

Original Research

Spatio-Temporal Emissions of Ammonia and Greenhouse Gases from Livestock and Its Relation to Atmospheric Particulate Matter Pollution in the Beijing-Tianjin-Hebei Region

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Abstract

Livestock farming sector is an important anthropogenic emission source of ammonia (NH₃) and greenhouse gases including nitrous oxide (N₂O) and methane (CH₄). The NH₃, not N₂O and CH₄, is known as an important gaseous precursor to cause atmospheric particulate matter (PM) pollution. However, the relationship between livestock-derived NH₃ and atmospheric PM pollution has not been sufficiently investigated, especially in the developed regions with serious air pollution. Here, we studied the spatio-temporal emissions of NH₃, N₂O and CH₄ from livestock farming as well as the relationship between livestock NH₃ emission and atmospheric PM pollution in the Beijing-Tianjin-Hebei Region, one of the most developed and air polluted area in China. Over the past 40 years, livestock-derived emissions of NH₃, N₂O and CH₄ had experienced four temporal stages (1978-1990: low level; 1991-1995: rapid growth; 1996-2005: reached hot moments; 2006-2018: stable at a high level). Livestock-derived emissions of NH₃, N₂O and CH₄ were 813, 19 and 499 Gg in 2017, respectively. The southeastern plain was the hotspot, and the pig and cattle were the main sources (78-99%) of NH₃, N₂O and CH₄ in livestock farming. The livestock-derived NH₃ emission explained approximately a quarter of the variations of atmospheric PM_{2.5} (size ≤2.5 μm) (24%) and PM₁₀ (size ≤10 μm) (22%) pollution. Based on the knowledge of atmospheric chemical processes, we concluded that the livestock-derived NH₃ emission significantly affected atmospheric PM_{2.5} and PM₁₀ pollution in the Beijing-Tianjin-Hebei Region. Consequently, the livestock farming, especially pig and cattle breeding, should be paid more attention

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in the context of atmospheric particulate matter pollution and regional greenhouse gases management.

Keywords: ammonia (NH₃), greenhouse gases (GHGs), methane (CH₄), nitrous oxide (N₂O), PM_{2.5}

Introduction

Livestock farming sector is one of the main anthropogenic emission sources of ammonia (NH₃) and greenhouse gases (GHGs) including nitrous oxide (N₂O) and methane (CH₄) [1-4]. Among the three gases, NH₃, as an important gaseous precursor of inorganic aerosol, plays an important role in atmospheric particulate matter formation [5-7]. For example, NH₃ can react with sulfur dioxide (SO₂), nitrogen oxides (NO_x), etc. to produce secondary particles and aerosols such as ammonium sulfate ((NH₄)₂SO₄), ammonium sulfite (H₈N₂O₃S) and ammonium nitrate (NH₄NO₃) [8-9]. In contrast, N₂O and CH₄ persist in the atmosphere for a relatively long time [10] and have little role in the formation of surface atmospheric particulate matter [6, 11]. Although N₂O can produce NO₂ (an important air particulate pollutant), this reaction mainly occurs in the stratosphere [10]. Therefore, it is of importance to understand the spatio-temporal emissions of livestock-derived NH₃, N₂O and CH₄ as well as the relationships between NH₃ and atmospheric particulate matter pollution in the context of atmospheric pollution and global climate change [12-14].

At a global scale, livestock farming as well as its-induced land use changes contributed 64%, 65% and 37% of anthropogenic NH₃, N₂O and CH₄ emissions, respectively [15]. The hotspots were in Asia, especially in India and China over recent decades [16-17]. The emissions of NH₃ and GHGs from China's animal husbandry showed an upward trend, with an average annual growth rate of 2.2% [18-19]. Even though there are some studies on livestock-derived NH₃ and GHGs emissions in several regions in China [20-22], the spatio-temporal patterns of the very developed and air polluted areas has not been sufficiently studied. Moreover, the impact of livestock-derived NH₃ emission on regional atmospheric particulate matter pollution is a hot research topic [23]. The current researches mainly focus on the three aspects of small region [24], facility farming [25-27] and atmospheric particulate matter formation mechanism [12]. On a regional scale, however, the spatio-temporal emissions of NH₃, N₂O and CH₄ from livestock farming as well as the relationships between NH₃ and atmospheric particulate matter pollution have not been sufficiently investigated.

The Beijing-Tianjin-Hebei region is one very developed and air polluted (especially particulate matter) areas in China [28]. In the global context of low-carbon economy, the Beijing-Tianjin-Hebei region is facing the double pressure of rapid economic & social development and reduction in NH₃ and GHGs emissions. The Beijing-Tianjin-Hebei region has intense livestock

farming, however, how it affects spatio-temporal emissions of NH₃, N₂O and CH₄ and the surface atmospheric particulate matter pollution is unclear. The objective of this study was to (i) investigate the spatio-temporal emissions of NH₃ and GHGs from livestock farming as well as its dominant sources, and (ii) explore the correlation between livestock-derived NH₃ emission and surface atmospheric particulate matter pollution in the Beijing-Tianjin-Hebei region.

Materials and Methods

Study Site

This study was conducted in the Beijing-Tianjin-Hebei (B-T-H) region (113°27'-119°50'E, 36°05'-42°40'N) in China, with a total area of 218,000 km². The B-T-H region gradually transitions from the northwest Yanshan-Taihang Mountain to the Southeast Plain. The mountain and plain accounted for 53.7% and 46.3% of the whole B-T-H region, respectively. The northwest mountain and southeast plain were characterized by a temperate continental climate and a warm temperate monsoon climate, respectively. There are four distinct seasons in this area, with the same period of rainfall and heat. The annual precipitation is approximately 300-800 mm, and the mean annual temperatures are 0-13°C in the B-T-H region. The regional soil types are mainly Burozem, Cinnamon Soil, Calcaric Cambisol and Chestnut Soil [3].

