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

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8 **Abstract**

9 An agricultural ammonia (NH₃) emission inventory in the North China Plain (NCP) on a prefecture level for the
10 year 2004, and a 5 × 5 km resolution spatial distribution map, have been calculated for the first time. The census
11 database from China's statistics datasets, and emission factors re-calculated by the RAINS model supported total
12 emissions of 3071 kt NH₃-N yr⁻¹ for the NCP, accounting for 27% of the total emissions in China. NH₃ emission
13 from mineral fertilizer application contributed 1620 kt NH₃-N yr⁻¹, 54% of the total emission, while livestock
14 emissions accounted for the remaining 46% of the total emissions, including 7%, 27%, 7% and 5% from cattle,
15 pigs, sheep and goats, and poultry, respectively. A high-resolution spatial NH₃ emissions map was developed
16 based on 1 × 1 km land use database and aggregated to a 5 × 5 km grid resolution. The highest emission density
17 value was up to 198 kg N ha⁻¹ yr⁻¹.

18
19 **Keywords:** Ammonia; Emission inventory; Livestock emission; Mineral fertilizer application; Spatial distribution

20
21 The first high resolution spatial distribution of ammonia emissions for the North China Plain showed rates up to
22 200 kg NH₃-N ha⁻¹ y⁻¹ (Capsule).

24 **1. Introduction**

25 As a natural component of the atmosphere, ammonia (NH₃) plays an important role in atmospheric chemistry and
26 ambient aerosol formation. Its emission rapidly increased during the 20th century due to the doubled or even
27 tripled intensification of agricultural production (Galloway et al., 2004; 2008; Erisman et al., 2008). Ammonia is a
28 major atmospheric pollutant, contributing to eutrophication, acidification and loss of biodiversity (Pearson and
29 Stewart, 1993; Fangmeier et al., 1994; Krupa, 2003). Most emitted reactive nitrogen (N) will be deposited back on
30 land (Goulding et al., 1998), even more so for NH₃ because of its short-distance transport (Asman et al., 1998).

31 This process, called 'N deposition', fertilizes ecosystems, influences the N cycle and introduces N saturation
32 (Aber et al., 1989; Matson et al., 2002; Adams, 2003). Increased N availability in ecosystems can lead to rapid
33 decline in species richness, even at the current N deposition level (Sala et al., 2000; Gotelli and Ellison, 2002;
34 Stevens et al., 2004). In addition, NH₃ is a key precursor to neutralize H₂SO₄ and HNO₃ in the air and form
35 (NH₄)₂SO₄, NH₄HSO₄ and NH₄NO₃ (Pinder and Adams, 2007; Erisman and Schaap, 2004; Walker et al., 2004;
36 Olszjan et al., 2005), which contribute to reduced visibility, regional haze and health impacts associated with fine
37 particular matter (PM).

38
39 Bouwman et al. (1997) estimated that the global NH₃ emission was about 54 Tg in 1990, half of which had been
40 estimated to derive from Asia. In the total global NH₃ emission, 70% is estimated to be related to food production,
41 and the other 30% is estimated to be related to natural sources, industrial processes, fossil fuels, etc. (Bouwman, et
42 al., 1997; Olivier et al., 1998). A host of evidence shows that agricultural sources, i.e. volatilization from livestock
43 manures and mineral fertilizer application, contribute to the major part of NH₃ emissions, approximately 80-90%
44 of the total anthropogenic emission (Bouwman et al., 1997; Asman et al., 1998; Van Der Hoek, 1998; Battye et
45 al., 2003).

46
47 Current researches on ammonia emissions inventories at large scales, such as global, Asian and national
48 inventories, reveal that approximate 20% of global NH₃ emission comes from China (Zhao and Wang, 1994;
49 Kilmont et al., 2001; Yamaji et al., 2004), especially from the intensive agricultural area (Yan et al., 2003; Wang
50 et al., 2005). To feed 22% of the global population on 9% of the world's arable land, China consumed more than
51 30% of the world's total N fertilizer in the last decade (<http://www.stats.gov.cn/>; IFA). Moreover, intensive
52 livestock husbandry has greatly developed in China due to the increasing requirement for livestock products. In
53 2006, China's annual production of meat, eggs and milk were 80.5, 29.5 and 33.0 Mt, respectively, an increase of
54 4.2, 5.5 and 11.4 times those in the early 1980s, respectively (<http://www.stats.gov.cn/>) (Fig. 1a). There was nearly
55 no change for the expenditure on grain and eggs, while the expenditure on meat and milk were 1.3 and 4.8 times
56 that in the previous decade, respectively (Fig. 1b). Meeting this demand was associated with a sharp increase in
57 the use of mineral nitrogen fertilizers and intensive livestock production. This, in turn, has led to the growth of
58 NH₃ emissions in China in the last decade.

59
60 The North China Plain (NCP) (Fig. 2a), which is called "China's granary", provides 40% and 25% of China's

61 wheat and corn production on 3.3% of the national area. Application rates of mineral nitrogen fertilizers in the
62 NCP are up to 600 kg N ha⁻¹ yr⁻¹ (Zhao et al., 1997; Cui, 2005). Less than 30% efficiency in N application
63 introduces about 40% N loss by various routes, including leaching of nitrate (NO₃⁻) and emissions of NH₃, nitrous
64 oxide (N₂O) and molecular nitrogen (N₂). In addition, about 30% of national animal products are also from this
65 area, which further increases the ammonia emission. Significantly high N deposition has been found in this area:
66 27 kg N ha⁻¹ yr⁻¹ deposited in inorganic forms, with 67% in the form of ammonium (Zhang et al., 2008a) , and 9
67 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
68 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
69 using ¹⁵N dilution method. All of this evidence supports the potential intensive NH₃ emission in the NCP.

70
71 In this study, estimated NH₃ emissions in the NCP were calculated at a 5 ×5 km grid resolution based on a
72 Chinese national land use database (1 × 1 km grid resolution). Ammonia emissions from livestock farms were
73 calculated using the agricultural model of RAINS (Klimont and Brink, 2004); emissions from mineral fertilizer
74 application were estimated from earlier measurements. This work represents the first estimate of NH₃ emissions in
75 China at high spatial resolution. In spite of some uncertainties, these data form the essential input for calculations
76 with an atmospheric transport model (ATM) to estimate atmospheric concentrations of NH₃ and deposition of N,
77 which can be applied to estimate exceedances of critical loads and levels and for testing the efficiency of future
78 ammonia emissions abatement strategies.

79

80 **2. Methodology**

81 2.1 Database

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

91 1 km latitude-longitude grid. Five provinces (Hebei, Henan, Shangdong, Jiangsu and Anhui) and two
92 municipalities (Beijing and Tianjin) (Fig. 2a) (Table 1), including fifty-three prefectures (Fig. 2b) were covered in
93 the study area. The average area of the prefectures is 8,452 km², with the minimum value of 2,153 km² for the
94 Hebi prefecture in the Henan province and maximum value of 14,132 km² for the Cangzhou prefecture in the
95 Hebei province (prefectures with only a part of their area in the NCP were not included).

