

Reply to Reviewer's Comments

We would like to thank the editor and two anonymous reviewers for their comments to help improve our manuscript. Below we give a point-to-point response to address the reviewers' comments. The original comments are in red and our responses are in black.

Comments from Anonymous Referee #1

General Comments:

This is an interesting and well-written manuscript describing the development of a higher spatial resolution emission-factor based ammonia inventory. I particularly appreciated the policy-focused trend discussion.

Response: We highly appreciated the referee's positive comments, and the recognition of the importance of our work.

This is a potentially important contribution for air quality modeling in China, but beyond comparison to previous inventories, it is missing discussions of the validity of many of the assumptions needed for its development beyond "it is what was done previously." It is also lacking comparison of emission estimates to observations, but adequate data sets may not be available.

Response: We thank the reviewer for the comment. In order to improve the performance of air quality modeling in China, a timely updated emission inventory with temporal and spatial variations is crucial (Skjott et al., 2011). In this study, a new Chinese agricultural fertilizer NH₃ emissions inventory has been compiled. However, it must be pointed out that currently, under the circumstances of lacking accurately activity data, detailed agricultural registers and practices, reliable input parameters, and field NH₃ measurement data in the air quality monitoring network widely existed in China, sound high resolution NH₃ inventory is hard to develop (Zheng et al., 2012). In this study, we have to adopt some assumptions which mainly referred to expert judgments or other similar studies, and we added the discussions of the validity of some of the assumptions in the revised version (Details are described in our responses to the Detailed Comments and Technical Comments below). More work should be done in the future in order to more accurately estimate the agriculture fertilizer NH₃ emissions in China.

In addition, in the revised version we added the temporal distribution comparison between observation and our study (Lines 318-319 on Page 12 in the revised version) (Ianniello et al., 2010; Meng et al., 2011). In the revised version, we also did some further discussions based upon other studies (Clarisse et al., 2009; Van Damme et al., 2014; Shephard et al., 2011; Van Damme et al., 2015) about the satellite measurement in order to evaluate the spatial and temporal distribution of NH₃ emissions inventory, such as the Tropospheric Emission Spectrometer (TES), Infrared Atmospheric Sounding Interferometer (IASI), and have revised the manuscript by referring to the results (See Lines 264-281 on Page 10 and Lines 319-323 on Page 12 in the revised version).

Lines 264-281 on Page 10 in the revised version: "We compared our results with the global NH₃ column distribution using satellite monitoring from the Infrared Atmospheric Sounding Interferometer (IASI) (Clarisse et al., 2009; Van Damme et al., 2014). Several emissions hotspots are observed in the Tarim basin, the North China Plain and western Heilongjiang province and

Jilin province by the IASI sensor, emission density is 4.2, 7.4, 5.2 and 9.6 tNH₃ km⁻² yr⁻¹, respectively. This result demonstrated excellent qualitative consistent with our estimated emissions. However, the higher emission areas were not observed by satellite monitoring because of clouds, water vapor, the surface temperature, high SO₂ emissions (Kharol et al., 2013; Wang et al., 2013; Garcia et al., 2008), land surface variation and the retrieval methods of NH₃ total columns (Xu et al., 2015). Higher plenty of cloud cover and precipitation could generate some uncertainties in the Sichuan Basin and Lianghu Plain. Additionally, NH₃ concentration distribution might not be always agreement with emission pattern due to its high reactive ability, solubility and short-lived in the atmosphere (Huang et al., 2012); Higher surface temperature and humidity could speed up NH₃ consumption simultaneously. And Sichuan Basin is high SO₂ pollution district where NH₃ gas could easily react with SO₂ (Zhang et al., 2009). Therefore, these factors lead the inconsistent between satellite monitoring and our inventory.”

Lines 318-319 on Page 12 in the revised version: “These higher emissions are virtually identical with several in situ data sets (Ianniello et al., 2010; Meng et al., 2011).”

Lines 319-323 on Page 12 in the revised version: “In addition, the seasonality of emissions from IASI and Tropospheric Emission Spectrometer (TES) satellite observations demonstrated excellent consistent with the temporal distribution of our inventory which is a summer maximum of NH₃ emissions in China (Shephard et al., 2011; Van Damme et al., 2015).”