The B-T-H region has two megalopolis (Beijing and Tianjin Municipalities) and 11 prefecture level cities of the Hebei Province, which is one developed area in China. By the end of 2018, the B-T-H had a permanent population of 113 million, with a population density of 517 people km⁻². The total gross domestic product of the B-T-H region was 8.5 trillion RMB, and the number of motor vehicles was approximately 24.6 million. The area of cropland was the largest, accounting for 34.8% of the whole area of B-T-H region. The forest land, grassland, construction land, water area and unused land accounted for 22.3%, 14.0%, 12.1%, 2.9% and 0.69% of the total area, respectively.

Data Collection of Livestock Farming

The livestock farming of the B-T-H region mainly includes five types: dairy cattle, meat cattle, pigs, broilers and laying hen. The long-term (1978-2018) data of the livestock farming were collected based on the official statistical yearbook of Beijing Statistics Yearbook 2003-2019 (data of 1978-2018), District

Statistical Yearbook of Beijing 2008-2019 (data of 2007-2018), Tianjin Statistical Yearbook 2002-2018 (data of 2000-2018), Hebei Economic Yearbook 2008-2018 (data of 1978-2017, the data of prefecture level cities were only during 2008-2017) and previous published articles [20, 29]. The annual breeding quantity of livestock was calculated as follows. If the breeding cycle was less than 1 year, the slaughtering amount was taken as the current year's breeding quantity, such as pigs, beef cattle and broilers. If the breeding period exceeded 1 year, the year-end stocks were the current farming amount, such as laying hens and dairy cattle [30].

Furthermore, a high-resolution spatio-temporal survey on the livestock farming in Beijing Municipality was conducted only during 2010-2014 [30]. The high-resolution spatial datasets were obtained by intensive field inventory survey of almost all livestock farming enterprises (including dairy cattle, meat cattle, pigs, broilers and laying hens) in the entire Beijing Municipality [30]. Our survey projects involved the number of annual reproduction (head), farming cycle, output amount, agricultural utilization and sewage treatment methods.

Estimation of Livestock-Derived NH₃, N₂O and CH₄ Emissions

The livestock-derived emissions of NH₃, N₂O and CH₄ in the B-T-H region were estimated based on the emission factor method. The total emissions of NH₃, N₂O and CH₄ in the B-T-H region were calculated by adding the emissions of all kinds of livestock in different years using the Equation (1) and Equation (2):

$$E_i = A_i \times EF_i \quad (1)$$

$$E = \sum_i (A_i \times EF_i) \quad (2)$$

where E_i is the emission of each livestock type (Gg); A_i is the annual breeding quantity of each livestock type (head); EF_i is the emission factor of each livestock type (kg head⁻¹ yr⁻¹), E is the total emission of NH₃, N₂O and CH₄ (Gg).

The NH₃ emission factors of the livestock farming were based on published literature [31-32]. Different management stages of livestock farming are considered, such as captive farming, fecal storage, returning to field application and grazing [21]. The N₂O and CH₄ emission factors are based on the emission factors for enteric fermentation and manure management of different livestock under different feeding methods given in the "Guidelines for the preparation of provincial greenhouse gas inventories" (No. [2011]1041) issued by China. All the emission factors are shown in Table 1.

Data Collection of Atmospheric Particulate Matter Concentration

The annual mean atmospheric particulate matter (PM_{2.5} and PM₁₀, size ≤2.5 μm and ≤10 μm, respectively) concentrations in the B-T-H region were all from the official website data bulletin of Beijing Environmental Statement (data of 2000-2018), Tianjin Ecology and Environment Statement (data of 2002-2018) and Hebei Province Ecology and Environment Condition Statement (data of 1998-2018).

Data Analysis

The temporal emissions of NH₃ and GHGs from livestock farming were shown using Origin 9.0 (OriginLab Corporation, Northampton, MA). The spatial emissions of NH₃ and GHGs from livestock farming were analyzed by Global Positioning System (GPS) and Geographic Information System (GIS) 10.2 (ArcMap, ESRI, MA). The linear relationship between the emission of NH₃ and atmospheric particulate matter (PM_{2.5} and PM₁₀) content was analyzed by SPSS 17.0.

Results

Spatio-Temporal Patterns of Livestock Farming

Over the past 40 years (1978-2018), the breeding structure of the five livestock farming types in the B-T-H region changed not much on this long time scale (Fig. 1). In the B-T-H region, the proportion of cattle and pigs breeding were relatively stable and reached

Table 1. The emission factors of the livestock farming (kg head⁻¹ yr⁻¹).

Livestock	NH ₃	N ₂ O	CH ₄	
			Enteric fermentation	Fecal management
Dairy cattle	37.61	1.846	88.1	7.46
Meat cattle	22.58	0.794	52.9	2.82
Pigs	11.55	0.227	1	3.12
Broilers	0.18	0.007	-	0.01
Laying hen	0.61	0.007	-	0.01