96

97 2.2 Emission factors

98 Although a number of studies have been undertaken on NH₃ emission inventories as well as the emission factors
99 (e.g. Klaassen, 1992, Bouwman et al., 1997, Misselbrook et al., 2000, Klimont, 2001, Doorn et al., 2002),
100 measured emission factors for different sources specific to China do not yet exist. Emission factors for livestock
101 (including poultry) used in European countries are summarized in Table 2. Many factors affect ammonia
102 production, such as species, gender, age, body weight, N content of the feed, the conversion of N in feed to N in
103 meat, milk and eggs, housing systems, litter/manure storage, spreading technique, proportion of time spent by
104 livestock indoor and outside and climatic conditions (EEA, 2007). Bouwman et al. (1997) calculated NH₃
105 emission factors for livestock excretion for developing countries with the assumption of lower feeding levels,
106 lower N content of the feed, a small portion of the livestock housed in stables and a higher temperature in the
107 developing areas. Klimont (2001) adjusted the emission factors for Chinese specific production efficiency of milk
108 or meat. Here, we re-calculated emission factors using the RAINS model methodology (Klimont and Brink, 2004)
109 but considering specific Chinese factors in the calculation:

$$110 \quad ef_1 = N_{x1} * v_1$$

$$111 \quad ef_2 = N_{x1} * (1 - v_1) * v_2$$

$$112 \quad ef_3 = N_{x1} * (1 - v_1 - (1 - v_1) * v_2) * v_3$$

$$113 \quad ef_4 = N_{x4} * v_4$$

$$114 \quad EF = ef_1 + ef_2 + ef_3 + ef_4$$

115 Where, $ef_{1,2,3,4}$ are NH₃-N loss at distinct emission stages, i.e., housing (1), storage (2), spreading (3), and grazing
116 (4); $N_{x1,4}$ are N excretion during housing (1) and grazing (4); $v_{1,2,3,4}$ are NH₃-N volatilization rates at distinct
117 emission stages; EF is the final emission factor. Livestock management specific to the Chinese situation was
118 considered in the study, such as the household scale and industrial scale, housing system, manure storage, as well
119 as the differences in different provinces.

120

121 Taking the emission factor for chicken as an example, all the parameters required in the RAINS model are listed
122 in Table 3. The NH₃-N loss rates at different stages were combinations of the parameters from references (Asman,
123 1992, ECETOC, 1994, Hutchings et al., 2001, EEA, 2007a) and the specific Chinese livestock management
124 systems. In the traditional households, chicken are free-range. NH₃-N loss rates are very high at the housing stage.
125 The main storage method, composting of the manure, increases ammonia emission as well. However, in the
126 intensive chicken farms, laying hens are caged, which gives a low NH₃-N loss rate in the housing stage.
127 Anaerobic digestion is the main storage method for the manure of laying hens in intensive farms, which greatly
128 reduces the ammonia emission. Broilers in intensive farms are kept with bedding material on the floor. For short
129 production cycles, usually 55 days, manure is not treated during the whole period. Bedding material is cleaned
130 once a production cycle finished. So, housing and storage processes are mixed for the broilers in intensive farms.
131 For manure from both the traditional households and intensive farms, 3.5% and 5% NH₃-N are lost in the spring
132 and early autumn spreading stages, respectively. There is no grazing stage for chicken, and the loss rates at this
133 stage were set to zero. Finally, the traditional household management systems introduced much higher emission
134 factors than the intensive farm management systems. In intensive chicken farms, the emission factors for broilers
135 were comparable with the data cited from the reference (Table 2), while the emission factors for laying hens are
136 lower than those for the caged housing stage.

137
138 Emission factors for all the other livestock sources from different provinces in the NCP were calculated by the
139 same methodology and are listed in Table 4.

140
141 Ammonia emission from mineral fertilizer application is another important part of the agricultural sources. In
142 China, the widely used N fertilizers are ammonium bicarbonate (ABC) and urea. Based on EEA (2007b) and
143 ECETOC (1994), the NH₃-N loss rate of urea is 15%. There has been no research on the emission factor for ABC
144 because of its small usage elsewhere. Bouwman et al. (1997) specially developed estimates of NH₃ loss from the
145 application of ABC, which were up to 20-30%. The emission factors of both these fertilizers are higher than for
146 other N fertilizers, e.g. ammonium sulphate, ammonium nitrate, calcium ammonium nitrate, anhydrous ammonia
147 and so on. Cai et al. (2002) measured ammonia loss in the NCP in different cropping systems: 30-39% of N
148 applied to rice, 11-48% of N applied to maize and 1-20% of N applied to wheat was lost as ammonia. Ding (2005)
149 measured ammonia emissions in winter wheat-summer maize rotation systems in Beijing, typical of agricultural
150 areas in the north part of the NCP, and found that 19.4% and 25.8% of N fertilizer applied was lost as ammonia

151 for wheat and maize systems, respectively. Using a wind-tunnel system, Su (2006) found that NH_3 volatilization
152 was up to 21.2% $\text{NH}_3\text{-N}$ of urea-N within fifteen days after fertilization. Yan et al. (2003) estimated the emission
153 from fertilizer application in Asian countries: $\text{NH}_3\text{-N}$ losses of 23.5% and 13.7% were applied for urea in paddy
154 fields and uplands, respectively, while $\text{NH}_3\text{-N}$ losses of 34.5% and 20.5% applied for ABC in paddy fields and
155 uplands, respectively. Instead of re-calculating emission factors, we used measured results directly and without
156 distinguishing differences among provinces for the smaller variation in fertilizer application practice in the NCP.
157 Considering 40% urea, 50% ABC and 10% of compound or other fertilizer applied in the NCP, 22% and 30%
158 were used as the fertilizer emission factors in northern wheat-maize rotation system and southern wheat-rice
159 rotation system in this study, respectively.

161 2.3 Spatial allocation

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

165
166 A methodology similar to the AENEID model for obtaining the geographic distribution of ammonia emissions
167 from livestock and fertilizer application (Dragosits et al., 1998; Dragosits, 1999; Hellsten et al., 2008) was used in
168 this study. The basic theory was to allocate all the livestock and fertilizer to the best suited land cover types. The
169 original NH_3 emission source for each category was distributed onto the appropriate area based on the
170 management type. Dragosits (1999) assigned the practices including housing, storage, spreading, grazing over
171 different land cover types, and then allotted the NH_3 emission from each source by the corresponding calculated
172 weighted indexes.