The article listed below may not have been available at the time this draft was first written, but I think it is important to include some discussion of this alternative method as it compares to the one in this paper. What are the strengths and weakness of the two methods? Are there situations where one method should be preferred over the other? I am particularly interested in the authors’ thoughts on the implications of bidirectional ammonia flux on their inventory estimates.

Response: We thank the reviewer for the suggestion. We strongly agreed with referee’s suggestion that it is important to include some discussion of the bi-directional flux of NH₃ estimation method as it compares to the one in this paper. However, our study does not include bi-directional exchange, and more work should be done in the future. In order to make the readers to understand this research better, in the revised version, we added the description for bidirectional exchange of NH₃ and the comparison with Fu et al. (2015) and Zhu et al. (2015) (See Lines 86-111 on Page 4 in the revised version), and the suggestion of our inventory for the future work (See Lines 497-500 on Page 18 in the revised version).

Lines 86-111 on Page 4 in the revised version: “In addition, studies have attempted to focus on implementing the bidirectional exchange of NH₃ in many air quality models, e.g., the Community Multi-scale Air Quality (CMAQ) model (Cooter et al., 2012; Bash et al., 2013; Pleim et al., 2013; Fu et al., 2015), and the GEOS-Chem global chemical transport model (Zhu et al., 2015). NH₃ deposition, emission, reemission, and atmospheric lifetime can be affected by rigorous treatment of the bidirectional flux of NH₃, and vegetation and soil can be either a sink or a source of atmospheric NH₃ (Sutton et al., 2007). Fu et al. (2015) provides the first online estimate of NH₃ emissions from agricultural fertilizer use China, based on coupling the CMAQ model with a bi-directional NH₃ exchange module and the Environmental Policy Integrated Climate (EPIC) model, this method considers an increased number of influencing factors, such as meteorological fields, soil and fertilizer application, and provides improved NH₃ emissions with higher spatial

and temporal resolution, whereas, gaps still exist for this method owing to the uncertainties of more model parameterization and input data. Zhu et al. (2015) developed the adjoint of bidirectional exchange in GEOS-Chem model which suggests that although the implementing bidirectional exchange greatly extends the lifetime of NH_3 in the atmosphere via deposition and reemission processes and conducts a better fundamental description of NH_3 emissions from fertilizers, whereas, it does not uniformly ameliorate estimation of NH_3 concentrations, NH_4^+ wet deposition and nitrate aerosol concentrations due to the NH_3 re-emissions from the ammonium soil pool that accumulates ammonium from previous months or the ammonium soil pool preserves ammonia/ammonium in the soil rather than emitting it directly after fertilizer application during the growing seasons (e.g., bidirectional exchange significant decreases NH_3 gross emissions in southeastern China and NH_3 concentrations in China in April of 2008, but changes in NH_4^+ wet deposition are not very large in April).”

Lines 497-500 on Page 18 in the revised version: “Our high resolution inventory can be applied to simulate atmospheric aerosol formation in air quality models with a bidirectional NH_3 exchange module, and then explore the effects of NH_3 emissions on China’s urban air pollution.”

I recommend publication with this addition and consideration of the detailed comments that follow.

Response: We thank the referee for favorable considerations of our manuscript. We have made major revisions as suggested by the referee, and details are described in our responses to the Detailed Comments and Technical Comments below.

Detailed Comments:

1. I suggest shortening the title to “High resolution inventory of ammonia emissions from agricultural fertilizer in China from 1978 to 2008”

Response: Followed the reviewer’s comment, the title has been changed into “High resolution inventory of ammonia emissions from agricultural fertilizer in China from 1978 to 2008”.

2. Pg 25304, line 5. Please describe “straw returning” as an emission source.

Response: We thank the reviewer for the suggestion. We added an explanation in lines 131-133 on Page 5.

Lines 131-133 on Page 5 in the revised version: “In this study, straw returning represents crop residue compost which adds soil nutrients in the rural China, and NH_3 is released during composting through aerobic and anaerobic microbial processes.”

3. Pg 25303, line 13. Does the function "f(.)" maintain a constant, climatological shape through time for each region, or does it respond to interannual weather variability? Please add to text.

Response: We apologize for our indistinct description of f(). We revised in lines 142-144 on Page 6 in the revised version: “f() represents a function whose shape depends on the source type, which responds to RP_{nijm} and C ”.

