material is cleaned once a production cycle finished. So, housing and storage processes are mixed for the broilers in intensive farms. For manure from both the traditional households and intensive farms, 3.5% and 5% NH_3-N are lost in the spring and early autumn spreading stages, respectively. There is no grazing stage for chicken, and the loss rates at this stage were set to zero. Finally, the traditional household management systems introduced much higher emission factors than the intensive farm management systems. In intensive chicken farms, the emission factors for broilers were comparable with the data cited from the reference (Table 2), while the emission factors for laying hens are lower than those for the caged housing stage.

Emission factors for all the other livestock sources from different provinces in the NCP were calculated by the same methodology and are listed in Table 4.

Ammonia emission from mineral fertilizer application is another important part of the agricultural sources. In China, the widely used N fertilizers are ammonium bicarbonate (ABC) and urea. Based on EEA (2007b) and ECETOC (1994), the NH_3-N loss rate of urea is 15%. There has been no research on the emission factor for ABC because of its small usage elsewhere. Bouwman et al. (1997) specially developed estimates of NH_3 loss from the application of ABC, which were up to 20-30%. The emission factors of both these fertilizers are higher than for other N fertilizers, e.g. ammonium sulphate, ammonium nitrate, calcium ammonium nitrate, anhydrous ammonia and so on. Cai et al. (2002) measured ammonia loss in the NCP in different cropping systems: 30-39% of N applied to rice, 11-48% of N applied to maize and 1-20% of N applied to wheat was lost as ammonia. Ding (2005) measured ammonia emissions in winter wheat-summer maize rotation systems in Beijing, typical of agricultural areas in the north part of the NCP, and found that 19.4% and 25.8% of N fertilizer applied was lost as ammonia for wheat and maize systems, respectively. Using a wind-tunnel system, Su (2006) found that NH_3 volatilization was up to 21.2% NH_3-N of urea-N within fifteen days after fertilization. Yan et al. (2003) estimated the emission from fertilizer application in Asian countries: NH_3-N losses of 23.5% and 13.7% were applied for urea in paddy fields and uplands, respectively, while NH_3-N losses of 34.5% and 20.5% applied for ABC in paddy fields and uplands, respectively. Instead of re-calculating emission factors, we used measured results directly and without distinguishing differences among provinces for the smaller variation in fertilizer application practice in the NCP. Considering 40% urea, 50% ABC and 10% of compound or other fertilizer applied in the NCP, 22% and 30% were used as the fertilizer emission factors in northern wheat-maize rotation system and southern wheat-rice rotation system in this study, respectively.

2.3 Spatial allocation

Ammonia emission from each source was allocated into a 5-km grid resolution using the ratio of area with different land use for the different sources. By the bottom-up process, all the NH_3 emissions from livestock and fertilizer use were distributed into the whole NCP region by the proportion of crops, grasslands, forests and so on.

A methodology similar to the AENEID model for obtaining the geographic distribution of ammonia emissions from livestock and fertilizer application (Dragosits et al., 1998; Dragosits, 1999; Hellsten et al., 2008) was used in this study. The basic theory was to allocate all the livestock and fertilizer to the best suited land cover types. The original NH_3 emission source for each category was distributed onto the appropriate area based on the management type. Dragosits (1999) assigned the practices including housing, storage, spreading, grazing over different land cover types, and then allotted the NH_3 emission from each source by the corresponding calculated weighted indexes.

Unlike in developed countries, livestock farms in the NCP operate at different scales, from household to industrial, and small farms are randomly scattered all over the place. It is difficult to calculate all the weighted indexes in detail. However, arable land appears in 95% of the total area in the NCP, while forests and grasslands only appear in 8% and 6% of the total area in the NCP, respectively (Fig. 3). There are overlaps of different land uses, such as agro-forestry and agro-pastoral ecosystems, as well as suburban areas with mixed properties of both rural and urban areas. Moreover, except for a tiny number of livestock which are kept part time outdoors by individual farmers, most of the livestock are kept indoors throughout the year in the livestock farms (except sheep and goats which are kept outdoors more time than other livestock). There is no NH_3 emission from agricultural sources considered in the forest and grassland areas excluding the grazing stage of cattle, sheep and goats, because most of the forest and grassland in the NCP are natural instead of semi-natural, as in developed countries. The intensive management practices in the agricultural areas (mainly arable land) greatly simplify the allocation process although some details are still missing. Ammonia emissions from housing and storage, spreading and grazing were allocated to probable rural residual areas, arable areas and grassland, respectively. The weighted index for each grid square was calculated by the percentage of the corresponding land uses. As for the mineral fertilizer application, NH_3 emission was allocated to the arable areas. The N fertilizer use is never homogenous because of the family operation on small plots, so an average value was applied. Although application of the same emission factors for mineral fertilizer use will introduce larger uncertainties, it is still the most advisable way for regional research at present. All emissions, independent of the management practices, were estimated within a prefecture region and mapped onto a 1-km grid resolution by the land use at first. Considering the areas of the prefectures in this study, 8,452 km^2 on average, and reduction of uncertainties, the resulting map was aggregated to a 5-km resolution.

3. Results and Discussion

3.1 NH_3 emission inventories

NH_3 emissions from different sources in every prefecture in the NCP are listed in Table 5, based on the census database and emission factors. The total NH_3 emission was as high as 3071 kt NH_3-N yr^{-1} in the NCP in 2004, including 1620 kt NH_3-N yr^{-1} from mineral fertilizer application and 1451 kt NH_3-N yr^{-1} from livestock sources. In all the livestock sources, 834 kt NH_3-N yr^{-1} was from pig emission, taking the largest part; the following 223 kt NH_3-N yr^{-1} and 228 kt NH_3-N yr^{-1} were from cattle and sheep (goats), respectively; and 166 kt NH_3-N yr^{-1} was from poultry.

Zhao and Wang (1994), Olivier et al. (1998) and Klimont (2001) estimated NH_3-N emissions in China corresponding to 11.1, 8.4 and 7.9 Tg in 1990. Klimont (2001) estimated NH_3-N emission in China to be 9.7 Tg in 1995. Streets et al. (2003) estimated NH_3-N emission in China to be 11.2 Tg in 2000. Regardless of the temporal variation and other minor emission sources in the NCP, the agricultural sources in the NCP contributed 27% of the recent NH_3 emission in China in 2000s. Comparing with the result from Hellsten et al., (2008), the total ammonia emission in the NCP was 15 times that in the UK, while the area ratio of the NCP to the UK is only 1.3. It means that the ammonia emission density based on grid cell in the NCP is more than 10 times that in the UK.