173
174 Unlike in developed countries, livestock farms in the NCP operate at different scales, from household to
175 industrial, and small farms are randomly scattered all over the place. It is difficult to calculate all the weighted
176 indexes in detail. However, arable land appears in 95% of the total area in the NCP, while forests and grasslands
177 only appear in 8% and 6% of the total area in the NCP, respectively (Fig. 3). There are overlaps of different land
178 uses, such as agro-forestry and agro-pastoral ecosystems, as well as suburban areas with mixed properties of both
179 rural and urban areas. Moreover, except for a tiny number of livestock which are kept part time outdoors by
180 individual farmers, most of the livestock are kept indoors throughout the year in the livestock farms (except sheep

181 and goats which are kept outdoors more time than other livestock). There is no NH₃ emission from agricultural
182 sources considered in the forest and grassland areas excluding the grazing stage of cattle, sheep and goats, because
183 most of the forest and grassland in the NCP are natural instead of semi-natural, as in developed countries. The
184 intensive management practices in the agricultural areas (mainly arable land) greatly simplify the allocation
185 process although some details are still missing. Ammonia emissions from housing and storage, spreading and
186 grazing were allocated to probable rural residual areas, arable areas and grassland, respectively. The weighted
187 index for each grid square was calculated by the percentage of the corresponding land uses. As for the mineral
188 fertilizer application, NH₃ emission was allocated to the arable areas. The N fertilizer use is never homogenous
189 because of the family operation on small plots, so an average value was applied. Although application of the same
190 emission factors for mineral fertilizer use will introduce larger uncertainties, it is still the most advisable way for
191 regional research at present. All emissions, independent of the management practices, were estimated within a
192 prefecture region and mapped onto a 1-km grid resolution by the land use at first. Considering the areas of the
193 prefectures in this study, 8,452 km² on average, and reduction of uncertainties, the resulting map was aggregated
194 to a 5-km resolution.

195 **3. Results and Discussion**

196 **3.1 NH₃ emission inventories**

197 NH₃ emissions from different sources in every prefecture in the NCP are listed in Table 5, based on the census
198 database and emission factors. The total NH₃ emission was as high as 3071 kt NH₃-N yr⁻¹ in the NCP in 2004,
199 including 1620 kt NH₃-N yr⁻¹ from mineral fertilizer application and 1451 kt NH₃-N yr⁻¹ from livestock sources.
200 In all the livestock sources, 834 kt NH₃-N yr⁻¹ was from pig emission, taking the largest part; the following 223 kt
201 NH₃-N yr⁻¹ and 228 kt NH₃-N yr⁻¹ were from cattle and sheep (goats), respectively; and 166 kt NH₃-N yr⁻¹ was
202 from poultry.
203

204
205 Zhao and Wang (1994), Olivier et al. (1998) and Klimont (2001) estimated NH₃-N emissions in China
206 corresponding to 11.1, 8.4 and 7.9 Tg in 1990. Klimont (2001) estimated NH₃-N emission in China to be 9.7 Tg in
207 1995. Streets et al. (2003) estimated NH₃-N emission in China to be 11.2 Tg in 2000. Regardless of the temporal
208 variation and other minor emission sources in the NCP, the agricultural sources in the NCP contributed 27% of
209 the recent NH₃ emission in China in 2000s. Comparing with the result from Hellsten et al., (2008), the total

210 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.
211 It means that the ammonia emission density based on grid cell in the NCP is more than 10 times that in the UK.
212
213 Sun et al. (1997) estimated NH₃ emission from the five provinces: Hebei, Henan, Shandong, Jiangsu and Anhui,
214 and found 3.0 Tg NH₃-N emitted in 1995. Wang et al. (2003) made the same estimation, and found 4.9 Tg NH₃-N
215 emitted in 2000. Taking the area ratios (Hebei: 43%; Henan: 46%; Shandong: 40%; Jiangsu: 35%; Anhui: 27%) of
216 the five provinces inside the NCP into account, it would give approximately 1.2 and 1.9 Tg NH₃-N emissions for
217 the areas in the NCP based on these two studies above, while our results gave 2.9 Tg NH₃-N from the same area,
218 much higher than the former estimates. However, we cannot simply calculate emissions from the product of the
219 emission and the area ratio of every province, because it was assumed that the emissions were homogeneously
220 distributed in every province during the calculation. Actually, intensive agriculture is concentrated in the NCP
221 instead of all the seven provinces (including the two municipalities, Beijing and Tianjin). The areas in these
222 provinces but outside the NCP were usually forests and grasslands with less ammonia emissions, which can be
223 seen in the land use category maps in Fig. 3 and the subsequent spatial distribution section.

224

225 3.2 Contributions of NH₃ emission from different sources and provinces

226 Contributions of NH₃ emissions from different sources in the NCP and different provinces are shown in Fig. 4.
227 Mineral fertilizer use contributed 54% to the total NH₃ emission in the NCP, while livestock sources contributed
228 the remaining 46% (Fig. 4a), consistent with the results for the whole national scale in former studies (Zhao and
229 Wang, 1994; Kilmont 2001; Streets, et al., 2003). In the different provinces, ammonia emissions from mineral
230 fertilizer application accounted for 29%-81% to the total emissions (Fig. 4b-h), with lower ratio in the Tianjin
231 municipality and higher ratio in the Jiangsu province. Livestock emissions constituted emissions from cattle (7%),
232 pigs (27%), sheep and goats (7%) and poultry (5%) (Fig. 4a). Pig emissions constituted the largest proportion of
233 the livestock emissions, which is related to people's dietary habits. In the different provinces, the ratios ranged
234 from 14%-39% to the total emissions, with lower ratio in the Jiangsu province and higher ratio in the Shandong
235 province. Contributions of emissions from cattle, sheep and goats, and chicken varied in different provinces,
236 related to the different development strategies. Emissions from cattle now make a more significant contribution
237 due to the sharply increased milk consumption in recent years (Fig. 1), although no temporal comparison was
238 carried out here.

239

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

249
250 Contributions of the total livestock emission from different provinces were proportioned to the area percentages of
251 each province to the NCP (Fig. 5b; Table 1): the Hebei, Henan and Shandong provinces took the larger part,
252 followed by the Jiangsu and Anhui provinces, then the Beijing and Tianjin municipalities. Among all livestock
253 sources, $223 \text{ kt yr}^{-1} \text{ NH}_3\text{-N}$ was from cattle emission. All cattle, buffalos, dairy cattle, horses, donkeys, and camels
254 were included in this part for the former category of draught cattle. Actually there are only a small number of
255 horses, donkeys and camels in the NCP nowadays. 67% of the emission came from dairy cattle because of the
256 rapidly increased consumption of milk (Fig 1b). The Hebei province provided the largest part, 40% of the cattle
257 emission. Unlike the fraction of the total emission, there was only 2% cattle emission from the Jiangsu province
258 (Fig. 5c). Ammonia emission from pigs account for 27% of the total emission, more than half of all the livestock
259 emissions. The Shandong province provided the largest part of pig and poultry emission, 36% and 37% of the two
260 parts, respectively (Fig. 5 d & f). For the emission from sheep and goats, the three main provinces, Hebei, Henan
261 and Shandong still took the leading parts (Fig. 5g). The Tianjin municipality shared 9% and the Jiangsu province
262 shared only 3% of the emission, which was asymmetrical to the area percentages.