4. Pg 25304, line 3. “We obtained the 2008 unavailable fertilizer, crop. . .” The sentence needs

clarification. How can you obtain unavailable data?

Response: We apologize that the description about this sentence is not clear. We revised this sentence in lines 158-161 on Page 6 “We obtained the 2008 unavailable fertilizer, crop, and livestock data for 332 counties in Mainland China and Taiwanese annual above-province-level data for 1978-1990 based on temporal interpolation (Zhou et al., 2014).”.

5. Pg 25306, line 3. Is “the CV of each activity data in 1978-2007 is equal to the CVs of the 2008 data” a good/valid assumption?

Response: In this study, we think “the CV of each activity data in 1978-2007 is equal to the CVs of the 2008 data” is a *good* assumption. Because the coefficient of variation (CV) of each activity data is assumed to be equal to the absolute value of the average difference between a given dataset for China used in to determine CAF_NH₃ and a default global dataset (e.g., IFA, FAO, World Bank) for 2008 which refers to Zhou et al. (2014), however, we cannot adopt the uniform method to obtain the CV of each activity data in 1978-2007 due to the limit of data availability. The CV of each activity data in 1978-2007 is equal to the CVs of the 2008 data based on expert judgments, which is reasonable and acceptable although uncertainties still exist. In addition, we further described the source of this assumption in Lines 220-222 on Page 8 “In addition, the CV of each activity data in 1978-2007 is assumed to be equal to the CVs of the 2008 data based on expert judgments.”

6. Pg 2506, line 9. Did you test the activity data to see if the normality assumption is valid?

Response: We thank the reviewer for the comment. We are sorry that we did not test the activity data to see if the normality assumption is valid. In this study, uniform distributions were assumed for the CV of each activity data which refers to Zhou et al. (2014) and Wang et al. (2012).

7. Pg 2506, line 20. The use of ammonium bicarbonate (ABC) is interesting as regards Green House Gas (GHG) production since it breaks down into CO₂, water and NH₃. You mention that China is moving away from this form of synthetic fertilizer. Could that result in a CO₂ emission decrease? Less nitrogen application should result in reduced N₂O emission (another GHG), but also could reduce a potential rural source of NO which might also affect ozone concentrations. Many possible nitrogen emission and air quality interactions are linked to fertilizer use. Will your inventory be able to respond to changes in these factors in a physically realistic manner? Will you be able to untangle the various interactions? Can you use this inventory for future projections?

Response: We thank the reviewer for the comment. We completely agreed that the use of ammonium bicarbonate (ABC) is interesting as regards Green House Gas (GHG) production since it breaks down into CO₂, water and NH₃. In this study, we mentioned that China is moving away from this form of synthetic fertilizer. However, synthetic fertilizer has been and will continue to be indispensable for China’s quest to produce sufficient food to meet its growing demands (Zhang et al., 2013). The industrial production and transport of synthetic fertilizer release abundant CO₂ into the atmosphere. Therefore, there is no doubt that cutting synthetic fertilizer application where available nitrogen exceeds crop requirements can decrease CO₂ emission (Huang et al., 2010). China needs a combination of reforms in the synthetic fertilizer industry and changes in management practices and technologies at the farm level to minimize excessive nitrogen use in the field (Kahrl et al., 2010; Zhang et al., 2013).

In addition, we also completely agreed that less nitrogen application should result in reduced N₂O emission (another GHG), but also could reduce a potential rural source of NO which might also affect ozone concentrations. Synthetic nitrogen fertilizer played a major role in N₂O emission from agricultural soil, with a linear or exponential effect of synthetic fertilizer rate on N₂O emission (Bouwman et al., 2002), which contributes 23% of the total N₂O emission in Zhou et al. (2014). N₂O is the largest stratospheric ozone-depleting substance through the formation of NO and is projected to remain so for the remainder of this century (Ravishankara et al., 2009). Reducing N₂O emissions offers the combined benefits of mitigating climate change and protecting the ozone layer (Kanter et al., 2013; Li et al., 2014).