Sun et al. (1997) estimated NH_3 emission from the five provinces: Hebei, Henan, Shandong, Jiangsu and Anhui, and found 3.0 Tg NH_3-N emitted in 1995. Wang et al. (2003) made the same estimation, and found 4.9 Tg NH_3-N emitted in 2000. Taking the area ratios (Hebei: 43%; Henan: 46%; Shandong: 40%; Jiangsu: 35%; Anhui: 27%) of the five provinces inside the NCP into account, it would give approximately 1.2 and 1.9 Tg NH_3-N emissions for the areas in the NCP based on these two studies above, while our results gave 2.9 Tg NH_3-N from the same area, much higher than the former estimates. However, we cannot simply calculate emissions from the product of the emission and the area ratio of every province, because it was assumed that the emissions were homogenously distributed in every province during the calculation. Actually, intensive agriculture is concentrated in the NCP instead of all the seven provinces (including the two municipalities, Beijing and Tianjin). The areas in these provinces but outside the NCP were usually forests and grasslands with less ammonia emissions, which can be seen in the land use category maps in Fig. 3 and the subsequent spatial distribution section.

3.2 Contributions of NH_3 emission from different sources and provinces

Contributions of NH_3 emissions from different sources in the NCP and different provinces are shown in Fig. 4. Mineral fertilizer use contributed 54% to the total NH_3 emission in the NCP, while livestock sources contributed the remaining 46% (Fig. 4a), consistent with the results for the whole national scale in former studies (Zhao and Wang, 1994; Kilmont 2001; Streets, et al., 2003). In the different provinces, ammonia emissions from mineral fertilizer application accounted for 29%-81% to the total emissions (Fig. 4b-h), with lower ratio in the Tianjin municipality and higher ratio in the Jiangsu province. Livestock emissions constituted emissions from cattle (7%), pigs (27%), sheep and goats (7%) and poultry (5%) (Fig. 4a). Pig emissions constituted the largest proportion of the livestock emissions, which is related to people's dietary habits. In the different provinces, the ratios ranged from 14%-39% to the total emissions, with lower ratio in the Jiangsu province and higher ratio in the Shandong province. Contributions of emissions from cattle, sheep and goats, and chicken varied in different provinces, related to the different development strategies. Emissions from cattle now make a more significant contribution due to the sharply increased milk consumption in recent years (Fig. 1), although no temporal comparison was carried out here.

Of the total agricultural emissions in the NCP, 23% derived from the Hebei province, 26% derived from the Henan province, 26% from derived the Shandong province, 13% from derived the Jiangsu province, 8% derived from the Anhui province and 4% derived from the Bejing and Tianjin municipalities together (Fig. 5a). The averaged emission densities, emission per unit area, of the seven provinces are shown in Fig. 6, which reflects the contributions at relative areas of the seven different provinces. High emission density, more than 120 kg N ha^{-1} yr^{-1}, was found in the Shandong province, while a low emission density, 40 kg N ha^{-1} yr^{-1}, was found in the Beijing municipality because, in Beijing, the large urban area with less NH_3 emission shared the emissions, and gave a low emission density for the whole area. If only the rural area in Beijing was taken into account, the emission densities would have been as high as those in other areas, i.e. even more than 100 kg N ha^{-1} yr^{-1} (Fig. 7).

Contributions of the total livestock emission from different provinces were proportioned to the area percentages of each province to the NCP (Fig. 5b; Table 1): the Hebei, Henan and Shandong provinces took the larger part, followed by the Jiangsu and Anhui provinces, then the Beijing and Tianjin municipalities. Among all livestock sources, 223 kt yr^{-1} NH_3-N was from cattle emission. All cattle, buffalos, dairy cattle, horses, donkeys, and camels were included in this part for the former category of draught cattle. Actually there are only a small number of horses, donkeys and camels in the NCP nowadays. 67% of the emission came from dairy cattle because of the rapidly increased consumption of milk (Fig 1b). The Hebei province provided the largest part, 40% of the cattle emission. Unlike the fraction of the total emission, there was only 2% cattle emission from the Jiangsu province (Fig. 5c). Ammonia emission from pigs account for 27% of the total emission, more than half of all the livestock emissions. The Shandong province provided the largest part of pig and poultry emission, 36% and 37% of the two parts, respectively (Fig. 5 d & f). For the emission from sheep and goats, the three main provinces, Hebei, Henan and Shandong still took the leading parts (Fig. 5g). The Tianjin municipality shared 9% and the Jiangsu province shared only 3% of the emission, which was asymmetrical to the area percentages.

About 1620 kt yr^{-1} NH_3-N was emitted from mineral fertilizer application to arable land based on the census data of N-fertilizer consumption and the emission factors. Emission from mineral fertilizer application constituted the largest proportion of the overall emission, contrasting with the ratios in developed countries which have more than 80% of emission from livestock husbandry (Sutton et al., 1995; Dragosits et al., 1998; Olivier et al., 1998; Battye et al., 2003). This illustrated the overall very high level of fertilizer application commonly used in Chinese agriculture. Mineral fertilizer application in the Jiangsu province contributed 20% ammonia emission in this area

(Fig. 5h), twice as much as the other provinces considering the percentage of the area of the NCP. On the one hand, paddy rice (or wheat-rice rotation system) is the main crop system in the southern the NCP, while winter wheat-summer maize is the main crop rotation system in the north; on the other hand, the temperature in the south is higher than in the north, which was a key factor for the NH_3 emission from mineral fertilizer application (EEA, 2007).

3.3 Spatial distribution

According to the estimation of NH_3 emissions from each source (Table 4), the emission from every prefecture was distributed over land use by the bottom-up process (Fig. 7). Variations of livestock emissions were significant (Fig 6a-d) for the regional priority development strategies. High cattle emission was found in the Hebei and the Henan provinces, especially the Shijiazhuang and Hengshui prefectures in the Hebei province (Fig. 7a). High pig emission was found in the Shandong province, especially the west part which was inside the NCP (Fig 6b). Although the total contribution of sheep and goats emission was equal to the total contribution of cattle emission, the sheep and goats emission was more concentrated in the mid-south of the NCP, with a maximum of up to 31 kg N ha^{-1} yr^{-1} (Fig. 7c). Poultry emissions were up 18 kg N ha^{-1} yr^{-1}, and mainly concentrated in the mid-north of the NCP (Fig. 7d). The highest livestock emission was 113 kg N ha^{-1} yr^{-1} at a 5×5 km grid resolution, with high values in the north of the Tianjin municipality, in the south of the Hebei province, in the west of the Shandong province and the north of the Henan province (Fig. 7e).