263
264 About $1620 \text{ kt yr}^{-1} \text{ NH}_3\text{-N}$ was emitted from mineral fertilizer application to arable land based on the census data
265 of N-fertilizer consumption and the emission factors. Emission from mineral fertilizer application constituted the
266 largest proportion of the overall emission, contrasting with the ratios in developed countries which have more than
267 80% of emission from livestock husbandry (Sutton et al., 1995; Dragosits et al., 1998; Olivier et al., 1998; Battye
268 et al., 2003). This illustrated the overall very high level of fertilizer application commonly used in Chinese
269 agriculture. Mineral fertilizer application in the Jiangsu province contributed 20% ammonia emission in this area

270 (Fig. 5h), twice as much as the other provinces considering the percentage of the area of the NCP. On the one
271 hand, paddy rice (or wheat-rice rotation system) is the main crop system in the southern the NCP, while winter
272 wheat-summer maize is the main crop rotation system in the north; on the other hand, the temperature in the south
273 is higher than in the north, which was a key factor for the NH_3 emission from mineral fertilizer application (EEA,
274 2007).

275 276 3.3 Spatial distribution

277 According to the estimation of NH_3 emissions from each source (Table 4), the emission from every prefecture was
278 distributed over land use by the bottom-up process (Fig. 7). Variations of livestock emissions were significant (Fig
279 6a-d) for the regional priority development strategies. High cattle emission was found in the Hebei and the Henan
280 provinces, especially the Shijiazhuang and Hengshui prefectures in the Hebei province (Fig. 7a). High pig
281 emission was found in the Shandong province, especially the west part which was inside the NCP (Fig 6b).
282 Although the total contribution of sheep and goats emission was equal to the total contribution of cattle emission,
283 the sheep and goats emission was more concentrated in the mid-south of the NCP, with a maximum of up to 31 kg
284 $\text{N ha}^{-1} \text{ yr}^{-1}$ (Fig. 7c). Poultry emissions were up to 18 kg $\text{N ha}^{-1} \text{ yr}^{-1}$, and mainly concentrated in the mid-north of the
285 NCP (Fig. 7d). The highest livestock emission was 113 kg $\text{N ha}^{-1} \text{ yr}^{-1}$ at a 5×5 km grid resolution, with high
286 values in the north of the Tianjin municipality, in the south of the Hebei province, in the west of the Shandong
287 province and the north of the Henan province (Fig. 7e).

288
289 The NH_3 emission from mineral fertilizer use was high, up to 124 kg $\text{N ha}^{-1} \text{ yr}^{-1}$, which meant a great loss of N
290 resources to the environment. High emissions appeared in the Shandong province, in the Jiangsu province and in
291 some parts of the Hebei and Henan provinces (Fig. 7f), consistent with the over-use of N fertilizer in these
292 intensive agricultural areas (Zhao et al., 1997; Cui, 2005). In contrast with the lower NH_3 emissions from
293 livestock husbandry in the Jiangsu province, it turned out to be higher in fertilizer use emission. The large amount
294 of N input, higher temperature and more precipitation in this area were all considered to be the main driving
295 factors.

296
297 The total ammonia emission was as high as 198 kg $\text{N ha}^{-1} \text{ yr}^{-1}$ (Fig. 7g). High emissions were found in the middle
298 of the NCP. Low emissions were found in the north and west of the Hebei province and most of the Anhui
299 province, as well as individual emission sources in these two regions. It also gave a realistic explanation of the

300 much higher NH₃ emission in the five main provinces in the NCP than the values simply calculated from the area
301 ratio in section 3.1 above.

302

303 Area ratios of the 5-km grid ammonia emission density for the NCP in 2004 are shown in Fig. 8. Most areas of the
304 NCP had an ammonia emission between 40-120 kg N ha⁻¹ yr⁻¹. Hellsten et al. (2008) modelled the ammonia
305 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%
306 of the area as ‘hotspots’, which was introduced by pig/poultry farming. Comparing with that result in the UK,
307 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
308 nearly the whole regional area for intensive agricultural practices. Such a high NH₃ emission rate plays a major
309 role in secondary particulate formation of (NH₄)₂SO₄, NH₄HSO₄ and NH₄NO₃ in this area (He et al, 2001; Yao, et
310 al., 2003; Wang et al., 2005; Duan et al., 2006). Furthermore, most of the emitted ammonia will be deposited back
311 to the land, and introduce ecological changes (Abler et al., 1989; Matson et al., 2002). Cape et al. (2009)
312 introduced a new annual critical level of NH₃ for higher vegetation of 2-4 μg m⁻³; sensitive plant species, such as
313 lichens and bryophytes, were even more sensitive, with a critical level of 1 μg m⁻³. As the preliminary output
314 results of the N deposition in the NCP (unpublished), which was modelled by the ATM model with the input of
315 the spatial distribution ammonia emission in this study, exceedance of the critical level for NH₃ appeared over
316 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
317 deposition across British acid grasslands. Although higher tolerance and less sensitivity of the crop system in the
318 intensive agricultural area in the NCP resulted in ecological effects which have been ignored for a long time, the
319 high NH₃ emissions will definitely bring in ecological changes even though there is little research to offer any
320 evidence to date.

321

322 3.4 Uncertainties

323 Uncertainties of NH₃-N emissions in the NCP are associated with both the emissions inventory and the spatial
324 allocation. The quality of the emissions inventory is further related to the quality of the input data and the
325 emission factors. The spatial allocation is affected by land use maps and the data for agricultural practices, which
326 are used to calculate the weight indexes for the different land use classes. Considerable uncertainty is associated
327 with the emission factors and the agricultural practices. The input census data were from the China Statistic
328 Bureau and local statistical bureaus, and the geographic maps were from the Institute of Geographic Sciences and

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

331

332 It was difficult to quantify the uncertainties in the emission factors, because there are few NH₃ emission
333 measurements available in China. In this study, the emission factors were re-calculated by the RAINS model,
334 taking the husbandry practices in the NCP into consideration, which was considered more consistent with reality
335 than simply using emission factors from elsewhere. Some of the emission factors were quite different compared
336 with previous studies, e.g. about twice those of the former sheep and goat factors. The main reason for this is the
337 lower ratio of lambs, which are associated with a smaller emission. In contrast to European countries, Chinese
338 people consume more mutton than lamb. In addition, instead of mostly indoor management like other livestock,
339 sheep and goats usually graze more outdoors, which also increased the NH₃ emission. The emission factors of
340 chicken in the intensive farms were also at the low level comparing with former studies in Table 2. The
341 explanation had been given in the example of the calculation of the emission factor. Mineral fertilizer application
342 emission factors were cited from measured results directly, without making any distinction among the provinces,
343 as was done for livestock emissions for the whole homogenous land use and agricultural practice.

