Mitigating ammonia emission from agriculture reduces $PM_{2.5}$ pollution in the Hai River Basin in China

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Abstract:

The Hai River Basin (HRB), one of the most populated areas in China, is experiencing high NH_3 emissions, mostly from agricultural sources, and suffering from strongly enhanced $PM_{2.5}$ concentrations in all urban areas. Further increase of population and urbanization projected until 2030 may exacerbate this situation. Here, the NUFER (NUtrient flows in Food chains, Environment and Resources use) and GAINS (Greenhouse gas – Air pollution Interactions and Synergies) models have been coupled for the first time to understand possible changes of agricultural NH_3 emission between 2012 and 2030 and their impacts on ambient $PM_{2.5}$ concentrations, and to explore options to improve this situation. Results show that agricultural ammonia emissions in the HRB were 1179 kt NH_3 in 2012, 45% of which was from the hotspots at or near conurbation areas, including Beijing-Tianjin, Tangshan-Qinhuangdao, Shijiazhuang-Baoding, Dezhou, Handan-Liaocheng, and Xinxiang. Without intervention, agricultural ammonia emissions will further increase by 33% by 2030. The impacts of several scenarios were tested with respect to air pollution. Compared to the business-as-usual scenario, a scenario of improved technology and management combined with human diet optimization could greatly reduce emission (by 60%), and lead to 22-43% and 9-24% decrease of the secondary inorganic aerosols and $PM_{2.5}$ concentrations,

respectively, in the hotspots of NH_3 emissions. Our results further confirmed that ammonia control is needed for air pollution abatement strategies (SO_2, NO_x and primary PM reduction) to be effective in terms of $PM_{2.5}$.

Key words: Air pollution, atmospheric model, Beijing-Tianjin-Hebei region, Nitrogen, $PM_{2.5}$ concentration.

1 Introduction

Ammonia (NH_3) emissions, mainly from agricultural production, are increasingly contributing to $PM_{2.5}$ pollution in China (Wu et al., 2016). Long-term exposure to high levels of $PM_{2.5}$ has been linked to premature death from cardiovascular and cardiopulmonary diseases (Pope et al., 2002; Beelen et al., 2014). NH_3 emissions and $PM_{2.5}$ ambient concentration were all high in the Hai River Basin (HRB) (Sun et al., 2015; Kang et al., 2016), one of the most important agricultural regions and also most densely populated regions in China. Intensive agricultural production, along with excess nitrogen use, has led to high NH_3 emissions in this region. However, there is little research to comprehensively evaluate the potential of agricultural NH_3 emission reductions and their impacts on ambient $PM_{2.5}$ concentrations in the HRB region.

An accurate and detailed NH_3 emissions inventory is the basis for such an evaluation. Considerable efforts have been made to develop such inventories (Kang et al., 2016; Huang et al., 2012; Zhang et al., 2010; Zhou et al., 2015; Xu et al., 2016). Most of the emission inventories were based on the low-resolution (provincial or prefecture level) activity data, limiting their reliability in gridded results. Although some studies used county-level activity data, the emission factors were unavailable at that level. Therefore, large uncertainties remain with the current understanding of NH_3 emissions (Wang et al., 2013).

There are a number of pathways available to mitigate agricultural NH_3 emissions. These start out from improving technology and management of agricultural production (Ma et al., 2013; Chen et al., 2014; Chadwick et al., 2015), but also include reducing food waste (Garnett et al., 2013), and preventing future increase of production by limiting the consumption of resource-intensive diets (Ma et al., 2013; Gu et al., 2015; Tilman and Clark, 2014). Increased food imports can also reduce the pressure of production on Chinese agriculture. Possible changes in food production and consumption in China have been explored for the period 2010 to 2030 by the NUFER (NUtrient flows in Food chains, Environment and Resources use) model (Ma et al., 2016), considering pathways mentioned above and assuming that China's population is expected to peak around 2030 (United Nations, 2015). Abatement technologies have been developed to reduce agricultural emissions (Bittman et al., 2014). With given or assumed knowledge on the stringency of current legislation and future ambition to reduce emissions, future pathways can be translated into emission scenarios (Rao et al., 2017).

Linking emissions of air pollutants to air quality requires the use of atmospheric models, specifically chemical transport models. More generalized information can be drawn from integrated models, which incorporate the results of such atmospheric models and produce source-receptor (or even emission-impact) relationships directly. Integrated assessment models have been applied on the national (Oxley et al., 2013) as well as regional (Kiesewetter et al., 2015) scales. For the latter case, the GAINS (Greenhouse gas – Air pollution Interactions and Synergies) model brings together bottom-up emission calculations from all economic sectors, emission control technologies and their emission factors for various pollutants, and further assesses the impacts of $PM_{2.5}$ concentration on human health (Kiesewetter et al., 2015). The GAINS model has been applied for several world regions, including China (Chen et al., 2015).

The objectives of this study are: (1) to develop a detailed and reliable agricultural NH_3

emission inventory for the HRB region for the year 2012; (2) to explore how the agricultural NH_3 emissions in the HRB region will change between 2012 and 2030 under a range of scenarios; and (3) to evaluate the impacts of the changes of agricultural NH_3 emissions on air pollution ($PM_{2.5}$ concentration).

2 Materials and methods

The study domain covers some of the most intensively used agricultural area in China, the Hai River Basin, encompassing the two municipalities Beijing and Tianjin, most of Hebei province, the eastern part of Shanxi province and the northern parts of Henan and Shandong provinces (Fig. S1 in the Supplementary Information). . We used county-level statistical data, local parameters, and coupled the NUFER with the GAINS model for the first time to develop and apply future emission scenarios used in this analysis. All details are described in the supplementary Information to this work, including data tables and figures. The following section presents a summary.