The NH_3 emission from mineral fertilizer use was high, up to 124 kg N ha^{-1} yr^{-1}, which meant a great loss of N resources to the environment. High emissions appeared in the Shandong province, in the Jiangsu province and in some parts of the Hebei and Henan provinces (Fig. 7f), consistent with the over-use of N fertilizer in these intensive agricultural areas (Zhao et al., 1997; Cui, 2005). In contrast with the lower NH_3 emissions from livestock husbandry in the Jiangsu province, it turned out to be higher in fertilizer use emission. The large amount of N input, higher temperature and more precipitation in this area were all considered to be the main driving factors.

The total ammonia emission was as high as 198 kg N ha^{-1} yr^{-1} (Fig. 7g). High emissions were found in the middle of the NCP. Low emissions were found in the north and west of the Hebei province and most of the Anhui province, as well as individual emission sources in these two regions. It also gave a realistic explanation of the much higher NH_3 emission in the five main provinces in the NCP than the values simply calculated from the area ratio in section 3.1 above.

Area ratios of the 5-km grid ammonia emission density for the NCP in 2004 are shown in Fig. 8. Most areas of the NCP had an ammonia emission between 40-120 kg N ha^{-1} yr^{-1}. Hellsten et al. (2008) modelled the ammonia emission in the UK at 1-km resolution, and found ammonia emission higher than 30 kg N ha^{-1} yr^{-1} only on 1.3% of the area as 'hotspots', which was introduced by pig/poultry farming. Comparing with that result in the UK, ammonia emission higher than 30 kg N ha^{-1} yr^{-1} appeared on 92% of the area of the NCP in our study, which was nearly the whole regional area for intensive agricultural practices. Such a high NH_3 emission rate plays a major role in secondary particulate formation of $(NH_4)_2SO_4$, NH_4HSO_4 and NH_4NO_3 in this area (He et al, 2001; Yao, et al., 2003; Wang et al., 2005; Duan et al., 2006). Furthermore, most of the emitted ammonia will be deposited back to the land, and introduce ecological changes (Abler et al., 1989; Matson et al., 2002). Cape et al. (2009) introduced a new annual critical level of NH_3 for higher vegetation of 2-4 μg m^{-3}; sensitive plant species, such as lichens and bryophytes, were even more sensitive, with a critical level of 1 μg m^{-3}. As the preliminary output results of the N deposition in the NCP (unpublished), which was modelled by the ATM model with the input of the spatial distribution ammonia emission in this study, exceedance of the critical level for NH_3 appeared over 99.6% of the area in the NCP. Stevens et al. (2004) found one species loss per 2.5 kg N ha^{-1} yr^{-1} increase in N deposition across British acid grasslands. Although higher tolerance and less sensitivity of the crop system in the intensive agricultural area in the NCP resulted in ecological effects which have been ignored for a long time, the high NH_3 emissions will definitely bring in ecological changes even though there is little research to offer any evidence to date.

3.4 Uncertainties

Uncertainties of NH_3-N emissions in the NCP are associated with both the emissions inventory and the spatial allocation. The quality of the emissions inventory is further related to the quality of the input data and the emission factors. The spatial allocation is affected by land use maps and the data for agricultural practices, which are used to calculate the weight indexes for the different land use classes. Considerable uncertainty is associated with the emission factors and the agricultural practices. The input census data were from the China Statistic Bureau and local statistical bureaus, and the geographic maps were from the Institute of Geographic Sciences and

Natural Resources Research, Chinese Academy Sciences. Both data sources are currently the most authoritative available.

It was difficult to quantify the uncertainties in the emission factors, because there are few NH_3 emission measurements available in China. In this study, the emission factors were re-calculated by the RAINS model, taking the husbandry practices in the NCP into consideration, which was considered more consistent with reality than simply using emission factors from elsewhere. Some of the emission factors were quite different compared with previous studies, e.g. about twice those of the former sheep and goat factors. The main reason for this is the lower ratio of lambs, which are associated with a smaller emission. In contrast to European countries, Chinese people consume more mutton than lamb. In addition, instead of mostly indoor management like other livestock, sheep and goats usually graze more outdoors, which also increased the NH_3 emission. The emission factors of chicken in the intensive farms were also at the low level comparing with former studies in Table 2. The explanation had been given in the example of the calculation of the emission factor. Mineral fertilizer application emission factors were cited from measured results directly, without making any distinction among the provinces, as was done for livestock emissions for the whole homogenous land use and agricultural practice.

The spatial allocation processes introduce another area of uncertainty. Theoretically, the NH_3 emission from each prefecture would be bottom-up according to the corresponding area based on the land use maps. The agricultural practices determined the weighted indexes of different land cover categories. It was effective for the fertilizer use, but more difficult for livestock husbandry, because more processes (housing, storage, spreading and grazing) and the special traditional household managements in the NCP have to be considered for the latter. However, the homogenous farmland land cover and the mostly indoor livestock keeping in the NCP largely simplified the process. The major uncertainties were not from the theoretical process, but rather associated with the local situation. First of all, the operation of livestock husbandry ranges from household scale to industrial scale and is randomly scattered in the rural area. Second, organic manure use has dramatically declined and been largely replaced by mineral fertilizer use. Only 18% of N was from organic manure in 47% of arable land which still received organic nutrient in China (Ju et al., 2005). A lot of animal manure was neither treated nor recycled, particularly the liquid manure, and directly discharged into surface waters, which introduced further environmental problems. Third, the organic manure was not necessarily applied in the local area. It is mostly transported to other areas after production. In this study, all these three possibilities were not taken into consideration, and instead the emissions were assigned onto the local area.

In addition, all the census data were at prefecture level, and distributed into each prefecture. So, there were artificial boundaries between higher and lower emission areas in the spatial variation maps, which were not physically realistic. However, it is true that there were different priorities in the development strategy in different regions which introduced variations in both livestock husbandry and fertilizer use in crop farms even on a prefecture level. Although we can eliminate this if we use bottom-up emissions on a province level or the NCP regional level, it will actually obliterate more spatial realities at a higher resolution.

4. Conclusions

An inventory of NH_3 emissions at prefecture level in the NCP was carried out for the year 2004. The total agricultural NH_3-N emission was high (3071 kt yr^{-1}), accounting for 27% of the total emission in China, while the area ratio was only 3.3%. 1620 kt yr^{-1} NH_3-N emission derived from fertilizer application, which was the largest emissions source, constituting more than half of the total agricultural emission. Livestock emissions contributed 1451 kt N yr^{-1}, constituting: emissions from cattle (7%), pigs (27%), sheep and goats (7%) and poultry (5%). The Henan, Hebei and Shandong provinces made the largest contribution to the total emissions in the NCP.