344

345 The spatial allocation processes introduce another area of uncertainty. Theoretically, the NH₃ emission from each
346 prefecture would be bottom-up according to the corresponding area based on the land use maps. The agricultural
347 practices determined the weighted indexes of different land cover categories. It was effective for the fertilizer use,
348 but more difficult for livestock husbandry, because more processes (housing, storage, spreading and grazing) and
349 the special traditional household managements in the NCP have to be considered for the latter. However, the
350 homogenous farmland land cover and the mostly indoor livestock keeping in the NCP largely simplified the
351 process. The major uncertainties were not from the theoretical process, but rather associated with the local
352 situation. First of all, the operation of livestock husbandry ranges from household scale to industrial scale and is
353 randomly scattered in the rural area. Second, organic manure use has dramatically declined and been largely
354 replaced by mineral fertilizer use. Only 18% of N was from organic manure in 47% of arable land which still
355 received organic nutrient in China (Ju et al., 2005). A lot of animal manure was neither treated nor recycled,
356 particularly the liquid manure, and directly discharged into surface waters, which introduced further
357 environmental problems. Third, the organic manure was not necessarily applied in the local area. It is mostly

358 transported to other areas after production. In this study, all these three possibilities were not taken into
359 consideration, and instead the emissions were assigned onto the local area.

360
361 In addition, all the census data were at prefecture level, and distributed into each prefecture. So, there were
362 artificial boundaries between higher and lower emission areas in the spatial variation maps, which were not
363 physically realistic. However, it is true that there were different priorities in the development strategy in different
364 regions which introduced variations in both livestock husbandry and fertilizer use in crop farms even on a
365 prefecture level. Although we can eliminate this if we use bottom-up emissions on a province level or the NCP
366 regional level, it will actually obliterate more spatial realities at a higher resolution.

367 368 **4. Conclusions**

369 An inventory of NH₃ emissions at prefecture level in the NCP was carried out for the year 2004. The total
370 agricultural NH₃-N emission was high (3071 kt yr⁻¹), accounting for 27% of the total emission in China, while the
371 area ratio was only 3.3%. 1620 kt yr⁻¹ NH₃-N emission derived from fertilizer application, which was the largest
372 emissions source, constituting more than half of the total agricultural emission. Livestock emissions contributed
373 1451 kt N yr⁻¹, constituting: emissions from cattle (7%), pigs (27%), sheep and goats (7%) and poultry (5%). The
374 Henan, Hebei and Shandong provinces made the largest contribution to the total emissions in the NCP.

375
376 A high-resolution map of NH₃ emissions in the NCP was developed for the first time based on census data and
377 land use maps. The highest 5-km grid emission density was 198 kg N ha⁻¹ yr⁻¹. High emissions were found in the
378 south of the Hebei province, in the west of Shandong province and in most of the Henan province. Generation of
379 this spatial distribution map of ammonia emissions will allow the opportunity to undertake atmospheric transport
380 modelling to provide spatial estimates of wet and dry deposition of N in the NCP. Although there are still some
381 uncertainties introduced by the lack of original emission factors in China, the emissions inventory and spatial
382 distribution of NH₃ emissions provide indispensable input data for atmospheric transport, N deposition, critical
383 load exceedance models and abatement strategies for China in future research.

384 385 **Acknowledgements**

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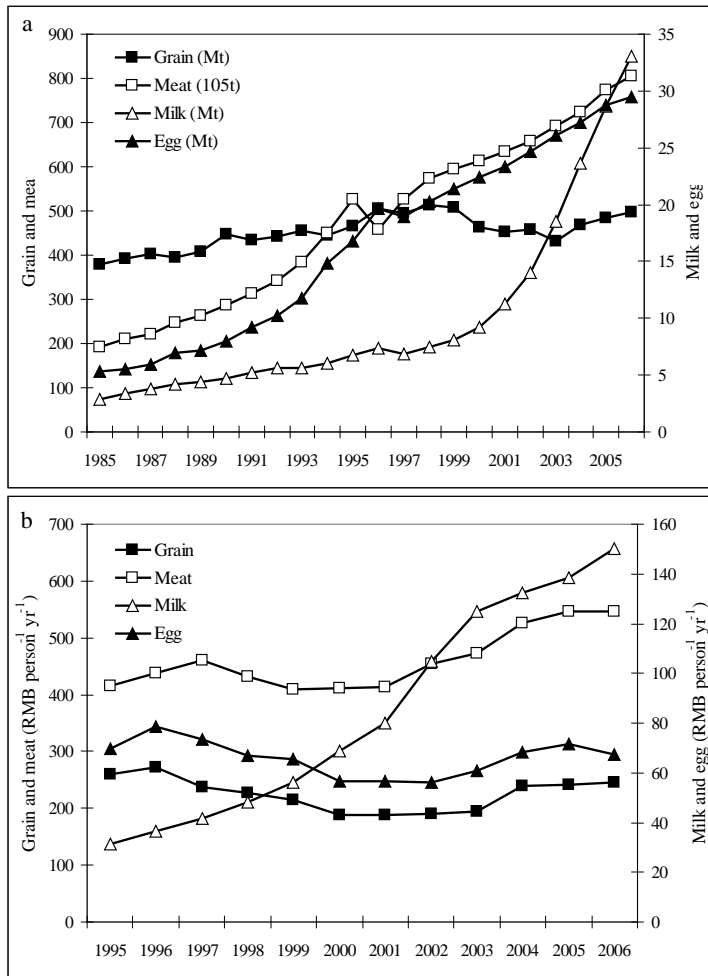


Figure 1. Agricultural products consumption in China in the last two decades (a) and the average human living expenditures in the last decade (b).