2.1 Ammonia emission estimation

Agricultural ammonia emissions were calculated from activity data on crop and livestock production and corresponding emission factors (EFs) specific for local conditions according to the following Eq. (1):

$$E_{NH_3} = \frac{17}{14}\sum(A_i \times EF_i) \ , \qquad (1)$$

Where E_{NH_3} is the estimated total agricultural NH_3 emissions; 17/14 is the conversion coefficient of NH_3-N emissions to NH_3 emissions; i represents the source type; A_i is the source specific activity data; EF_i is the corresponding NH_3 emission factor (often presented as the share of N used/applied, hence expressed in NH_3-N).

2.1.1 Activity data sources

Here county-level agricultural activity data, derived from a variety of local statistical yearbooks, for 2012 were used to generate an inventory of agricultural NH_3 emissions. County-level activity data provided more detail on spatial variation of mineral fertilizer application and livestock production than provincial and prefectural level data.

Arable land emissions depend on crop-specific fertilizer application, fertilizer type, and soil properties (as pH). In this study we considered five main crop types (wheat, maize, soybean, vegetable, and fruit), seven mineral fertilizer types (urea, ammonium bicarbonate – ABC, ammonium nitrate – AN, ammonium sulfate – AS, diammonium phosphate – DAP, compound fertilizer – NPK, and other), and two fertilization methods (basal and top dressing). The National Development and Reform Commission (NDRC) (NDRC, 2013) provided the application rate of each crop-specific synthetic N fertilizer for each province (Table S3). Furthermore, the information on fertilization methods of each synthetic fertilizer type for each crop type (Table S4) was derived from a comprehensive farm survey covering 400 individual farms in 11 typical counties in the Hai River Basin (Fig. S1). These farms represented the typical topography of the region that varies from piedmont plains, low plains to peri-urban, and also covered the representative crop-based farm types, including cereal farms that only grew wheat in winter and maize in summer in rotation, vegetable farms that grew vegetables only or vegetables as well as cereals, and fruit farms that grew fruits only or fruits as well as cereal crops.

For the NH_3 emissions related to livestock production, we considered six main animal classes: pigs, beef cattle, dairy cattle, laying poultry, meat poultry, and sheep & goats.

Pigs, beef cattle and dairy cattle manure management systems were further divided into liquid and solid sub-systems, respectively. Grazing excreta were also considered in the calculation for dairy cattle, beef cattle, and sheep & goat systems. We used the provincial-level N excretion rates in different livestock systems (Table S6) estimated by Bai et al. (2016) using the NUFER model. A mass-flow approach, livestock stage-specific feeding structures based on farm interviews, and nitrogen retention in milk, egg, and body weight gain from statistical data were used by the NUFER model to estimate the average nitrogen excretion at provincial scale, so the N excretion rates were more reasonable and detailed than the averages from literature.

2.1.2 Emission factors

The emission factors described above specifically consider the Chinese situation. For the mineral fertilzier application, the crop- and fertilizer-specified emission factors (Tabe S7) were developed from published literature taking advantage of local conditions, specific shares of fertilizers, and fertilization methods as described above.

NH_3 emissions related to livestock production were assessed from a mass-consistent N-flow model on manure. Here the NUFER model provided information required to describe the losses to the respective environmental media in each of the stages (housing, storage & treatment, and application), including NH_3, N_2O, N_2, leaching, and direct discharge (Bai et al., 2016). The assessment specifically accounts for N lost in discharge to surface water, which led to the decrease of NH_3 emissions from manure management chain. We further updated the manure direct discharge factors in manure management chain for different livestock systems based on the results of farm interview (Table S8) on landless and mixed crop-livestock farms. Other emission factors could be found in the study by Bai et al. (2016). These emission factors were used as inputs to the GAINS model and, together with activity numbers specifically developed from Chinese statistical information (see activity data sources above) used for further calculation.

2.1.3 Spatial allocation

Land use data on 1 km ×1 km grid, provided by the Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (RESDC) (http://www.resdc.cn), were used to explore the spatial distribution of agricultural NH_3 emissions. The NH_3 emissions from arable lands were then evenly allocated to the arable areas in each county, assuming that all the available animal manure in each county was applied to the arable lands within this county. NH_3 emissions from animal housing, manure storage, and treatment stages were evenly allocated to the rural settlement areas in each county. To reduce the uncertainties, the resulting 1-km maps were aggregated to 5-km resolution.

2.2 Evaluation of impacts of ammonia emissions on $PM_{2.5}$

GAINS uses a source-receptor matrix to assign $PM_{2.5}$ concentrations (annual mean values) and concentration changes to the emissions derived according to the respective scenarios. The underlying methodology has been described in detail (Kiesewetter et al., 2015). The source-receptor matrix used here is the result of multiple sensitivity simulations of the European Monitoring and Evaluation Programme (EMEP) Chemical Transport Model (CTM), a hemispheric model that also covers the area of China (Simpson et al., 2012) and describes the impact of changes in emissions of primary PM and PM precursors SO_2, NO_x, NH_3, and VOC, on ambient $PM_{2.5}$ concentrations on a 0.5 degree grid. Emissions of other compounds (SO_2, NO_x, and VOC) than NH_3 were obtained from the scenarios developed for the International Energy Agency (IEA, 2016). We evaluated the impact of NH_3 emission changes on secondary inorganic aerosols (SIA) and in consequence also $PM_{2.5}$ concentrations for 2030 under several scenarios previously developed to reflect future pathways of the Chinese agricultural system.