A high-resolution map of NH_3 emissions in the NCP was developed for the first time based on census data and land use maps. The highest 5-km grid emission density was 198 kg N ha^{-1} yr^{-1}. High emissions were found in the south of the Hebei province, in the west of Shandong province and in most of the Henan province. Generation of this spatial distribution map of ammonia emissions will allow the opportunity to undertake atmospheric transport modelling to provide spatial estimates of wet and dry deposition of N in the NCP. Although there are still some uncertainties introduced by the lack of original emission factors in China, the emissions inventory and spatial distribution of NH_3 emissions provide indispensable input data for atmospheric transport, N deposition, critical load exceedance models and abatement strategies for China in future research.

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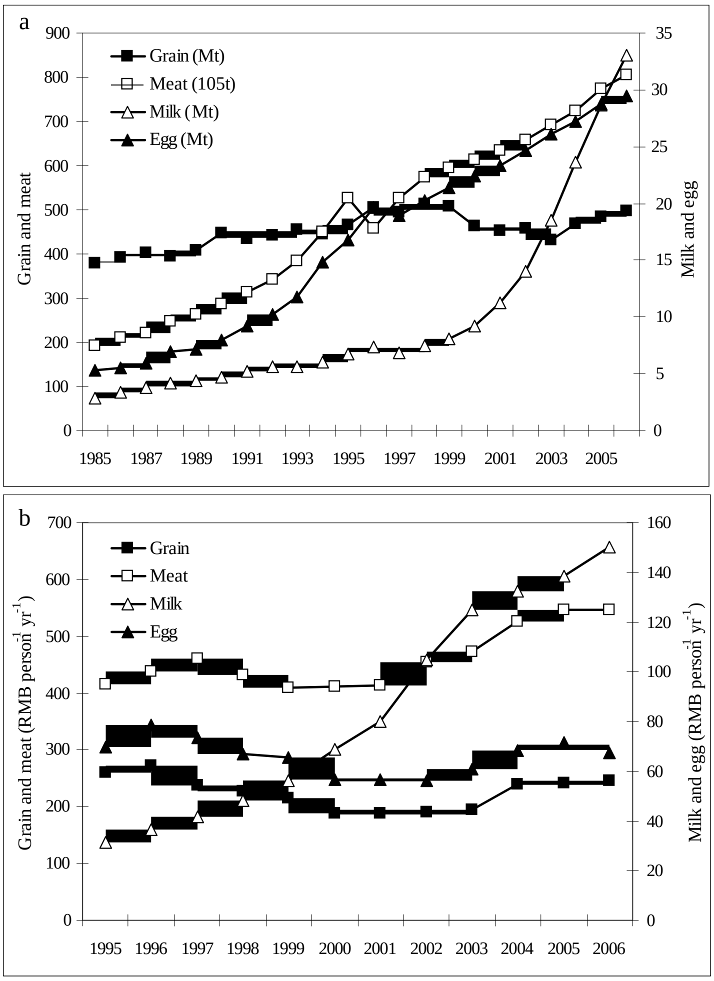

Figure 1. Agricultural products consumption in China in the last two decades (a) and the average human living expenditures in the last decade (b).

Note: RMB means Chinese Ren-Min-Bi (Yuan), the currency of the People's Republic of China

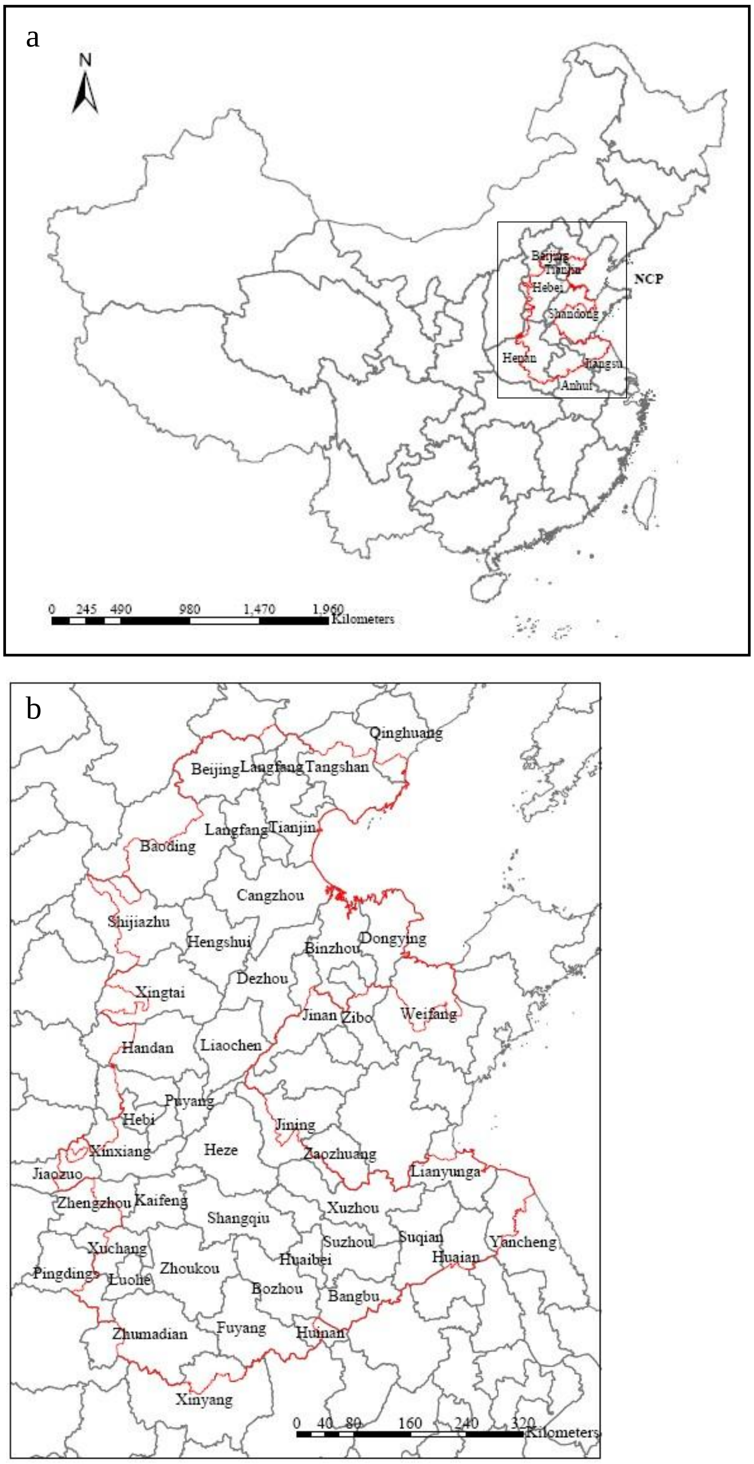

Figure 2. Location of the North China Plain (NCP) on a province scale (a) and prefecture scale (b) (Database for Beijing and Tianjin are on district level but not displayed here; red line is the boundary of the NCP)