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

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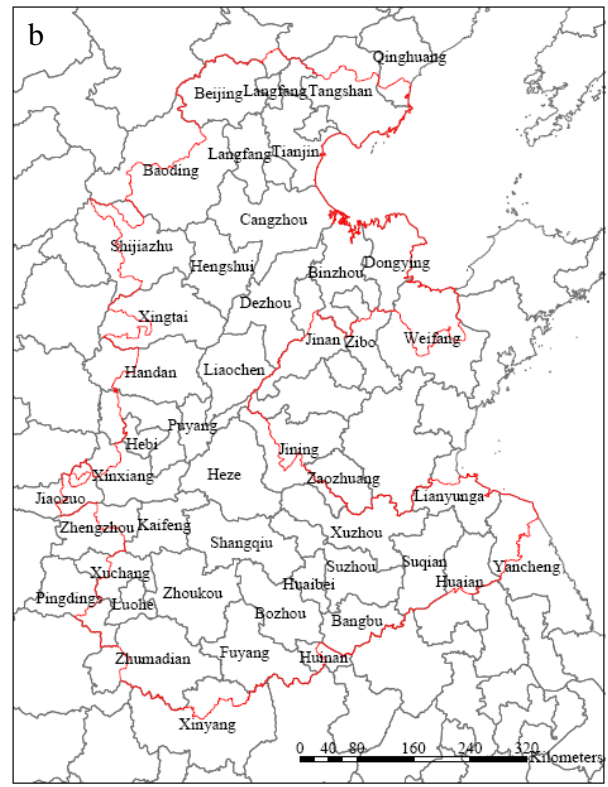
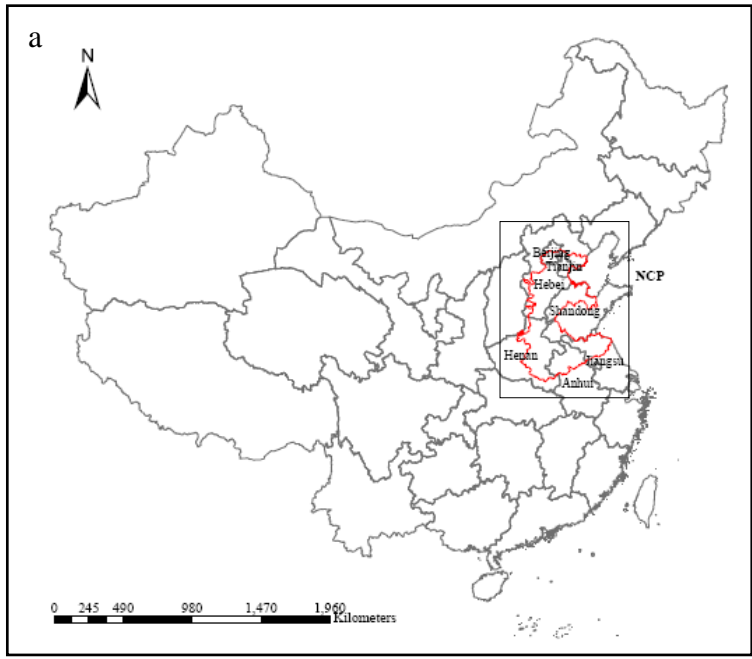


Figure 2. Location of the North China Plain (NCP) on a province scale (a) and prefecture scale (b) (Database for Beijing and Tianjin are on district level but not displayed here; red line is the boundary of the NCP)

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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

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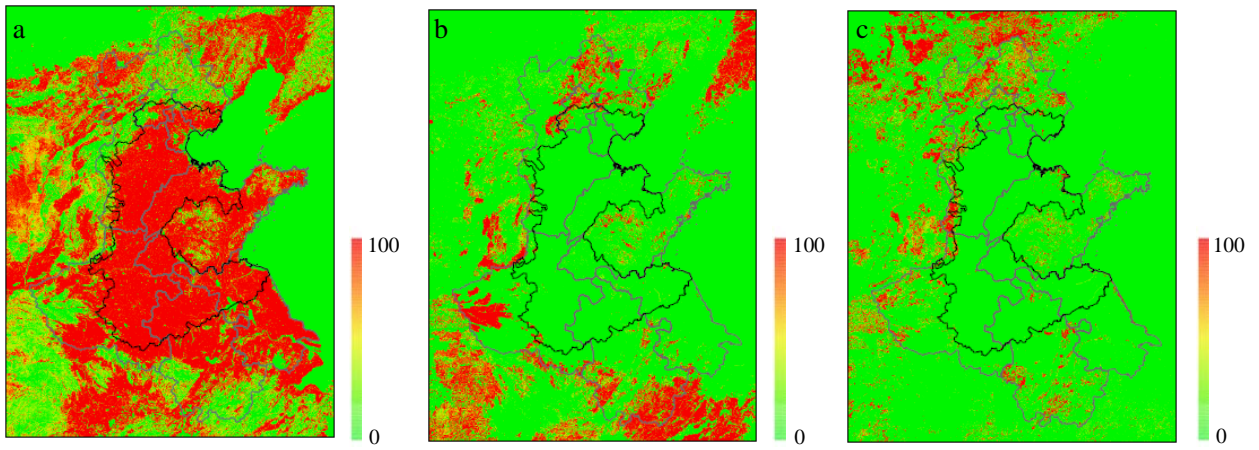


Figure 3. Land cover in the NCP (a. arable land; b. forest; c. grassland) (grey lines are the boundaries of the provinces involved in; black line is the boundary of the NCP)

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Table 2. NH₃ emission factors (kg N head⁻¹ yr⁻¹) from different references.

Category	Europe	UK	Europe	Developed countries	Developing countries	China
	Van der Hoek, 1998	Misselbrook et al., 2000	Klaassen, 1992	Bouwman et al., 1997		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

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

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Table 3. NH₃-N loss rates at different stages and the calculated emission factor in chicken farms in the NCP.

		Type of housing	N _{x1}	v ₁	v ₂	v ₃	v ₄	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_{x1}- kg N head⁻¹ yr⁻¹; EF- kg N hear⁻¹ yr⁻¹

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Table 4. NH₃ 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

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Table 5. NH₃ emission estimates from different sources in the NCP at prefecture level (kt NH₃-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
Hebei	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
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
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.