Responses of $PM_{2.5}$ concentrations to NH_3 emission changes are calculated via linear transfer coefficients which have been generated by reducing NH_3 emissions by 15% under present day (2015) atmospheric conditions in the CTM. Under strong reductions of NH_3 emissions as envisaged in some of the scenarios discussed below, we may assume that SIA formation becomes so strongly NH_3-limited that no ammonium nitrate is formed any more. Hence a lower boundary of SIA concentrations was calculated by setting the contribution of NO_x emissions to zero. We note that this lower range still does not consider that also ammonium sulphate formation may become NH_3-limited, i.e. that the contribution of SO_2 emissions to SIA formation may decrease more strongly than suggested by the present-day SO_2 transfer coefficient (Kiesewetter et al., 2015).

2.3 Scenarios

For sectors other than agriculture, anthropogenic activities and emissions used in this study were kept identical to those in the New Policies Scenario of the International Energy Agency (IEA) (IEA, 2016) assessment, assuming economic development as projected by the Organization for Economic Co-operation and Development (OECD) and application of currently agreed emission control legislation. Projected population growth rate for the whole of China taken from United Nations (2015) was used in the Hai River Basin.

Six scenarios developed previously (Ma et al., 2016) were used to estimate the demand changes for domestic agricultural production between 2012 and 2030 in the HRB region, such as the area of each crop type, the amount of synthetic fertilizer application, and animal numbers. Also the impacts of these changes on agricultural NH_3 emissions and air quality were quantified. The key information of these scenarios is explained below.

Scenario 0: Business As Usual (BAU). This scenario assumes that (1) with a rapid increase of income in rural areas, the diet (specifically, the meat consumption) of Chinese rural people in 2030 will be the same as that of the urban population in 2010, while the diet for the urban population will not change; (2) there is no change in food import rates between 2010 and 2030; (3) the agricultural practice in 2030 will also be the same as that in 2010.

Scenario 1: Consume and Waste Less (CWL). This scenario assumes that (1) the recommendations of the Chinese Dietary Guidelines (2007) will be fully adopted by rural and urban populations in 2030; (2) at the same time, food waste during food production and supply chain will be reduced by 20% relative to 2010; (3) the food import rates and the practice of agricultural production in 2030 will be the same as that in 2010.

Scenario 2: Import More Food (IMF). With the same demand for agricultural products in 2030 as that in the BAU scenario, the IMF scenario assumes that in 2030, (1) the soybean import rate will be expected to be stable at 84%; (2) the share of milk imported will increase to 20%; (3) self-sufficiency in all other plant-based and animal-based foods and feeds will reach 90%.

Scenario 3: Producing More and Better (PMB). Under the same demand for domestic agricultural products and food import rate in 2030 as in the BAU scenario, the PMB scenario assumes for the management of livestock production that, compared to 2010, (1) the shares of liquid and solid systems for dairy, beef cattle and pig systems will not change in 2030; (2) livestock productivity will increase by 20% for pig and meat poultry, and 40% for other animal types via improved feeding in 2030; (3) manure discharge into rivers will completely disappear in 2030, manure will be applied on fields instead; (4) the NH_3 emission rates in animal housing, manure storage, treatment, and application stages will all reduce by 50%, due to the extensive application of several technologies, such as covered storage of manure, low ammonia application (deep application, etc.), bio-filtration, animal housing adaption, and their combination. For crop production management, it is assumed that relative to 2010, (1) the productivity of rice, wheat, maize, vegetable and fruit production will increase by 17%, 45%, 70%, 25% and 25% in 2030, assuming that the Integrated Soil-Crop System Management (ISSM) technology (Chen et al., 2014) is fully adopted; (2) the fertilizer N application rates for rice, wheat and maize will not change, and the fertilizer N application rates for vegetable, soybean and fruit will decrease by 30% in 2030. The increased animal manure availability by reducing animal manure losses is expected to reduce the application of synthetic fertilizer.

Scenario 4: PMB+CWL. Scenario 4 combines the assumptions developed for PMB and CWL.

Scenario 5: PMB+CWL+IMF. This scenario is a combination of PMB, CWL and IMF scenarios.

3 Results

3.1 The agricultural ammonia emission inventories in HRB

The total agricultural NH_3 emissions in the HRB region were estimated as 1179 kt NH_3 yr^{-1} in 2012. Animal manure and synthetic fertilizer application accounted for almost equal shares of total agricultural emissions, 51% and 49%, respectively. Pigs, laying poultry, beef cattle, dairy cattle, sheep & goat, and other poultry contributed 32%, 19%, 18%, 14%, 10%, and 7% to livestock emissions, respectively. Urea and ABC accounted for 89% of total NH_3 emissions from synthetic fertilizer (Fig. 1).

Under the BAU scenario, total agricultural NH_3 emissions would increase by 33% relative to 2012, reaching 1571 kt NH_3 yr^{-1} in 2030. NH_3 emissions from livestock manure and synthetic fertilizer application would increase by 59% and 7%, respectively (Fig. 1); livestock manure would thus contribute 60% of total agricultural NH_3 emissions. This mainly resulted from the demand increase for the domestic animal-derived food (meat, milk, eggs) by 33-93%, and therefore for feed products by 27-34% (Table S8).

Human diet optimization in parallel with reduction of food waste (CWL scenario) would significantly reduce total agricultural NH_3 emissions in 2030 by 27% compared to BAU scenario, synthetic fertilizer and livestock manure responsible for 12% and 15%, respectively. Relative to 2012, the reduction of synthetic fertilizer emissions in 2030 (24%) would be offset by the increase of livestock emissions (18%), and consequently the agricultural emissions in 2030 would be similar to that in 2012 (Fig. 1). The dairy cattle system, approximately five times the 2012 level, is the only contributor to the elevated livestock NH_3, due to the huge increase in recommended intakes of milk.