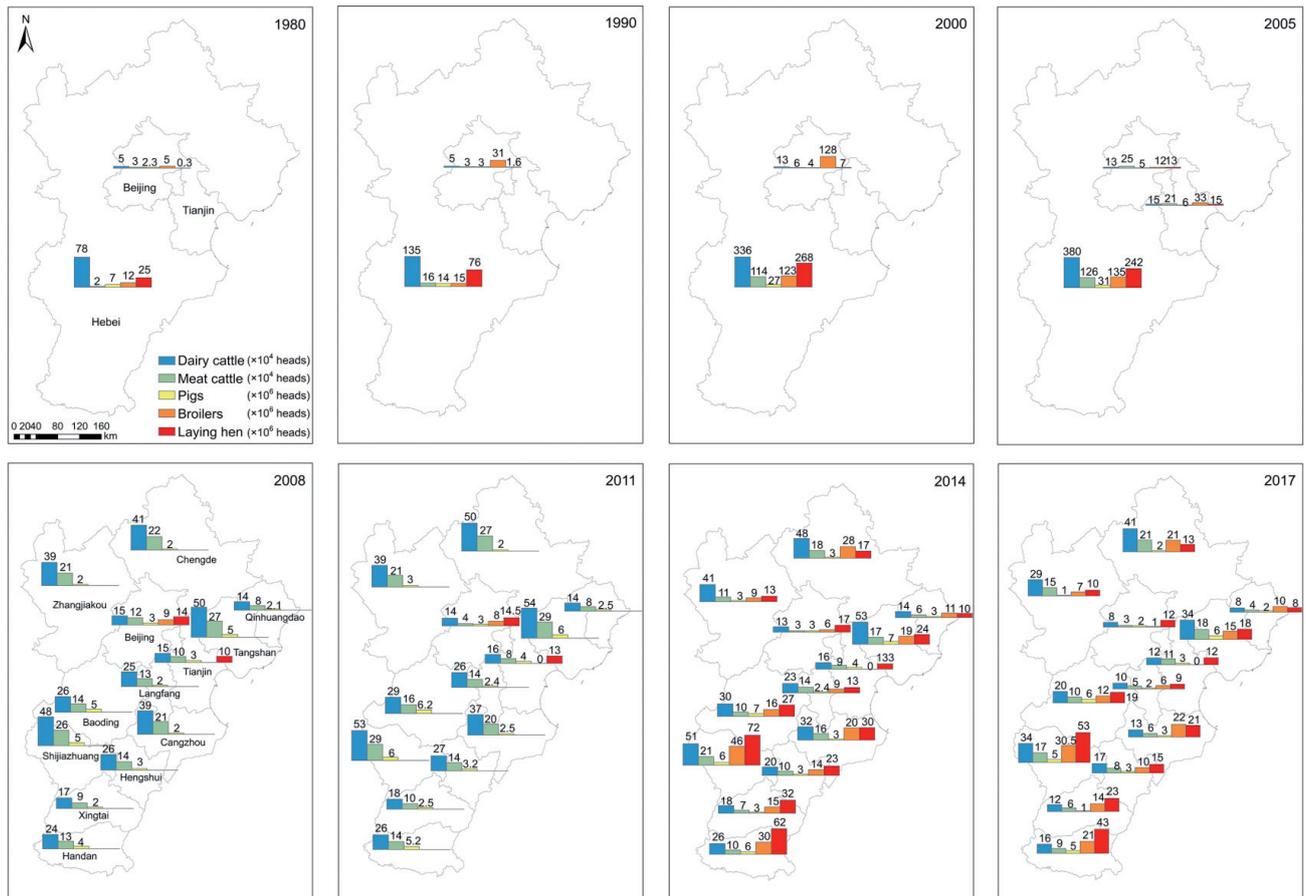


Fig. 1. The breeding amount of the five livestock types in the Beijing-Tianjin-Hebei region.

the peak in 2005 and 2014, respectively. In contrast, the production of pigs and broilers in Beijing reached its peak in 2003 and then declined slowly; and laying hens peaked in 2014 (Fig. 1).

The livestock farming enterprises were mainly distributed in suburbs of Beijing during 2010-2014 (Fig. 2). The number and distribution of livestock farming enterprises in Beijing changed little in the past 5 years. The southeast plain was dominated by dairy cattle,

meat cattle, pigs and laying hens. Broilers were mostly distributed in the northwest mountainous area (Fig. 2). During 2010-2014, the breeding quantity of pigs and laying hens increased slowly, while the rest showed a downward trend (Fig. 2). The number of dairy cattle, meat cattle and broilers in 2014 decreased by 8.8%, 40.4% and 46.8% compared with 2010, respectively. On the contrary, the number of pigs and laying hens increased by 1.5% and 60.1% respectively. In addition,

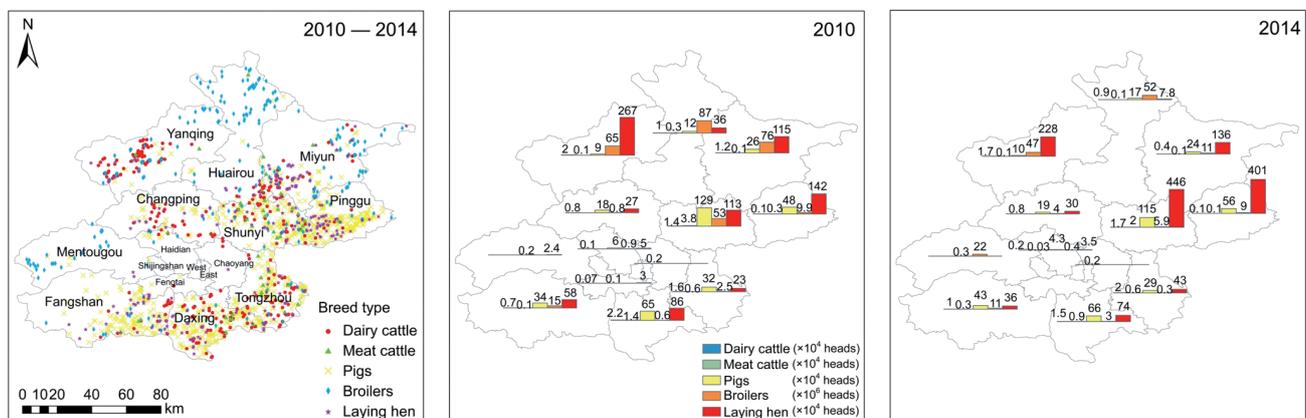


Fig. 2. Distribution of livestock farming enterprises and breeding amounts of the five livestock types in the Beijing Municipality during 2010-2014.