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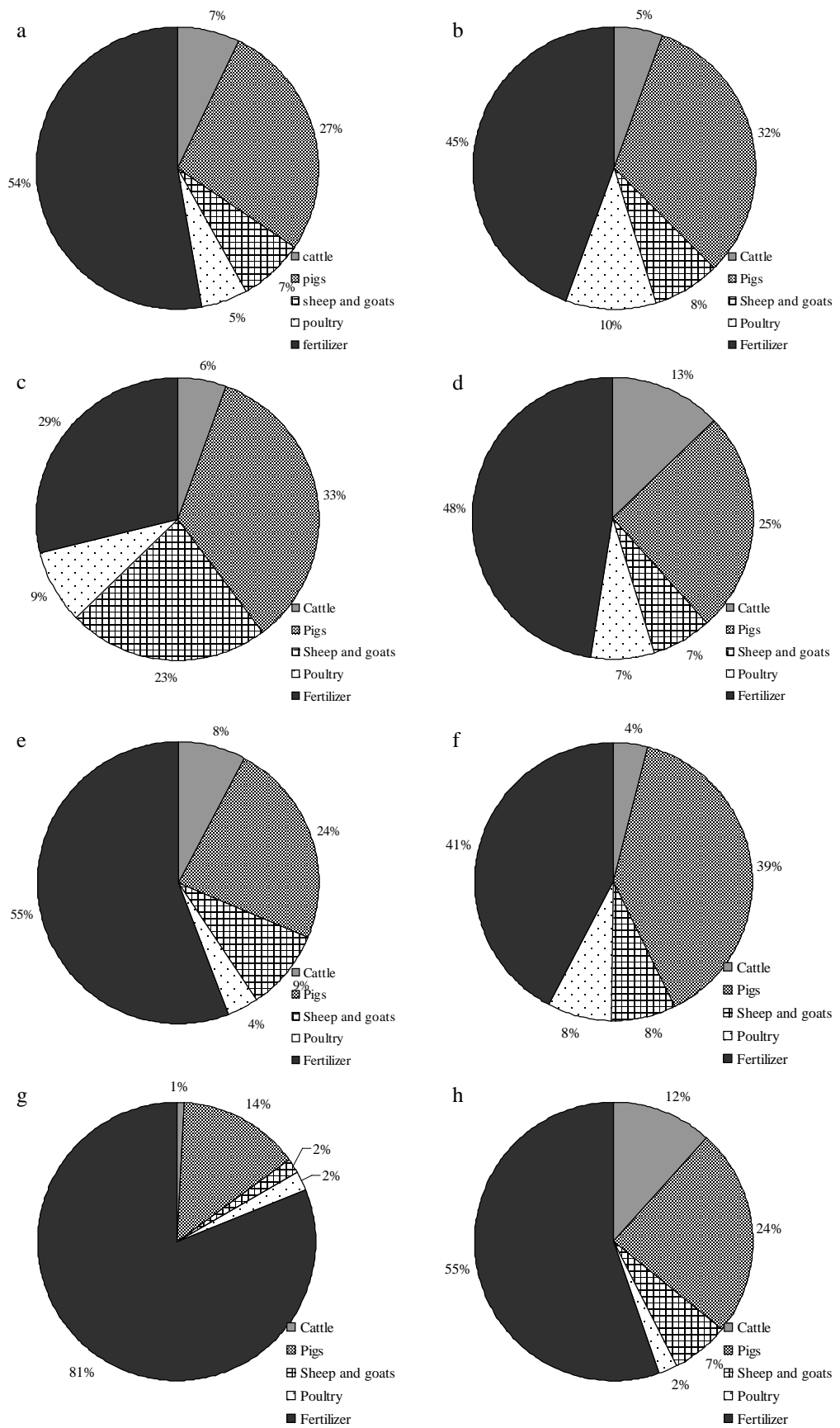


Figure 4. Contributions of NH₃ emission from different sources in the NCP and different provinces (a. NCP; b. Beijing; c. Tianjin; d. Hebei; e. Henan; f. Shandong; g. Jiangsu; h. Anhui)

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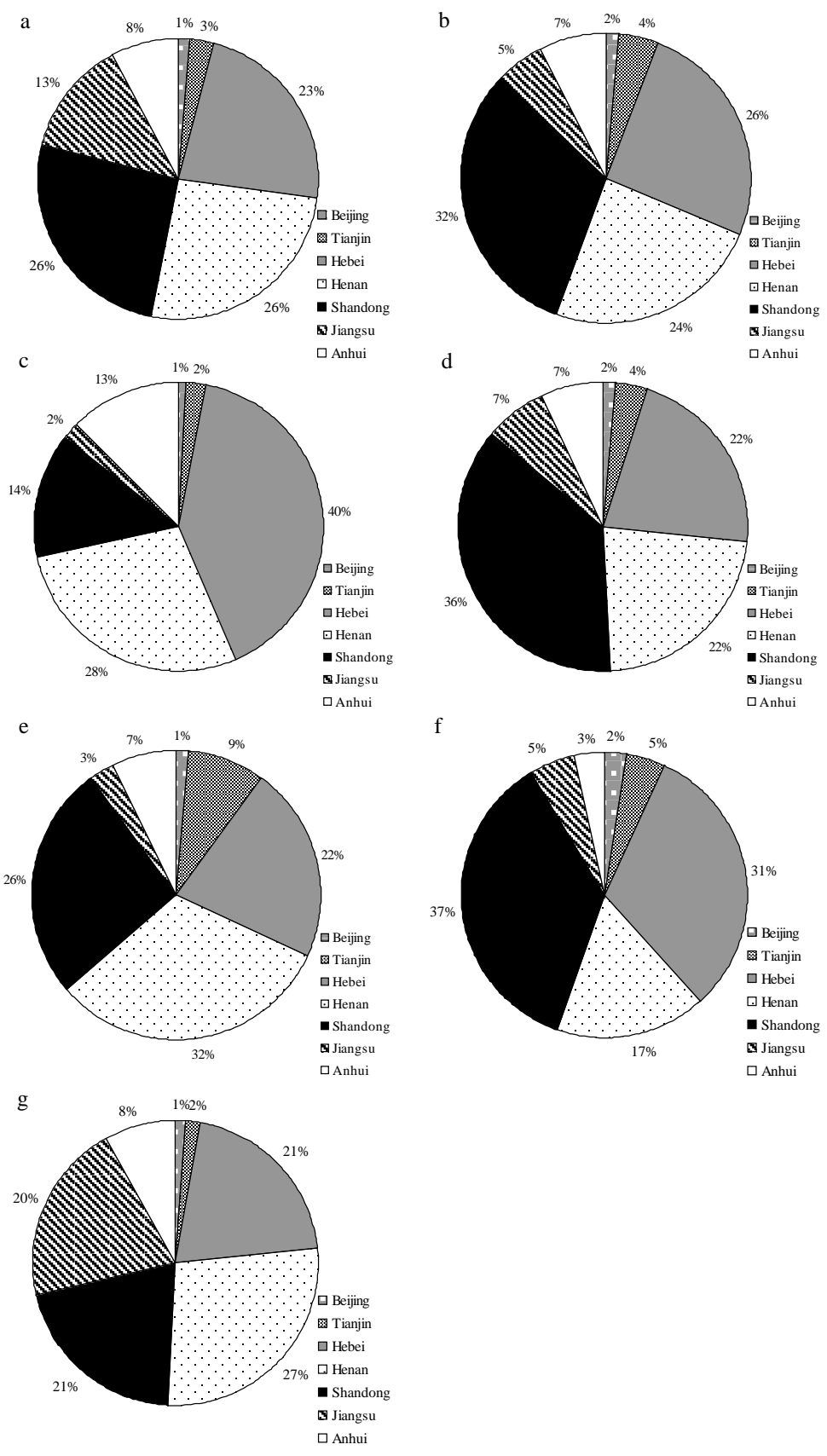


Figure 5. Contributions of NH₃ emission from different provinces for each source in the NCP (a. all the agricultural emissions; b. emission from livestock in total; c. emission from cattle; d. emission from pigs; e. emission from sheep and goats; f. emission from poultry; g. emission from fertilizer use)