The increase of food imports (IMF scenario) could also reduce agricultural NH_3 emissions by 31% and 8%, relative to the BAU scenario in 2030 and 2012, respectively. The emissions from synthetic fertilizer application and livestock manure would be 25% and 34% lower than those under BAU scenario, respectively. Compared to 2012, emissions from synthetic fertilizer application would decrease by 20%, while livestock emissions would increase by 4% (Fig. 1).

Improvements of technology and management (PMB scenario) could greatly reduce NH_3 emissions by 45% and 26% relative to the BAU scenario in 2030 and 2012, respectively, being more successful in emission reductions than the CWL and IMF scenarios. Urea and ABC were the main contributors to the emission reductions, accounting for 35% and 70% of the total reductions relative to 2030 of the BAU scenario and 2012, respectively (Fig. 1).

The combination of different strategies could further reduce the agricultural NH_3 emissions. The combination of the improvements of technology and management with change in human diet (PMB+CWL scenario) could decrease total NH_3 emissions by 60% and 47% relative to 2030 of the BAU scenario and 2012. Urea and ABC were also the main contributors to the emission reductions, accounting for 34% and 53% of the total reductions relative to 2030 of the BAU scenario and 2012, respectively. Among all agricultural sources, only dairy cattle emissions would increase, due to the human diet change. Considering improvement of food import in addition could lead to a further decrease of NH_3 emissions of 6 - 8% (Fig. 1).

3.2 The spatial distribution of ammonia emissions

Fig. 2 shows the spatial patterns of agricultural NH_3 emissions in 2012 and, for comparison, in 2030 under strong emission reductions (PMB+CWL scenario). Several NH_3 emissions hotspots appeared in 2012, which cover 17% of the basin area but contributed approximately 45% of the total agricultural NH_3 emissions. The NH_3 emission densities from arable land and animal manure in hotspots were consistently higher than in other regions. This was mainly due to the higher nitrogen fertilizer application rates and livestock density than other regions (Fig. S2). These emissions hotspots are situated close to the big cities, such as Beijing, Tangshan, Shijiazhuang, Baoding, Dezhou, Handan, Liaocheng, and Xinxiang. The NH_3 hotspot emissions in 2030 under the PMB+CWL scenario would be clearly lower than those in 2012.

Fig. 3 reveals that the NH_3 emissions densities and sector contributions vary among the hotspots, as well as between 2012 and 2030. The lowest average emission density in 2012 occurred in Tangshan-Qinhuangdao with 7 t km^{-2}, and the highest value in Shijiazhuang-Baoding with 12 t km^{-2} (km^2 represents the land area). Synthetic fertilizers and animal manure accounted for almost equal shares of emissions for all hotspots other than Tangshan-Qinhuangdao and Dezhou regions, where animal manure was the dominant emission source. Urea and ABC contributed more than 80% of synthetic fertilizer emissions for all hotspots. Dairy cattle, beef cattle, laying poultry, and pigs were the largest livestock emission sources for Tangshan-Qinhuangdao, Dezhou, Handan-Liaocheng, and other hotspots, respectively. Under the PMB+CWL scenario, Shijiazhuang-Baoding would still have the highest average emission density with 7 t km^{-2} in 2030, while the lowest value with 4 t km^{-2} would occur in Beijing-Tianjin, Handan-Liaocheng, and Xinxiang regions. All hotspots other than Handan-Liaocheng region, would be dominated by livestock emissions, especially dairy cattle emissions.

3.3 Impact of NH_3 emissions on $PM_{2.5}$ concentration

We estimated the population-weighted concentrations of $PM_{2.5}$ (annual mean values, results obtained by model grid cell were averaged over HRB weighted by population) and SIA for 2012 and 2030 under each scenario. Fig. S3 shows the distributions of $PM_{2.5}$ and SIA concentrations for 2012 and 2030 under the BAU and PMB+CWL scenarios. We further evaluated the impacts of NH_3 emissions reductions on $PM_{2.5}$ and SIA concentrations in the hotspots of NH_3 emissions (Fig. 4). Although the $PM_{2.5}$ concentrations in 2030 under all scenarios would be lower than those in 2012, the future NH_3 emissions reductions can further decrease the $PM_{2.5}$ concentrations via reduction of SIA. Taking the comparison between the BAU and PMB+CWL scenarios as an example, 60% agricultural NH_3 emissions reduction would lead to a decrease in mean $PM_{2.5}$ concentrations by 9-14% in the standard calculation, or by 12-24% in the "lower boundary" case (see methods section) in the hot spots – which corresponds to 22-27% (or 29-43% for the "lower boundary" case) reduction of SIA. The greatest reductions of $PM_{2.5}$ concentrations occur in the Xinxiang region with 12 μg m^{-3} (18 μg m^{-3}). Shijiazhuang-Baoding would still have the highest $PM_{2.5}$ concentration among all the hotspots at 77 (75) μg m^{-3} (calculated at the 0.5° resolution, which ignores smaller-scale urban concentration increments that are mostly related to low-level primary PM sources in cities).