Table 1. Areas and percentages of the provinces in the NCP

Province	Area (km^2)	Area in the NCP (km^2)	Percentage in the NCP (%)	Percentage of the NCP (%)
Beijing	16389	10059	61	3
Tianjin	11620	11620	100	4
Hebei	187292	79995	43	26
Henan	165541	75730	46	24
Shandong	154227	62062	40	20
Jiangsu	100929	35466	35	11
Anhui	140299	38363	27	12
Total	776297	313295	40	100

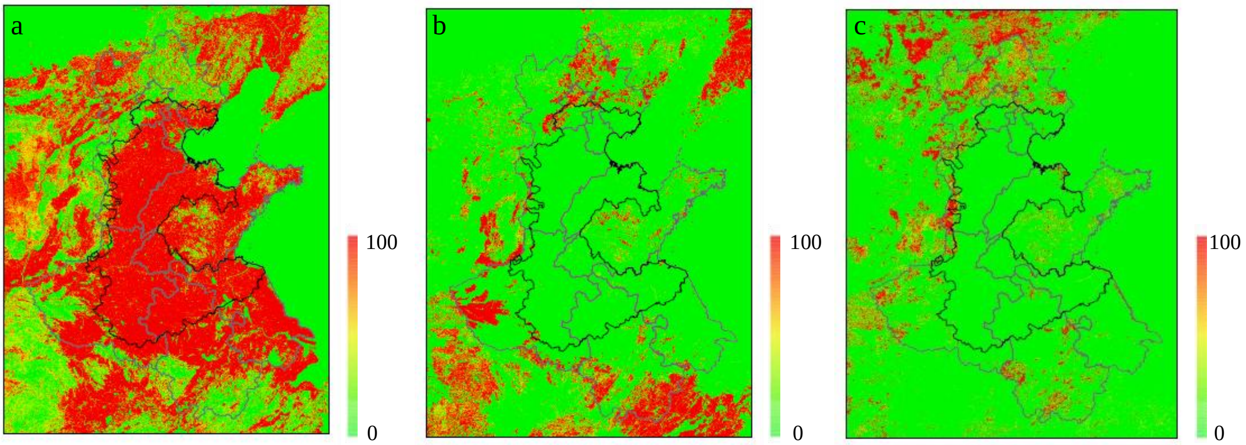

Figure 3. Land cover in the NCP (a. arable land; b. forest; c. grassland) (grey lines are the boundaries of the provinces involved in; black line is the boundary of the NCP)

Table 2. NH_3 emission factors (kg N head^{-1} yr^{-1}) from different references.

Category	Europe Van der Hoek, 1998	UK Misselbrook et al., 2000	Europe Klaassen, 1992	Developed counties Bouwman et al., 1997	Developing countries Bouwman et al., 1997	China Klimont, 2001
Dairy cattle	23.47	21.79	29.26	20.39	17.42	16.98-20.40
Other cattle	11.78	5.62	10.29	7.79	8.11	7.82-8.15
Pigs	5.26	3.95	4.22	3.95	3.95	3.95
Sow	13.53					
Sheep and goats	1.10	0.60	1.70	1.00	0.92	0.99
Horses (including donkeys)	6.59		12.29	7.58	8.70	8.73
Camels	8.65			7.58	8.70	10.62
Chicken (including other poultry)						
Laying hen	0.30	0.36	0.29	0.18	0.18	0.26
Broiler	0.23	0.19	0.15			0.15

Note: all the units have been transferred to kg N head^{-1} yr^{-1}, calculated from the units used in the original references.

Table 3. NHrN loss rates at different stages and the calculated emission factor **in** chicken farms **in** the NCP.

		Type of housing	N 1		Vz			EF
Traditional household	Laying hen	Free-range	0.80	40%	25%	3.5%-5%	0	0.46
	Broilers	Free-range	0.63	40%	25%	3.5%-5%	0	0.36
Intensive chicken farm	Laying hen	Caged	0.80	**11%**	2%	3.5%-5%	0	0.13
	Broilers	Floored	0.63	22%		3.5%-5%	0	0.16

*Units: N_1- kg N head· yr·; EF- kg N hear· yr·

Table 4. NH_3 emission factors (kg N $head^{-1}$ yr^{-1}) used in this study.

Category	Beijing	Tianjin	Hebei	Henan	Shandong	Jiangsu	Anhui
Dairy cattle							
(1-5 heads)	22.20	28.33	23.08	29.06	19.36	24.74	21.81
(>5 heads)	17.85	18.15	19.89	20.67	19.61	21.16	20.02
Meat cattle							
(1-50 heads)	6.54	8.35	6.8	8.57	5.71	7.29	6.43
(>50 heads)	5.26	5.35	5.86	6.09	5.78	6.24	5.90
other cattle	8.58	10.95	8.92	11.24	7.49	9.57	8.43
Horses	5.40	6.89	5.61	7.07	4.71	6.02	5.30
Donkeys	4.57	5.84	4.76	5.99	3.99	5.10	4.49
Pigs							
(1-50 heads)	5.06	6.46	5.27	6.63	4.42	5.65	4.98
(>50 heads)	3.38	3.44	3.77	3.92	3.72	4.01	3.79
Sheep and goats	2.14	2.73	2.22	2.80	1.86	2.38	2.10
Poultry *							
Laying hen (traditional house hold)	0.46	0.46	0.46	0.46	0.46	0.46	0.46
Broilers (traditional house hold)	0.36	0.36	0.36	0.36	0.36	0.36	0.36
Laying hen (intensive chicken farm)	0.13	0.13	0.13	0.13	0.13	0.13	0.13
Broilers (intensive chicken farm)	0.16	0.16	0.16	0.16	0.16	0.16	0.16

Table 5. NH_3 emission estimates from different sources in the NCP at prefecture level (kt NH_3-N) in 2004.