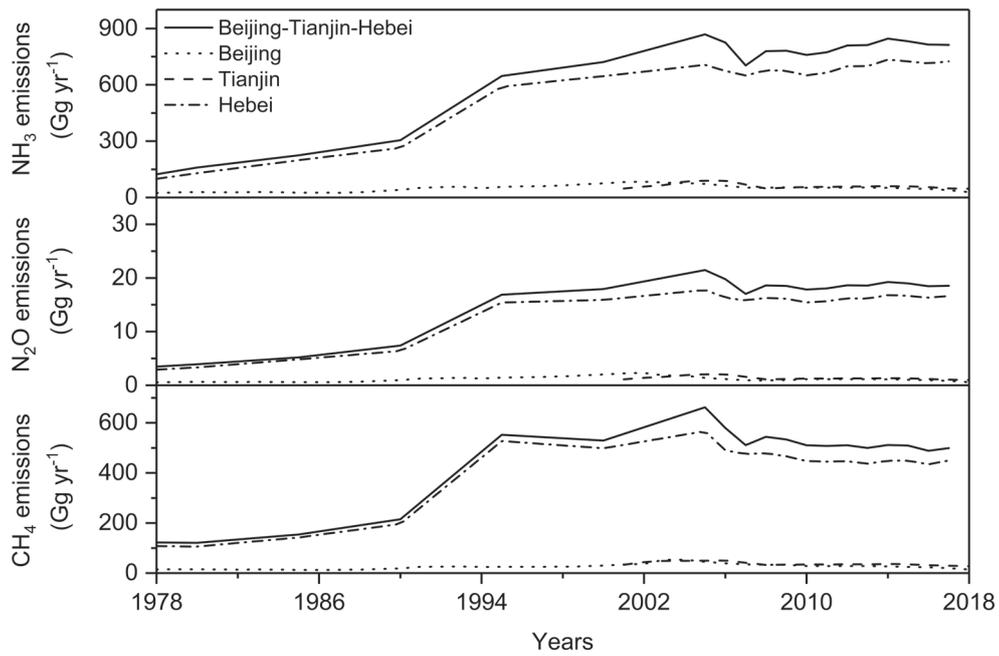


Fig. 3. Temporal emissions of NH_3 , N_2O and CH_4 from the livestock farming in the Beijing-Tianjin-Hebei region during 1978-2018.

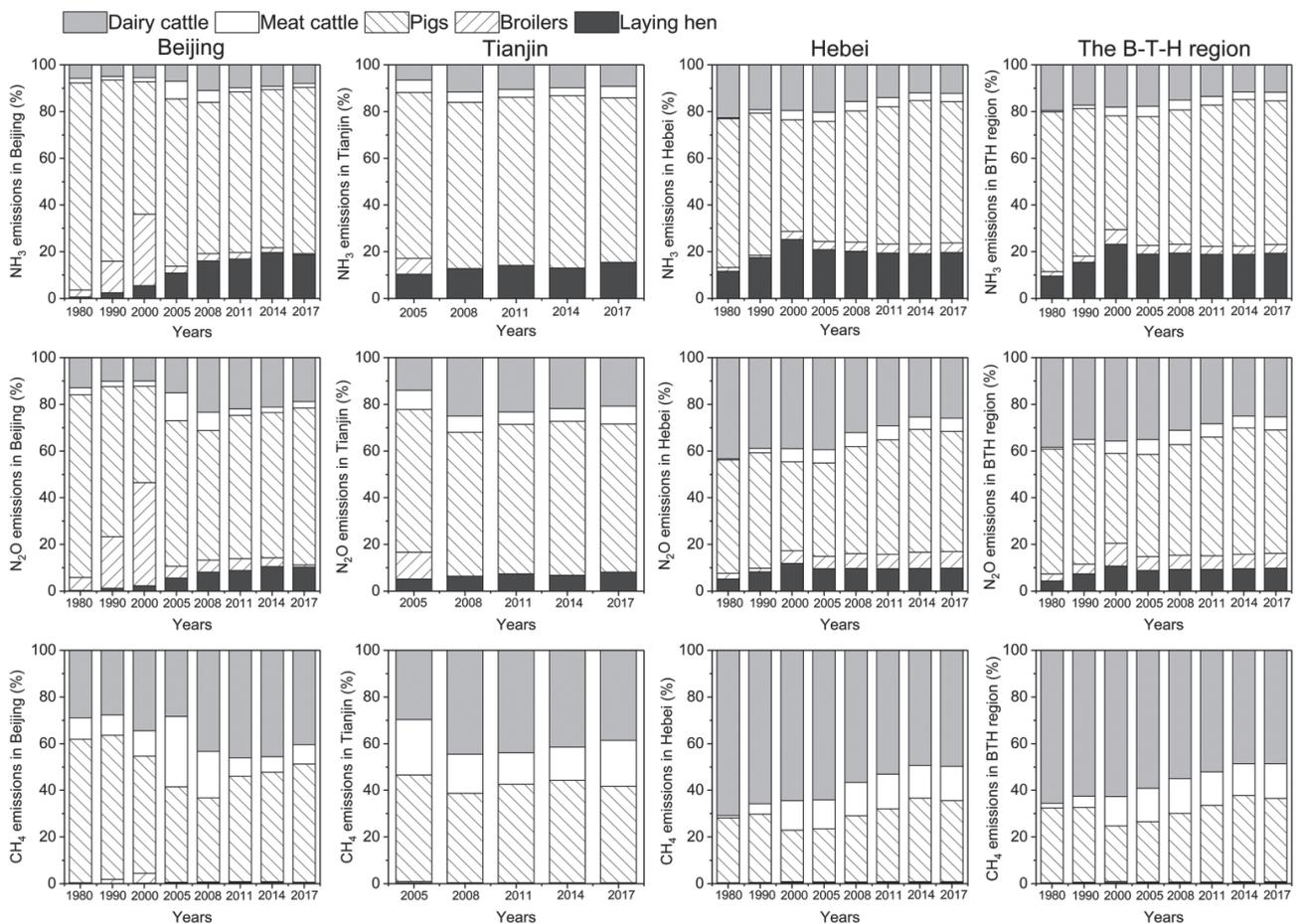


Fig. 4. The proportion of livestock-derived NH_3 , N_2O and CH_4 emissions in Beijing, Tianjin, Hebei and the Beijing-Tianjin-Hebei region.

the farming enterprises moved from the central urban area to the suburbs in Beijing.