4 Discussion

4.1 Comparison with previous studies

In order to indicate the levels of the emission inventory uncertainty, we compared in detail our results with a number of available studies. Fig. 5 presents a comparison of the NH_3 emissions from synthetic fertilizer application in this study in 2012 with literature data on the province level. We chose two existing inventories originating from Chinese institutions (Huang et al., 2012; Zhou et al., 2015), and a previous implementation of NH_3 emissions in the GAINS model deriving from the ECLIPSE V5 scenario (Klimont et al., 2016). Our estimates were 9%-28% lower than 2010 emissions reported for Beijing, Tianjin, and Hebei province (Zhou et al., 2015). The disparity was mainly caused by the consideration of basal dressing, especially for N compounds application in our study, which resulted in the lower emission factors and thus ammonia volatilization (Cai et al., 2002; Li and Ma, 1993; Zhang et al., 1992). Compared to the

2010 emissions of the GAINS/ECLIPSE V5 scenario, our results were lower for all the provinces other than Tianjin. The differences of data sources and emission factors could explain the lower emissions in this study compared to the 2010 estimate in that GAINS/ECLIPSE V5 scenario, which used the data of mineral fertilizer from FAO (FAO, 2016). Compared to this study, total mineral N fertilizer applications were higher in the GAINS/ECLIPSE V5 scenario for all provinces other than Tianjin and Henan province, but the shares of urea and ABC were lower for all provinces. The ammonia emission factors were specified for urea (including ABC) and other mineral N fertilizer in GAINS, while in this study we collected information on seven mineral fertilizer categories with two fertilization methods. Compared to the published estimates for 2006 (Huang et al., 2012), our results were lower for Henan province, but higher for other provinces. The disparities might be mainly attributed to the changes of fertilizer composition, fertilization method between 2006 and 2012, and the differences in emission factors. Compound fertilizer consumptions in each province were all higher in 2012 than 2006. According to the farm interview, most of compound fertilizer were deep applied to the fields. In our study, the emission factors were mainly based on reported local measurement results (Table S7), while Huang et al. used the emission factors parameterized by fertilizer–N application rate, temperature, and fertilization method. The changes of fertilizer composition between 2006 and 2012, and the different sources of fertilizer application rates and fertilization method all contributed to the disparities of emission factors.

We also compared the NH_3 emissions from the animal manure management chain to the previous estimates to analyze the role of the consideration of manure discharge, activity data and parameters. According to the farm interview, about 19-57% of manure N were directly discharged to surface water from animal housing, which caused considerable water pollution while reducing NH_3 emissions. Selected parameters, including N excretion rates and emission factors, were determined by NUFER model based on national statistical data and local farm interview. Our results for 2012 were 45-58% smaller than 2010 estimations for Beijing, Tianjin and Hebei province reported by Zhou et al. (Zhou et al., 2015). The consideration of only main livestock types, including dairy cattle, beef cattle, pig, sheep & goat, meat poultry and laying poultry, in our calculation is to some extent offset by the increase of animal numbers in 2012 compared to 2010, so the disparities are explained by these authors ignoring losses from animal manure management chain and applying different parameters. All these were also responsible for the higher contributions of domestic animals (77-78%%) and lower contributions of poultry (22-23%) to the total livestock NH_3 emissions in Beijing, Tianjin and Hebei province in this study than those estimated by Zhou et al. (2015).

Our emission estimates were 10% and 58% lower for Beijing and Shanxi, and 26-50% higher for Shandong, Henan and Tianjin than the 2010 estimates by the GAINS - ECLIPSE V5 scenario. These disparities could be attributed to the different data sources and emission factors, as well as the consideration of N losses via animal manure discharge, which was ignored by that previous GAINS scenario. Our results on livestock emissions were higher than those estimated by Huang et al. (Huang et al., 2012) for Beijing, Tianjin, while lower for other provinces. The consideration of animal manure direct discharge to surface water and the different parameters, including annual total ammonia nitrogen (TAN) excretion per animal for each livestock class and NH_3 emission factors from each stage of livestock manure management, contributed to the disparities. The N losses via animal manure discharge were not included in their estimation. The parameters applied by Huang et al. were mainly derived from literature or foreign studies, such as EEA's guidelines (EEA, 2013) and the same among all the Chinese provinces.

In order to verify our estimation for the $PM_{2.5}$ concentration, we compared our results with the monitoring data in 2014 provided by China Air Quality Online Monitoring and Analysis Platform (https://www.aqistudy.cn/) (Table S10), considering the monitoring data available. For the main cities, the annual mean concentrations of $PM_{2.5}$ were similar between 2012 and 2014.

4.2 The effect of the changes in NH_3 emissions

Considering the increase of NH_3 emissions in 2030 under the BAU scenario relative to 2012, projected possible emission reductions of particulate matter, SO_2 and NO_x (IEA, 2016) would lead to 14-21% decrease of $PM_{2.5}$ concentrations in the hot spots. The decrease of $PM_{2.5}$ concentrations confirms that NH_3 emission reduction should be paid more attention in the future for the control of $PM_{2.5}$, especially of SIA (Wang et al., 2013). Besides the effect of NH_3 emissions on air quality, the nitrogen use efficiency of agricultural production will be greatly improved by such reduction measures. However, the population-weighted $PM_{2.5}$ concentrations in the hotspots would be still higher than the global population-weighted mean (20 μg m^{-3}) (Van Donkelaar et al., 2010) and the Chinese annual average standard (35μg m^{-3}) (GB 3095-2012, 2012). Therefore the further control of emissions of particulate matter, SO_2 and NO_x cannot be ignored in the future.