Provinces	Prefectures	Cattle	Pigs	Sheep and goats	Poultry	Fertilizer	Total
Beijing	Total	2.2	12.7	3.0	4.1	17.6	39.6
	City	0.0	0.0	0.0	0.0	1.2	1.2
	Changping	0.1	0.6	0.5	0.2	1.0	2.4
	Daxing	0.7	2.2	1.0	0.9	4.3	9.1
	Fangshan	0.2	1.2	0.4	0.6	1.8	4.2
	Mentougou	0.0	0.1	0.1	0.0	0.0	0.3
	Pinggu	0.1	1.3	0.2	0.6	1.9	4.1
	Shunyi	0.7	5.6	0.6	1.4	3.1	11.4
	Tongzhou	0.3	1.8	0.2	0.3	4.3	6.9
Tianjin	Total	4.9	30.0	20.0	7.5	25.2	87.6
Heibei	Total	90.4	180.2	49.5	51.8	335.1	706.8
	Shijiazhuang	20.1	36.1	6.0	12.3	60.9	135.4
	Qinhuangdao	3.1	8.2	2.2	0.9	14.4	28.8
	Tangshan	11.9	26.4	2.8	4.7	39.3	85.0
	Langfan	6.3	10.6	5.5	3.1	19.9	45.4
	Baoding	6.2	32.2	6.5	6.0	51.2	102.1
	Cangzhou	13.2	11.1	6.9	4.6	33.4	69.1
	Hengshui	11.9	14.3	5.1	2.3	27.2	60.8
	Xingtai	10.4	14.9	4.0	7.3	38.7	75.2
	Handan	7.3	26.4	10.4	10.8	50.1	105.0
Henan	Total	62.4	188.2	72.7	28.4	445.5	797.1
	Zhengzhou	2.3	8.6	2.5	1.7	21.3	36.3
	Kaifeng	3.6	14.1	7.4	1.6	24.3	51.0
	Pingdingshan	5.2	12.2	4.2	1.6	26.6	49.8
	Anyan	1.6	7.9	2.7	2.1	30.3	44.6
	Hebi	0.2	3.6	1.7	1.4	7.5	14.5
	Xinxiang	2.6	11.0	3.0	2.4	39.1	58.0
	Jiaozuo	1.9	7.6	1.6	1.7	19.9	32.6
	Puyang	1.3	7.4	3.9	2.0	26.3	40.9
	Xuchang	4.2	13.1	3.8	1.5	18.6	41.2
	Luohe	1.1	9.3	1.0	1.1	13.7	26.2
	Shangqiu	8.2	20.9	16.2	2.8	44.3	92.4
	Xinyang	7.7	16.5	3.8	2.9	51.9	82.8
	Zhoukou	10.3	24.9	11.2	2.5	71.6	120.6
	Zhumadian	12.1	31.0	9.7	3.2	50.1	106.1
Shandong	Total	31.1	308.3	59.6	59.6	334.2	792.9
	Jinan	5.5	35.8	4.9	4.9	26.7	77.8
	Zibo	0.9	10.7	1.5	1.5	13.2	27.7
	Zaozhuang	0.4	14.1	2.9	2.9	17.4	37.6
	Dongying	1.1	7.0	2.3	2.3	13.3	26.1
	Weifang	3.0	44.1	2.9	2.9	49.9	102.7
	Jining	2.3	48.4	9.2	9.2	43.8	112.9
	Dezhou	10.7	51.1	6.1	6.1	42.8	116.8
	Liaocheng	5..86	32.2	6.3	6.3	41.5	86.2
	Binzhou	3.5	16.4	2.3	2.3	27.8	52.3
	Heze	3.8	48.6	21.3	21.3	57.8	152.7
Jiangsu	Total	3.9	57.3	6.9	9.0	330.1	407.2
	Xuzhou	0.9	13.8	1.7	2.2	98.5	117.0
	Lianyungang	0.5	6.8	0.8	1.1	48.0	57.1
	Huaian	0.6	9.3	1.1	1.5	49.1	61.5
	Yancheng	1.3	19.3	2.3	3.0	85.9	111.8
	Suqian	0.6	8.3	1.0	1.3	48.6	59.7
Anhui	Total	28.0	57.7	16.2	5.4	132.7	239.9

	Huaibei	0.7	3.5	1.2	0.4	7.7	13.4
	Bozhou	8.4	14.1	3.9	0.9	24.5	51.9
	Suzhou	4.3	13.2	5.7	1.8	32.3	57.3
	Bangbu	3.3	6.0	1.1	0.8	27.2	38.4
	Fuyang	9.6	18.7	4.0	1.1	29.7	63.1
	Huainan	1.7	2.1	0.3	0.4	11.3	15.8
NCP		222.8	834.3	227.8	165.7	1620.4	3071.0
	(Ratio)	7%	27%	7%	5%	54%	100%

Note: Total value for every province only refers to the emissions from prefectures in the NCP, while the emissions from prefectures in the seven provinces but outside the NCP were not calculated. Emissions in Tianjin municipality were not divided into districts for the 100% of area in the NCP and relative smaller area which means a prefecture in other province.