Temporal Emissions of NH_3 , N_2O and CH_4 from Livestock Farming

The livestock-derived emissions of NH_3 , N_2O and CH_4 in the Beijing, Tianjin, Hebei and whole B-T-H region had similar patterns over the past 40 years (Fig. 3). They all experienced four development stages (1978-1990: low level; 1991-1995: rapid growth; 1996-2005: reached hot moments; 2006-2018: stable at high level). The livestock-derived emissions of NH_3 , N_2O and CH_4 were 813, 19 and 499 Gg in 2017, respectively. The Hebei Province accounted for the largest proportion of NH_3 , N_2O , and CH_4 emissions (80.3%-95.4%), especially in 1995. In contrast, the proportions of Beijing and Tianjin Municipality constantly decreased (Fig. 3).

The NH_3 and GHGs emissions of the five livestock types in the B-T-H region were dominated by the Hebei Province (Fig. 4). The contribution of broilers to total livestock in NH_3 and N_2O peaked in 2000 in Beijing (30.6% and 44.1% respectively) and then gradually decreased. On the contrary, the contribution of laying

hens increased year by year and reached the peak in 2014 (19.6% and 10.5% for NH_3 and N_2O , respectively). The culture structure in other areas changed little over time. In general, the NH_3 and N_2O emissions of livestock (except for dairy cattle and laying hens) decreased in Beijing and Tianjin Municipality. In contrast, the proportion of meat cattle, pigs, broilers and laying hens increased in Hebei, while only dairy cattle decreased (Fig. 4). This was consistent with the overall pattern of the B-T-H region.

Spatial Emissions of NH_3 , N_2O and CH_4 from Livestock Farming

The hotspots for NH_3 and N_2O emissions were the cities of Shijiazhuang, Tangshan and Baoding during 2008-2017 (Fig. 5). In terms of CH_4 emission, the cities of Tangshan, Shijiazhuang and Chengde had the highest emissions. In contrast, the cities of Qinhuangdao, Xingtai and Beijing had the lowest average emissions during 2008-2017. In general, the areas with higher NH_3 and N_2O emissions were mainly located at the junction of mountain and plain, while CH_4 emissions increased in the Northwest Mountainous Areas (Fig. 5).

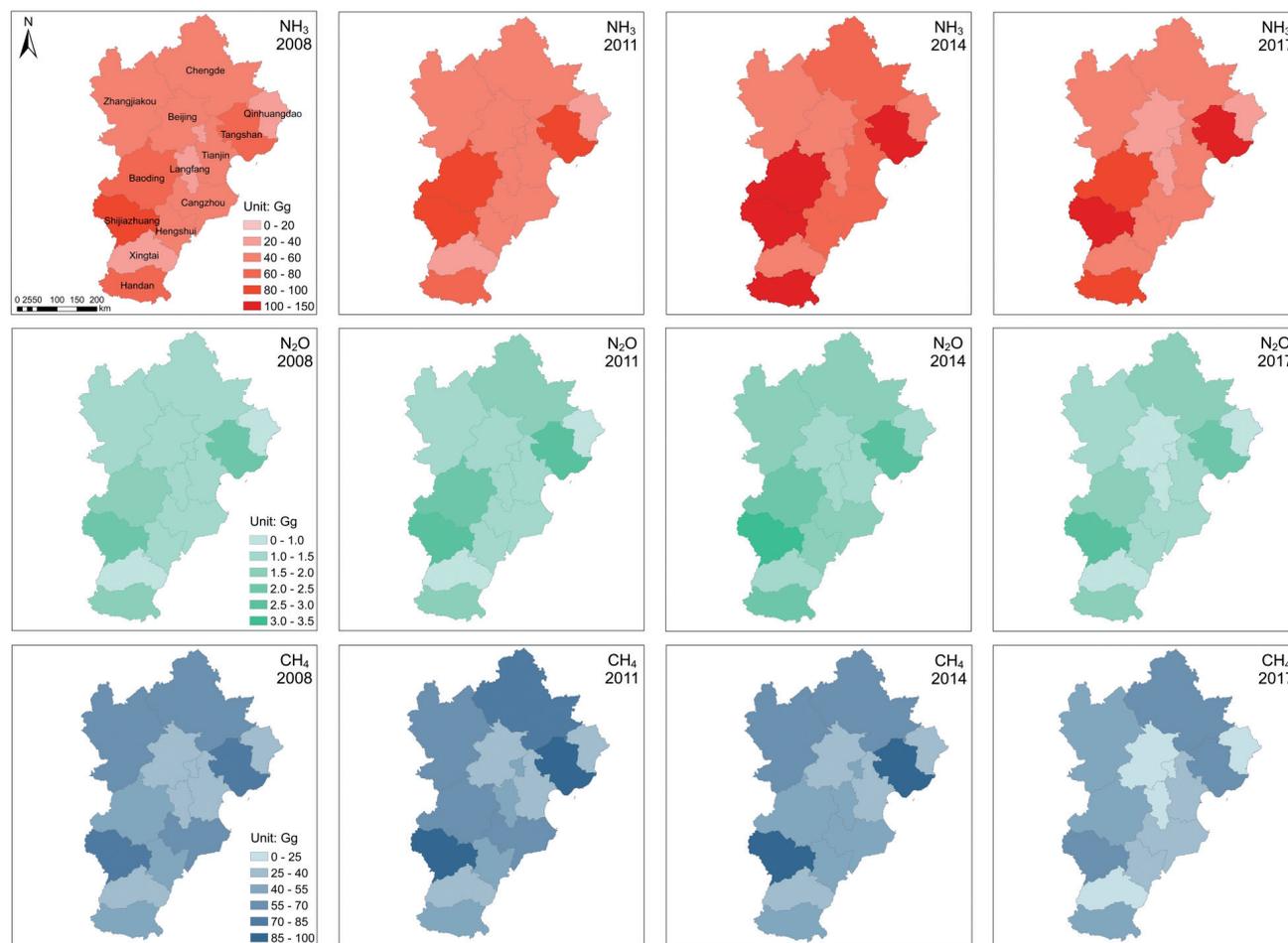


Fig. 5. Spatial emissions of NH_3 , N_2O and CH_4 (Gg) from livestock farming in the Beijing-Tianjin-Hebei region in 2008, 2011, 2014 and 2017.