4.3 Pathways of NH_3 emission reduction

It is clear that structural changes in food production and consumption, as well as the improvements in nutrient management have an impact on NH_3 emissions. The increase of NH_3 emissions in 2030 under the BAU scenario can be almost offset by increasing food import (IMF scenario) or optimizing the human diet (CWL scenario). Compared to the increasing food import, the dietary change and reduced food losses scenario is more advisable, because it is beneficial to human health and the environment simultaneously. Our analysis was based on the dietary recommendations of 2007. As the updated dietary guidelines (Chinese Nutrition Society, 2016) recommend reduced intakes of meat and fruit, the ammonia emissions under those circumstances would decrease even further. However, the challenge lies in how the optimized diet can be adopted by all the people. Although human health awareness continues to grow with the ageing of populations and increase in diet-related diseases (Popkin et al., 2001), the behavioral patterns are not always consistent with these attitudes, due to the lack of knowledge on nutrition. Therefore, nutrition education has been demonstrated to be one effective way to healthy diet and physical activity (Wang et al., 2015; Aikman et al., 2006) and offers benefits also in terms of reducing ammonia emissions.

The improvement of nitrogen management is the key for NH_3 emission reductions, according to the results under the PMB scenario. The ISSM technology in the crop production sector has illustrated the possibility to increase crop yields, therefore reduce NH_3 emissions under the policy of "Zero Growth in Synthetic Fertilizer Use from 2020" (MOA, 2015). However, smallholder farming with small parcels of land (around 0.1 ha) (Chen et al., 2011) dominates the agricultural production, as a result of the household contract responsibility system in China. Low agricultural incomes and the rapid economic development drive the farmers to make a living from off-farm work rather than manage crop or soil. Meanwhile, most of the farmers lack knowledge. As a result, non-optimal N management is common in China, and implementation of these new techniques and measures by farmers faces a big challenge. Some pathways have been explored to disseminate new knowledge and technologies. For instance, the Science and Technology Backyard (STB) platform (Zhang et al., 2016), enabling the farmers, university, industry, and government to work together, is an illustrated pathway. In order to popularize the new knowledge and technologies, the agro-technical team should be further scaled up, stabilized, and perfected, however, this will to a large extent enhance the running cost under the current circumstances. The combination of STB platform with enlarging farm size and decreasing fertilizer subsidies might be more cost-efficient for the reduction of nitrogen fertlizer application (Ju et al., 2016). For animal manure management, we used the information on the ammonia emission abatement technology in the GAINS model, including low nitrogen feed, low ammonia application, animal house adaption, bio-filtration, covered outdoor storage of manure, and their combination. Although they may not fully reflect the Chinese situation, clear potentials exist to further reduce NH_3 emissions. Animal manure management options that allow low emissions thus need to be further studied for situations typically encountered in China.

5 Conclusions

Overall, agricultural ammonia emissions were 1179 kt NH_3 in 2012 in HRB, with approximately 45% of the total agricultural NH_3 emissions coming from the six hotspots regions which covered only 17% of the basin area. This also means much higher ammonia emission densities occurred in these regions – from 7 to 12 t NH_3 km^{-2}. NH_3 emissions will further increase by 33% by 2030 and still with large regional uneven distribution, if there is no intervention. However, considerable emission reductions are possible and also benefit for air pollution abatement in terms of $PM_{2.5}$. The most effective strategy is the combination of improved technology and management with human diet optimization, which can reduce ammonia emissions by 60% and therefore 22-27% to 29-43% and 9-14% to 12-24% of the secondary inorganic aerosols and $PM_{2.5}$ concentration, respectively, relative to 2030 under the BAU scenario. This is a considerable potential to be achieved by ammonia emission controls.

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References

National Development and Reform Commission (NDRC), 2013. Cost and Income of Chinese Farm Produce (In Chinese). China Statistics Press: Beijing.

International Energy Agency (IEA): Energy and Air Pollution - World energy outlook Special Report. http://www.iea.org/publications/freepublications/publication/weo-2016-special-report-energy-and-air-pollution.html (acessed 20 January 2016).

Aikman, S. N., Min, K. E., and Graham, D., 2006. Food attitudes, eating behavior, and the information underlying food attitudes. Appetite. 47, 111-114. doi: 10.1016/j.appet.2006.02.004.

Bai, Z., Ma, L., Jin, S., Ma, W., Velthof, G. L., Oenema, O., Liu, L., Chadwick, D., and Zhang, F., 2016. Nitrogen, Phosphorus, and Potassium Flows through the Manure Management Chain in China. Environ. Sci. Technol. 50, 13409–13418. doi: 10.1021/acs.est.6b03348.

Beelen, R., Raaschou-Nielsen, O., Stafoggia, M., Andersen, Z. J., Weinmayr, G., Hoffmann, B., Wolf, K., Samoli, E., Fischer, P., Nieuwenhuijsen, M., Vineis, P., Xun, W. W., Katsouyanni, K., Dimakopoulou, K., Oudin, A., Forsberg, B., Modig, L., Havulinna, A. S., Lanki, T., Turunen, A., Oftedal, B., Nystad, W., Nafstad, P., Faire, U. D., Pedersen, N. L., Östenson, C.-G., Fratiglioni, L., Penell, J., Korek, M., Pershagen, G., Eriksen, K. T., Overvad, K., Ellermann, T., Eeftens, M., Peeters, P. H., Meliefste, K., Wang, M., Bueno-de-Mesquita, B., Sugiri, D., Krämer, U., Heinrich, J., Hoogh, K. d., Key, T., Peters, A., Hampel, R., Concin, H., Nagel, G., Ineichen, A., Schaffner, E., Probst-Hensch, N., Künzli, N., Schindler, C., Schikowski, T., Adam, M., Phuleria, H., Vilier, A., Clavel-Chapelon, F., Declercq, C., Grioni, S., Krogh, V., Tsai, M.-Y., Ricceri, F., Sacerdote, C., Galassi, C., Migliore, E., Ranzi, A., Cesaroni, G., Badaloni, C., Forastiere, F., Tamayo, I., Amiano, P., Dorronsoro, M., Katsoulis, M., Trichopoulou, A., Brunekreef, B., and Hoek, G., 2014. Effects of long-term exposure to air pollution on natural-cause mortality: an analysis of 22 European cohorts within the multicentre ESCAPE project. Lancet. 383, 785-795.