Many possible nitrogen emission and air quality interactions are linked to fertilizer use. In this study, the NH₃ total emissions will alter when the types of fertilizer application change and the corresponding NH₃ emission factors also follow change. Moreover, we have described about NO_x (NO_x=NO₂+NO) and NH₃ emission and air quality interactions in the section 4.2 Impacts of NH₃ Emissions on Urban Air pollution. So our inventory can be able to complete these work. In spite of the large NH₃ emissions in China, some studies, and the methods similar with us, have been conducted to assess the mitigation potentials (Klimont et al., 2011; Amann et al., 2013; Sejian et al., 2015; Shen et al., 2014), therefore we believe that we can use this inventory for future projections, and this is also what we are trying to do in the future.

8. Pg 25312, section 4.1. Nice comparison to previous inventories. Is your method “better” or just different from the others? Do you have evidence that the higher spatial resolution is giving you more valid information as opposed to a larger number of poor estimates? Comparison of your emission estimates with satellite data is a good start, but is not sufficient because of the limitations you mention. Are there ground-based flux measurements available for any locations that you could compare with your source inventory? If not, would that be a good recommendation for future government investment?

Response: We thank the reviewer for the comment and suggestion. In this study, we only expressed that our method is just different from the others in section 4.1. We apologize that the purpose description about comparison to previous inventories is not clear. We revised the section 4.1 “Differences with previous NH₃ emissions inventories”.

In addition, to test the sensitivity of the NH₃ emissions spatial patterns to input activity data (high resolution activity data), an emissions inventory (PRO-NH₃(China)) was developed using the same methods that were employed to create CAF_NH₃ except county-level activity data for provincial disaggregation using regression models (Zhang et al., 2007; Zhou et al., 2014). This section can give us some evidence that higher spatial resolution is giving us more valid information as opposed to a larger number of poor estimates. Please see the revised manuscript in Lines 282-301 on Pages 10-11.

We totally agreed with the referee’s suggestion about the validation of our inventory by using ammonia ground-based flux measurements, and in fact this is the best way, ideally. Although the observation data for ammonia flux was very spare and not publicly available in China as the referee said, we added the temporal distribution comparison between observation and our study (Lines 318-319 on Page 12 in the revised version) (Ianniello et al., 2010; Meng et al., 2011). Additionally, in the revised version, we also did some further discussions based upon other studies

(Clarisse et al., 2009; Van Damme et al., 2014; Shephard et al., 2011; Van Damme et al., 2015) about the satellite measurement in order to evaluate the spatial and temporal distribution of NH₃ emissions inventory, such as the Tropospheric Emission Spectrometer (TES), Infrared Atmospheric Sounding Interferometer (IASI), and have revised the manuscript by referring to the results (See Lines 264-281 on Page 10 and Lines 319-323 on Page 12 in the revised version). We also suggest that the local measured NH₃ should be conducted in the future government investment (See Line 531 on Page 19 in the revised version).

Lines 282-301 on Pages 10-11 in the revised version: “To test the sensitivity of the NH₃ emissions spatial patterns to input activity data, an emissions inventory (PRO-NH₃(China)) was developed using the same methods that were employed to create CAF_NH₃ except county-level activity data for provincial disaggregation using regression models (Zhang et al., 2007; Zhou et al., 2014). The E_{total} of PRO-NH₃ is 7.3 TgNH₃ yr⁻¹, which is 12.5% less than the CAF_NH₃ value. For a more detailed comparison, the relative difference was defined as $RD = (E_1 - E_2) / ((E_1 + E_2)/2)$ (Wang et al., 2012), where E_1 and E_2 are the E_{total} for agricultural fertilizer of the counties for CAF_NH₃ and for PRO-NH₃ for each county, respectively. Fig. 3 shows all counties’ frequency and spatial distributions of the RDs. The spatial bias of the provincial disaggregation increases as the absolute RDs. A negative (positive) RD suggests an overestimation (underestimation) of a county’s emissions by utilizing the provincial disaggregation approach (PRO-NH₃). The mean absolute RD was 48.7% for all counties. In 37% of the countries, the absolute RDs were found higher than 50%. In addition, the PRO-NH₃ emission pattern is lowly correlated with the CAF-NH₃ pattern ($R = 0.49$, $p < 0.01$). These results indicate that spatial bias can be substantially reduced using the county-level activity data and that provincial disaggregation using regression models cannot determine the county-scale structure of the spatial distribution of activity data within provinces. Large RDs were often observed in provinces and regions in which the development status significantly varies, such as Sichuan, Qinghai, Inner Mongolia and Tibet.”