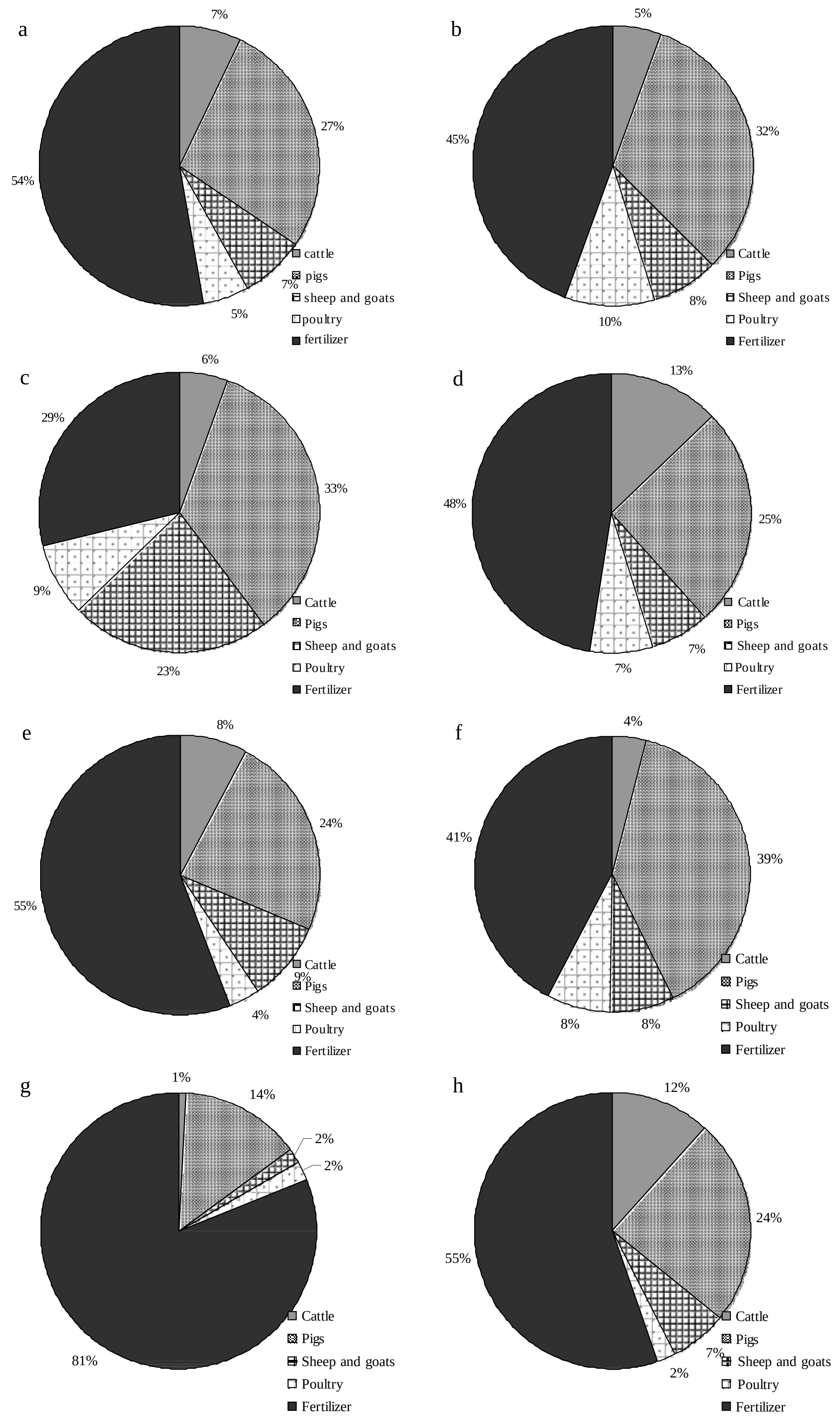

Figure 4. Contributions of NH_3 emission from different sources in the NCP and different provinces (a. NCP; b. Beijing; c. Tianjin; d. Hebei; e. Henan; f. Shandong; g. Jiangsu; h. Anhui)

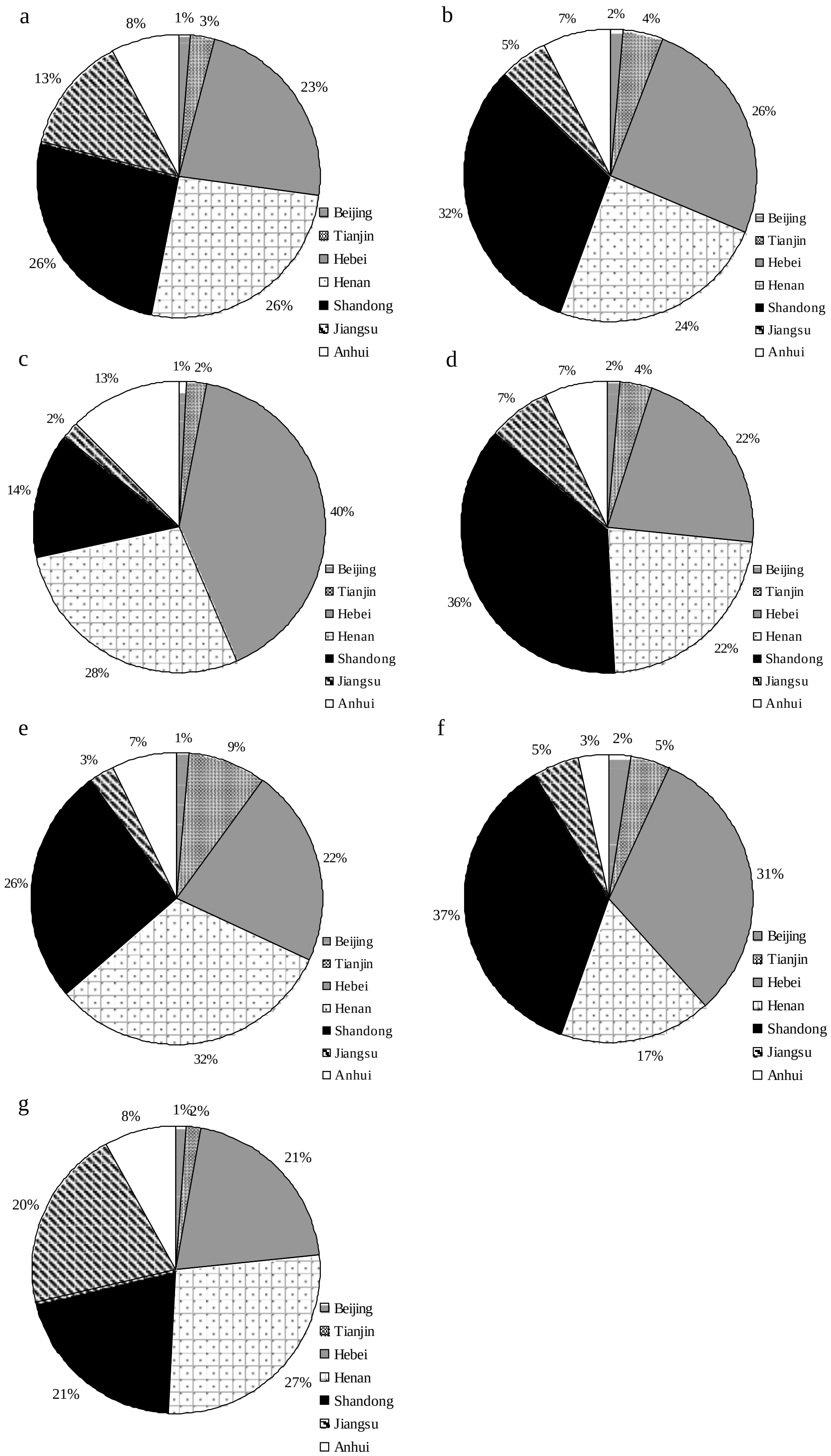

Figure 5. Contributions of NH_3 emission from different provinces for each source in the NCP (a. all the agricultural emissions; b. emission from livestock in total; c. emission from cattle; d. emission from pigs; e. emission from sheep and goats; f. emission from poultry; g. emission from fertilizer use)

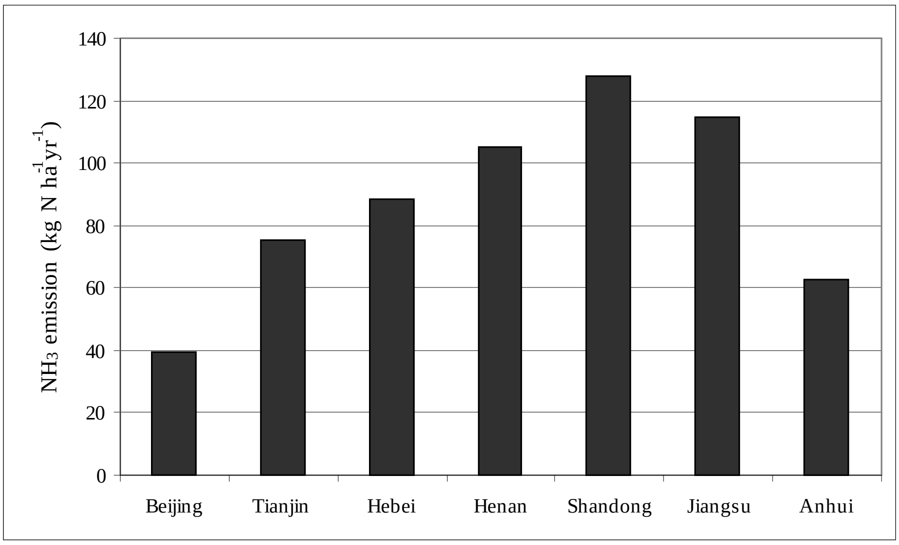

Figure 6. Averaged NH_3 emission densities in different provinces in 2004.