Bittman, S., Dedina, M., Howard, C. M., Oenema, O., and Sutton, M. A., 2014. Options for ammonia mitigation: Guidance from the UNECE Task Force on Reactive Nitrogen. NERC/Centre for Ecology & Hydrology.

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

Chadwick, D., Wei, J., Yan'an, T., Guanghui, Y., Qirong, S., and Qing, C., 2015. Improving manure nutrient management towards sustainable agricultural intensification in China. Agr. Ecosyst. Enviro. 209, 34-46. doi: 10.1016/j.agee.2015.03.025.

Chen, F., Yamashita, K., Kurokawa, J., and Klimont, Z., 2015. Cost–Benefit Analysis of Reducing Premature Mortality Caused by Exposure to Ozone and $PM_{2.5}$ in East Asia in 2020. Water Air Soil Poll. 226, 108. doi: 10.1007/s11270-015-2316-7.

Chen, X., Cui, Z., Fan, M., Vitousek, P., Zhao, M., Ma, W., Wang, Z., Zhang, W., Yan, X., Yang, J., Deng, X., Gao, Q., Zhang, Q., Guo, S., Ren, J., Li, S., Ye, Y., Wang, Z., Huang, J., Tang, Q., Sun, Y., Peng, X., Zhang, J., He, M., Zhu, Y., Xue, J., Wang, G., Wu, L., An, N., Wu, L., Ma, L., Zhang, W., and Zhang, F., 2014. Producing more grain with lower environmental costs. Nature. 514, 486-489. doi: 10.1038/nature13609.

Chinese Nutrition Society, 2016. The Food Guide Pagoda for Chinese Residents. Standards Press of China, Beijing (In Chinese).

European Environment Agency (EEA), 2013. Air pollutant emission inventory guidebook 2013. Copenhagen.

Food and Agriculture Organization (FAO), 2016. Food and Agriculture Data. http://faostat.fao.org/site/291/default.aspx (accessed 20 December 2016).

Garnett, T., Appleby, M. C., Balmford, A., Bateman, I. J., Benton, T. G., Bloomer, P., Burlingame, B., Dawkins, M., Dolan, L., Fraser, D., Herrero, M., Hoffmann, I., Smith, P., Thornton, P. K., Toulmin, C., Vermeulen, S. J., and Godfray, H. C. J., 2013. Sustainable Intensification in Agriculture: Premises and Policies. Science. 341, 33-34.

Gu, B., Ju, X., Chang, J., Ge, Y., and Vitousek, P. M., 2015. Integrated reactive nitrogen budgets and future trends in China. Proc. Natl. Acad. Sci. USA. 112, 8792–8797. doi: 10.1073/pnas.1510211112.

Huang, X., Song, Y., Li, M., Li, J., Huo, Q., Cai, X., Zhu, T., Hu, M., and Zhang, H., 2012. A high-resolution ammonia emission inventory in China. Global Biogeochem. Cy. 26, 2-14. doi: 10.1029/2011gb004161.

Kang, Y., Liu, M., Song, Y., Huang, X., Yao, H., Cai, X., Zhang, H., Kang, L., Liu, X., Yan, X., He, H., Zhang, Q., Shao, M., and Zhu, T., 2016. High-resolution ammonia emissions inventories in China from 1980 to 2012. Atmos. Chem. Phys. 16, 2043-2058. doi: 10.5194/acp-16-2043-2016.

Kiesewetter, G., Schoepp, W., Heyes, C., and Amann, M., 2015. Modelling $PM_{2.5}$ impact indicators in Europe: Health effects and legal compliance. Environ. Modell. Softw. 74, 201-211. doi: 10.1016/j.envsoft.2015.02.022.

Klimont, Z., Kupiainen, K., Heyes, C., Purohit, P., Cofala, J., Rafaj, P., Borken-Kleefeld, J., and Schöpp, W., 2016. Global anthropogenic emissions of particulate matter including black carbon. Atmos. Chem. Phys. Discuss. doi: 10.5194/acp-2016-880.

Li, S., and Ma, S., 1993. Ammonia volatilization from calcareous soil II. The relationship between ammonia loss from N fertilizers and applied methods (In Chinese). Agricultural Research in the Arid Areas. 11, 130-134.

Ma, L., Wang, F., Zhang, W., Ma, W., Velthof, G., Qin, W., Oenema, O., and Zhang, F., 2013. Environmental assessment of management options for nutrient flows in the food chain in China. Environ. Sci. Technol. 47, 7260-7268. doi: 10.1021/es400456u.

Ma, L., Bai, Z., Ma, W., Guo, M., Jiang, R., Liu, J., Oenema, O., Velthof, G. L., Whitmore, A. P., Crawford, J., Dobermann, A., Schwoob, M. H., and Zhang, F., 2016. Assessing pathways to sustainable food production and consumption. Submitted to Global Environ. Chang.

Oxley, T., Dore, A. J., ApSimon, H., Hall, J., and Kryza, M., 2013. Modelling future impacts of air pollution using the multi-scale UK Integrated Assessment Model (UKIAM). Environ. Int. 61, 17-35. doi: 10.1016/j.envint.2013.09.009.