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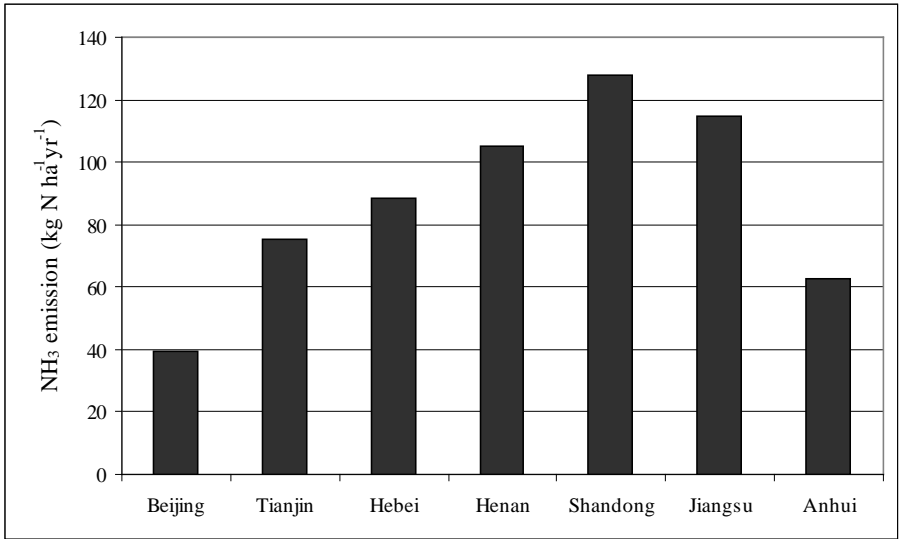
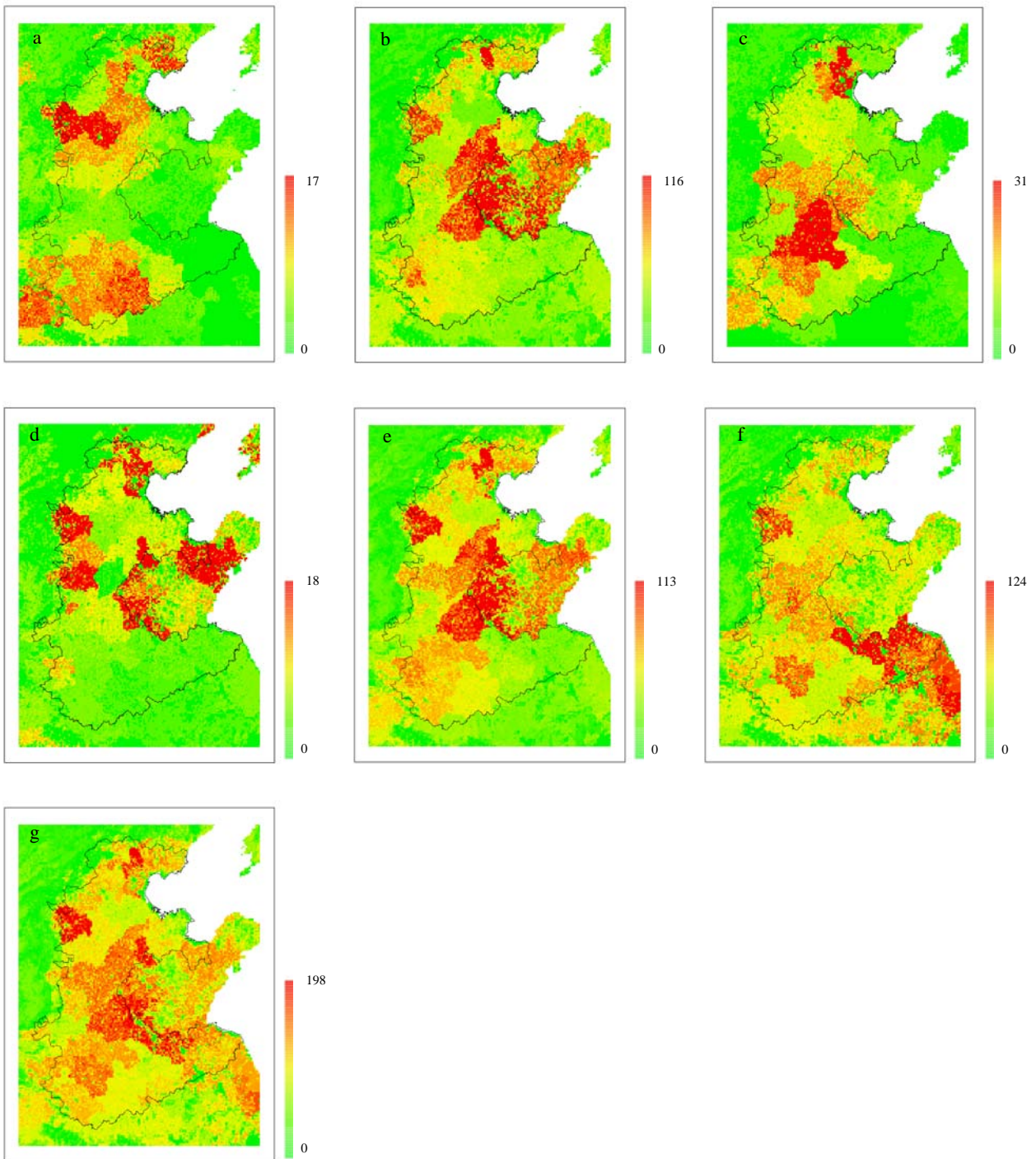


Figure 6. Averaged NH₃ emission densities in different provinces in 2004.



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Figure 7. Spatial distribution of the NH_3 emissions from different sources in the NCP ($\text{kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$) 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)

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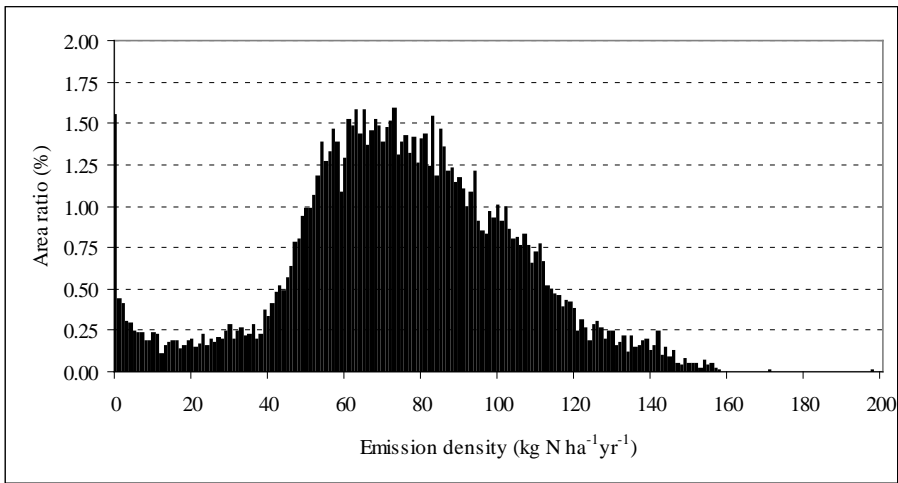


Figure 8. Distribution in terms of area ratio of the total ammonia grid emission density in the NCP in 2004.