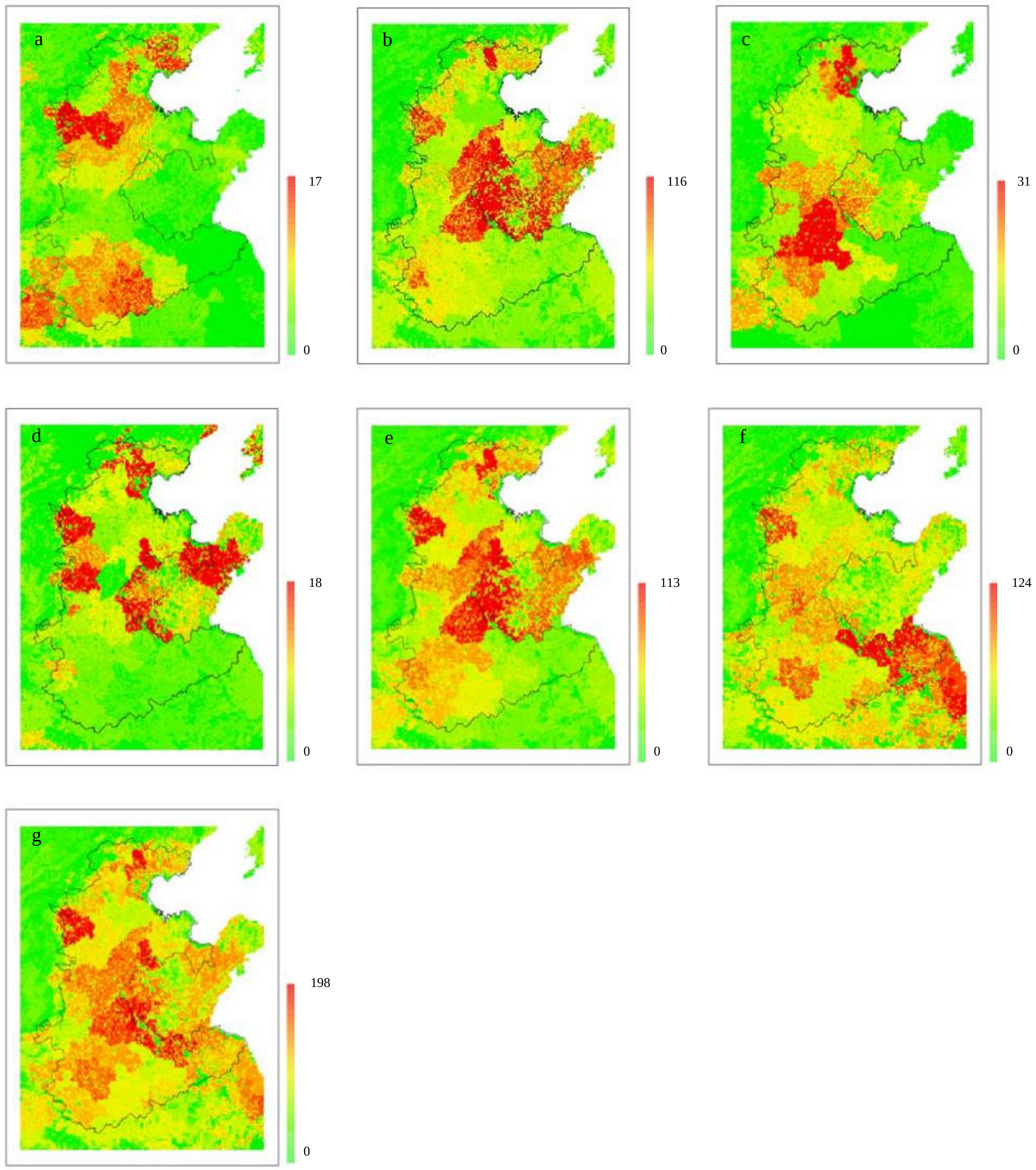

Figure 7. Spatial distribution of the NH_3 emissions from different sources in the NCP (kg NH_3-N ha^{-1} yr^{-1}) in 2004 (a. cattle (including horse, donkey and camels); b. pigs; c. sheep and goats; d. poultry; e. all the livestock sources; f. fertilizer use; g. all the agricultural sources)

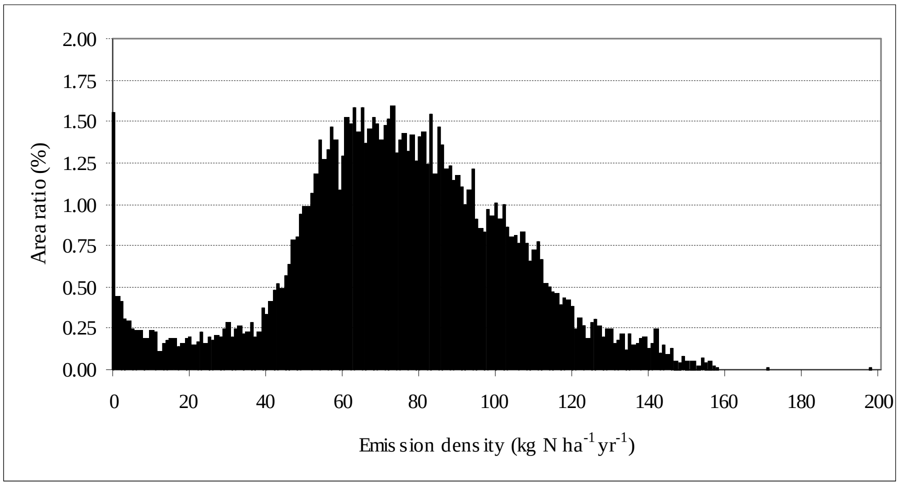

Figure 8. Distribution in terms of area ratio of the total ammonia grid emission density in the NCP in 2004.