Lines 264-281 on Page 10 in the revised version: “We compared our results with the global NH₃ column distribution using satellite monitoring from the Infrared Atmospheric Sounding Interferometer (IASI) (Clarisse et al., 2009; Van Damme et al., 2014). Several emissions hotspots are observed in the Tarim basin, the North China Plain and western Heilongjiang province and Jilin province by the IASI sensor, emission density is 4.2, 7.4, 5.2 and 9.6 tNH₃ km⁻² yr⁻¹, respectively. This result demonstrated excellent qualitative consistent with our estimated emissions. However, the higher emission areas were not observed by satellite monitoring because of clouds, water vapor, the surface temperature, high SO₂ emissions (Kharol et al., 2013; Wang et al., 2013; Garcia et al., 2008), land surface variation and the retrieval methods of NH₃ total columns (Xu et al., 2015). Higher plenty of cloud cover and precipitation could generate some uncertainties in the Sichuan Basin and Lianghu Plain. Additionally, NH₃ concentration distribution might not be always agreement with emission pattern due to its high reactive ability, solubility and short-lived in the atmosphere (Huang et al., 2012); Higher surface temperature and humidity could speed up NH₃ consumption simultaneously. And Sichuan Basin is high SO₂ pollution district where NH₃ gas could easily react with SO₂ (Zhang et al., 2009). Therefore, these factors lead the inconsistent between satellite monitoring and our inventory.”

Lines 318-319 on Page 12 in the revised version: “These higher emissions are virtually identical with several in situ data sets (Ianniello et al., 2010; Meng et al., 2011).”

Lines 319-323 on Page 12 in the revised version: “In addition, the seasonality of emissions from IASI and Tropospheric Emission Spectrometer (TES) satellite observations demonstrated excellent consistent with the temporal distribution of our inventory which is a summer maximum of NH₃ emissions in China (Shephard et al., 2011; Van Damme et al., 2015).”

Line 531 on Page 19: “Therefore, they should be implemented in the future research.”

9. Pg 25314, line 20. Please clarify this sentence. What do you mean by “..replaced by compound and organic..” Isn't ABC a compound fertilizer?

Response: We thank the reviewer for the comment. We apologize that the description about this sentence is not clear. In this sentence, compound fertilizer represents compound nitrogen-phosphorous-potassium fertilizer. We revised this sentence in the revised version in Lines 511-514 on Page 19.

Lines 511-514 on Page 19 in the revised version: “Because of their high volatility, urea and ABC have been gradually replaced by compound nitrogen-phosphorous-potassium and organic fertilizers in the wake of the country attaching greater importance to the food security problem.”

10. Does the current inventory need further improvement, i.e., future direction of inventory research? Is there a need for field campaigns in China? I think you make a very valid point that using research findings based on European farming systems is not necessarily what is needed to understand agricultural ammonia emissions from Chinese crop and livestock management systems.

Response: We thank the reviewer for the comment. We think that the current inventory need further improvement, because our inventory still exist several uncertainties especially in the emissions from synthetic fertilizer application and livestock manure spreading due to the exceedingly high values and large amount of parameters related to the emission factors adjustment. It has been demonstrated that a dependable data-driven approach and local experiments or process-based models can substantially help increase the spatial and temporal resolution and decrease the uncertainties of emissions inventories. Therefore, they should be implemented in the future research. We added the description about this comment in Lines 525-531 on Page 19 in the revised version.

Lines 525-531 on Page 19 in the revised version: “Nevertheless, our inventory still exist several uncertainties especially in the emissions from synthetic fertilizer application and livestock manure spreading due to the exceedingly high values and large amount of parameters related to the emission factors adjustment. It has been demonstrated that a dependable data-driven approach and local experiments or process-based models can substantially help increase the spatial and temporal resolution and decrease the uncertainties of emissions inventories. Therefore, they should be implemented in the future research.”

Technical Comments:

1. Figure 2, Use the same scale and font size on figure A and B legends. Larger is better.

Response: We thank the reviewer for the comment. Figure 2 has been revised based on the

comments, as shown in the following figure.