Based on the emissions per unit area, the spatial emissions of NH_3 , N_2O and CH_4 changed a little bit during 2008-2017 (Fig. 6). The southeastern plain was the hotspot area, especially the cities of Shijiazhuang, Tangshan, Langfang and Handan (Fig. 6). The emissions of NH_3 , N_2O and CH_4 from livestock farming in Beijing, Tianjin and surrounding cities decreased in the past decade (2008-2017).

The spatial emissions of NH_3 , N_2O and CH_4 in Beijing changed little over the past 5 years (Fig. 7). The hotspots of NH_3 and N_2O emissions from livestock farming were mainly located in the west and northwest of Beijing, such as the town of Qianjiadian, Tanghekou and Changshaoying. In contrast, the hotspots of CH_4 emission were mainly in the eastern and southeastern Beijing, such as the town of Yangzhen, Dasungezhuang and Huoxian (Fig. 7).

Relationship between Livestock-Derived NH_3 Emission and Atmospheric Particulate Matter Concentration

There were linear relationships between the livestock-derived NH_3 emissions and the atmospheric

$\text{PM}_{2.5}$ and PM_{10} pollution in the B-T-H reign, especially for $\text{PM}_{2.5}$ (Fig. 8). The livestock-derived NH_3 emission per unit area explained 24% of the variation of atmospheric $\text{PM}_{2.5}$ concentration (Fig. 8a) and 22% of the variation of atmospheric PM_{10} concentration (Fig. 8 b).

Discussion

Spatio-Temporal Emissions of Livestock-Derived NH_3 , N_2O and CH_4

From 1960s to early 2000s, global livestock farming increased by 2 times, of which poultry and pigs increased by 3 and 1.2 times, respectively [33-34]. At the same period, the stock of cattle, pigs and poultry (laying hens and broilers) in the B-T-H region increased by 5.4, 3.2 and 12.6 times (Fig. 1), which were higher than the global level [33-34] and other countries [35-36]. Since then, the livestock of cattle, pigs, laying hens and broilers in the Beijing and Tianjin Municipality decreased sharply and moved to the suburbs. This was basically consistent with the research results of some

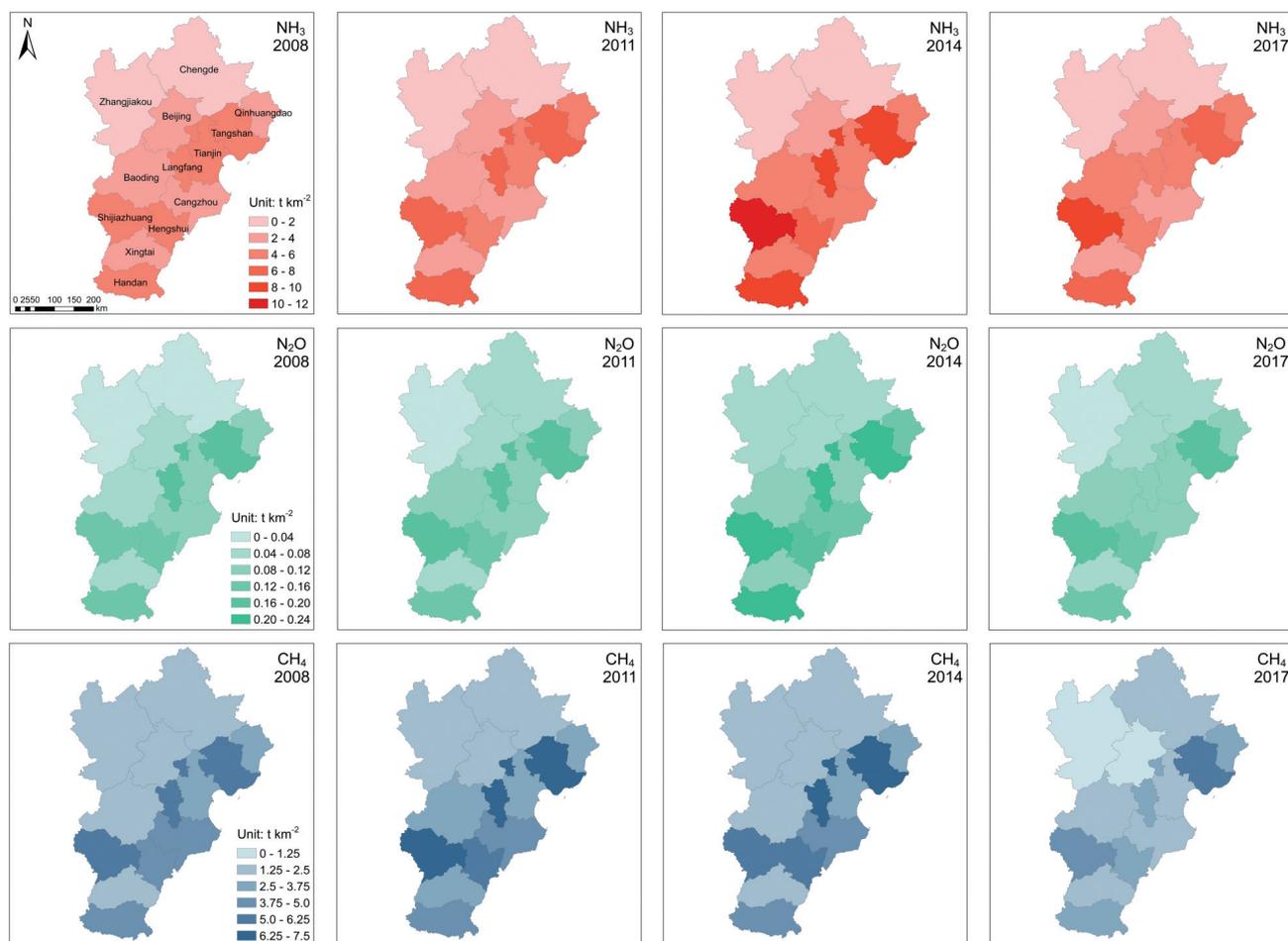


Fig. 6. Spatial emissions of livestock-derived NH_3 , N_2O and CH_4 (Gg km^{-2}) in the Beijing-Tianjin-Hebei region in 2008, 2011, 2014 and 2017.