Pope, C. A., Burnett, R. T., Thun, M. J., Calle, E. E., Krewski, D., Ito, K., and Thurston, G. D., 2002. Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. Jama. 287, 1132-1141. doi:10.1001/jama.287.9.1132.

Popkin, B. M., Horton, S., Kim, S., Mahal, A., and Jin, S., 2001. Trends in diet, nutritional status, and diet-related noncommunicable diseases in China and India: The economic costs of the nutrition transition. Nutr. Rev. 59, 379-390. doi: https://doi.org/10.1111/j.1753-4887.2001.tb06967.x.

Rao, S., Klimont, Z., Smith, S. J., Van Dingenen, R., Dentener, F., Bouwman, L., Riahi, K., Amann, M., Bodirsky, B. L., van Vuuren, D. P., Reis, L. A., Calvin, K., Drouet, L., Fricko, O., Fujimori, S., Gernaat, D., Havlik, P., Harmsen, M., Hasegawa, T., Heyes, C., Hilaire, J., Luderer, G., Masui, T., Stehfest, E., Strefler, J., van der Sluis, S., and Tavoni, M., 2017. Future air pollution in the Shared Socio-economic Pathways. Global Environ. Chang. 42, 346-358. doi: 10.1016/j.gloenvcha.2016.05.012.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J. P., Valdebenito, Á., and Wind, P., 2012. The EMEP MSC-W chemical transport model-technical description. Atmos. Chem. Phys. 12, 7825-7865. doi: 10.5194/acp-12-7825-2012.

Sun, Y. L., Wang, Z. F., Du, W., Zhang, Q., Wang, Q. Q., Fu, P. Q., Pan, X. L., Li, J., Jayne, J., and Worsnop, D. R., 2015. Long-term real-time measurements of aerosol particle composition in Beijing, China: seasonal variations, meteorological effects, and source analysis. Atmos. Chem. Phys. 15, 10149-10165. doi: 10.5194/acp-15-10149-2015.

Tilman, D., and Clark, M., 2014. Global diets link environmental sustainability and human health. Nature. 515, 518-522. doi: 10.1038/nature13959.

United Nations, 2015. World Population Prospects, New York City, USA. http://esa.un.org/unpd/wpp/ (accessed on 8 November 2016).

Van Donkelaar, A., Martin, R. V., Brauer, M., Kahn, R., Levy, R., Verduzco, C., and Villeneuve, P. J., 2010. Global estimates of ambient fine particulate matter concentrations from satellite-based aerosol optical depth: development and application. Environ. Health Pers. 118, 847-855. doi: 10.1289/ehp.0901623.

Wang, D., Stewart, D., Chang, C., and Shi, Y., 2015. Effect of a school-based nutrition education program on adolescents' nutrition-related knowledge, attitudes and behaviour in rural areas of China. Environ. Health Prev. Med. 20, 271-278. doi: 10.1007/s12199-015-0456-4.

Wang, Y., Zhang, Q. Q., He, K., Zhang, Q., and Chai, L., 2013. Sulfate-nitrate-ammonium aerosols over China: response to 2000–2015 emission changes of sulfur dioxide, nitrogen oxides, and ammonia. Atmos. Chem. Phys. 13, 2635-2652. doi: 10.5194/acp-13-2635-2013.

Wu, Y., Gu, B., Erisman, J. W., Reis, S., Fang, Y., Lu, X., and Zhang, X., 2016. $PM_{2.5}$ pollution is substantially affected by ammonia emissions in China. Environ. Pollut. 218, 86-94. doi: 10.1016/j.envpol.2016.08.027.

Xu, P., Liao, Y. J., Lin, Y. H., Zhao, C. X., Yan, C. H., Cao, M. N., Wang, G. S., and Luan, S. J., 2016. High-resolution inventory of ammonia emissions from agricultural fertilizer in China from 1978 to 2008. Atmos. Chem. Phys. 16, 1207-1218. doi: 10.5194/acp-16-1207-2016.

Zhang, S. L., Cai, G. X., Wang, X. Z., H., X. Y., Zhu, Z. L., and Freney, J. R., 1992. Losses of urea-nitrogen applied to maize grown on a calcareous fluvo-aquic soil in North China Plain. Pedosphere. 2, 171-178.

Zhang, W., Cao, G., Li, X., Zhang, H., Wang, C., Liu, Q., Chen, X., Cui, Z., Shen, J., Jiang, R., Mi, G., Miao, Y., Zhang, F., and Dou, Z., 2016. Closing yield gaps in China by empowering smallholder farmers. Nature. 537, 671-674. doi: 10.1038/nature19368.

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

Zhou, Y., Shuiyuan, C., Lang, J., Chen, D., Zhao, B., Liu, C., Xu, R., and Li, T., 2015. A comprehensive ammonia emission inventory with high-resolution and its evaluation in the Beijing–Tianjin–Hebei (BTH) region, China. Atmos. Environ. 106, 305-317. doi: 10.1016/j.atmosenv.2015.01.069.

MOA, 2015. Zero Growth in Synthetic Fertilizer Use from 2020 Onwards (in Chinese).

Ministry of Agriculture of the People's Republic of China.

GB 3095-2012, 2012. Ambient Air Quality Standard. National Standard of the People’s Republic of China.

Chen, X., Cui, Z., Vitousek, P. M., Gassman, K. G., Matson, P. A., Bai, J., Meng, Q., Hou, P., Yue, S., R ömheld, V., Zhang, F., 2011. Integrated soil-crop system management for food security. Proc Natl Acad Sci USA. 108:6399-6404.

Ju, X., Gu, B., Wu, Y., Galloway, JN., 2016. Reducing China’s fertilizer use by increasing farm size. Global Environ Chang. 41: 26-32.