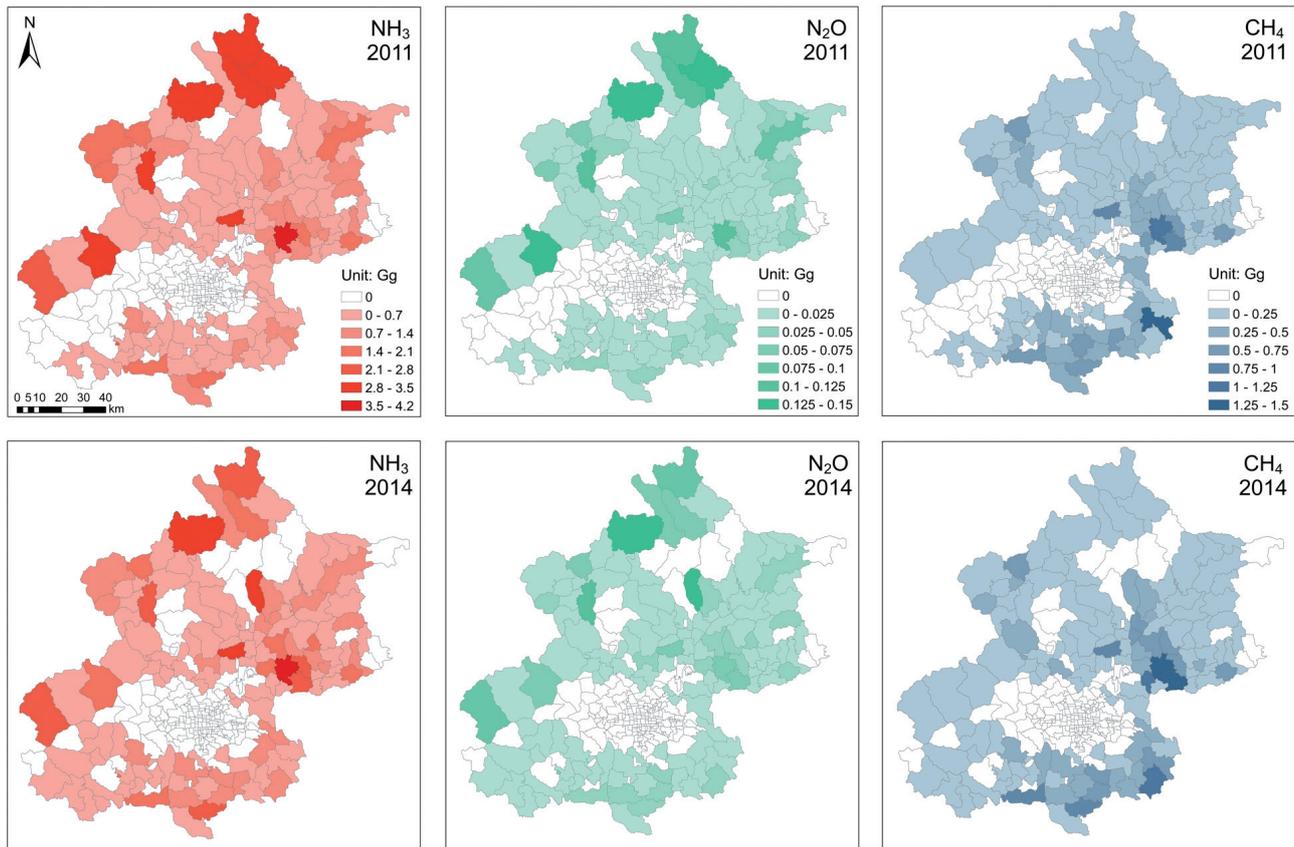


Fig. 7. Spatial emissions of livestock-derived NH_3 , N_2O and CH_4 (Gg) in the Beijing Municipality in 2011 and 2014.

scholars [13, 20]. This may be explained by the changes in population and livestock products increments, urbanization and green and sustainable development [13, 37].

The livestock-derived emissions of NH_3 , N_2O and CH_4 in the B-T-H region experienced four temporal periods with peaks in 2005 (Fig. 3), which was basically consistent with the pattern of entire China [34, 38-40] and other East Asian countries [36]. Our estimation of livestock-derived NH_3 emissions in the B-T-H region in 2010 (759 Gg) and 2015 (832 Gg) (Fig. 3) were 18% and 13% less than that (897 Gg in 2010) by Zhou et al. [21] and (942 Gg in 2015) by Ye et al. [3], respectively. However, our estimations of NH_3 and N_2O emissions in Beijing were 1.2 and 2.5 times higher than that of Liang et al. [29], which was consistent with the estimation results of Wei et al. [13]. These differences may be mainly due to different sources of data (field surveys), livestock types (such as including donkeys, mules, horses, rabbits, ducks and geese or not) and the use of slightly different emission factors.

At spatial scale, the hotspots were located in the southeast plain such as Shijiazhuang, Tangshan, Langfang and Handan (Fig. 6). This is similar to the relevant research in the B-T-H region [21, 29, 41] and some results in South Asia [39]. Among them, cattle and pigs were the main sources of NH_3 and GHGs emissions from livestock, accounting for 70-88% (NH_3),

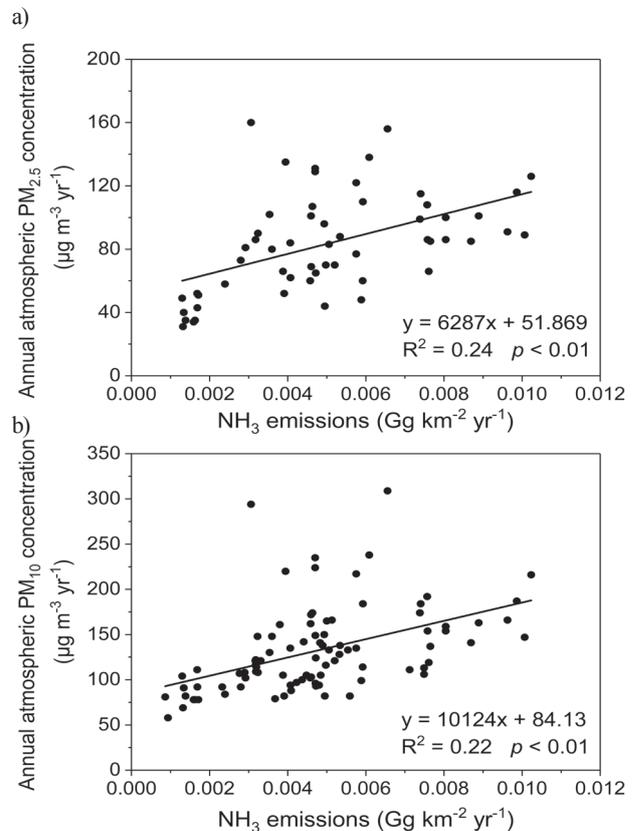


Fig. 8. Relationships between the livestock-derived NH_3 emission and annual mean atmospheric particulate matter ($\text{PM}_{2.5}$ and PM_{10}) concentration.

83-92% (N_2O) and 99% (CH_4) of the total emissions, respectively (Fig. 4). This is consistent with previous studies in the Red River Delta [36] and Jilin Province [42]. This spatial pattern may be related to the natural terrain advantage for southeast plain, the population and economic development degree [37].

Our research used a constant emission factor of each livestock type because of the long-term scale and unavailable dynamic emission factors. Even though this kinds of constant emission factors were from Chinese official guidelines (No. [2011]1041) and literature [31-32] thus should have reliabilities. As known, however, the actual emission factor of each specific livestock is related to the age and weight [26, 34, 43-44], geographical location [45], farming temperature [21] and feed composition [46] and other factors. Therefore, the constant emission factor of each livestock type certainly brought some uncertainty. In future research, dynamic emission factors [30, 47] should be employed, especially under the situation of developing livestock farming sector in China [48].

Relationship between Livestock-Derived NH_3 Emissions and Atmospheric Particulate Matter Pollution

We found that the livestock-derived NH_3 emission explained a quarter of variation of atmospheric $\text{PM}_{2.5}$ pollution and 22% variation of atmospheric PM_{10} pollution in The B-T-H region (Fig. 8). This is reasonable because NH_3 can react to form secondary particles such as ammonium nitrate (NH_4NO_3), ammonium hydrogen sulfate (NH_4HSO_4), ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) [49-50]. In addition, NH_4^+ can be combined with Pb, Cu, Fe, Zn, etc. through complexation to form secondary particles with larger diameters [51]. A substantial proportion of $\text{PM}_{2.5}$ pollution was caused by aerosol formation driven by NH_3 emission [5, 23]. Therefore, we conclude that livestock-derived NH_3 emission significantly affects the atmospheric particulate matter pollution in the B-T-H region.

Research conducted by the Ministry of Ecology and Environment indicated that coal consumption, industry activities, motor vehicles and dust mainly (90%) responded for atmospheric particulate matter pollution in the B-T-H region. The Tianjin Environmental Protection Bureau believed that other emissions such as livestock farming, catering and construction contributed approximately 7% to PM_{10} in 2014. In addition, livestock farming contributed approximately 10% to $\text{PM}_{2.5}$ in the B-T-H region [3, 28] and less than 4.5% to PM_{10} formation in Shijiazhuang [52]. Based on our result (Fig. 8), however, the influence of livestock-derived NH_3 emission on atmospheric particulate matter pollution in the B-T-H region may be underestimated. Therefore, more attention needs to pay on the effect of livestock-derived NH_3 emission on atmospheric particulate matter pollution in the future.

We found that the contribution of livestock-derived NH_3 emissions to $\text{PM}_{2.5}$ was higher than PM_{10} (Fig. 8). Compared with PM_{10} , $\text{PM}_{2.5}$ is more controlled by reaction-generated atmospheric particles, droplets and secondary particles. In contrast, the PM_{10} may be more derived from relative larger size of mineral particles such as sand dust, straw burning and so on [53]. In support, the NH_3 emission from pig farms increased the concentration of $\text{PM}_{2.5}$, especially the amount of nitrogen-containing particles [54-55]. Therefore, it is reasonable for the better relationship between the livestock-derived NH_3 emission and $\text{PM}_{2.5}$, compared with that of PM_{10} . Consequently, it is important to pay attention to the contribution of livestock-derived NH_3 emissions to $\text{PM}_{2.5}$ pollution.

“The European Air Pollutant Emission Inventory” [56] pointed out that intensive poultry houses (50%) and pig houses (30%) in Europe are important sources of atmospheric particulate matter. This indicated that livestock farming was one of the important sources of atmospheric particulate matter [12, 57]. Similarly, pigs and dairy cattle were also the main sources of NH_3 and GHGs emissions from livestock farming in the B-T-H region (Fig. 4). This is consistent with the sources of GHGs from China's livestock farming [58-59]. Therefore, the reduction of NH_3 and GHGs emissions should be tailored to different species of livestock (especially the pigs and cattle) and different regions.

Conclusions

Over the past 40 years, the livestock-derived emissions of NH_3 and GHGs (N_2O and CH_4) increased greatly until 2005 and subsequently remained stable in the Beijing-Tianjin-Hebei region. The southeastern plain was the hotspot, and the pig and cattle were the main sources (>78%) of NH_3 , N_2O and CH_4 emissions from livestock farming. Livestock-derived NH_3 emission accounted for approximately a quarter of the variation of atmospheric $\text{PM}_{2.5}$ and PM_{10} pollution, especially for the $\text{PM}_{2.5}$. In conclusion, the livestock farming, especially the breeding of pigs and cattle, should be paid more attention in the context of atmospheric $\text{PM}_{2.5}$ pollution and regional GHGs reduction.

Acknowledgments

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Conflict of Interest

The authors declare no conflict of interest.

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