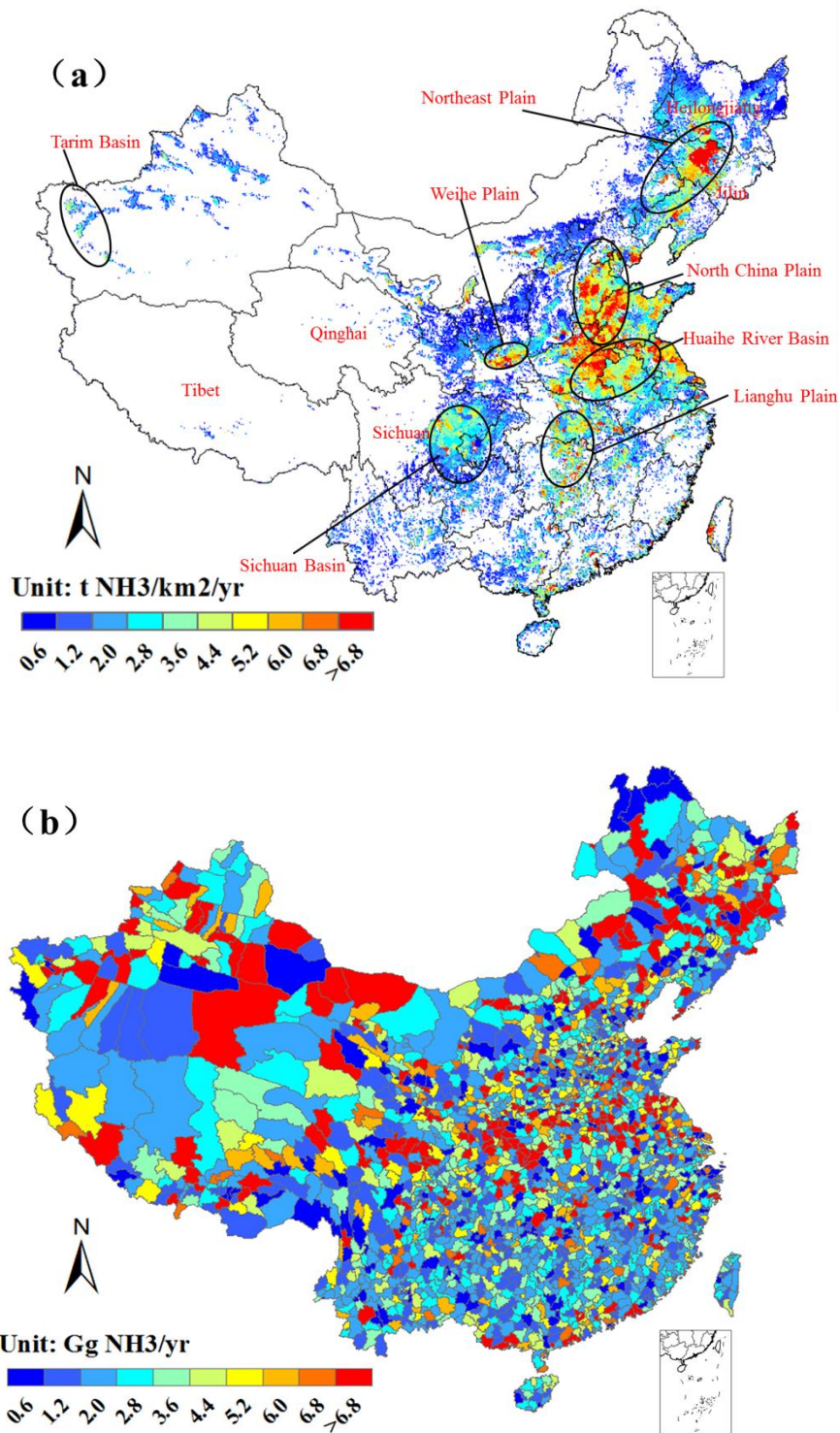


Figure.2 NH₃ emission map of China's agricultural fertilizer at 1 km ×1 km (a) and the county level (b) for 2008. Major emission areas are circled.

2. Figure 3, Define "RDs" in caption.

Response: We thank the reviewer for the comment. Lines 800-804 on Page 31 in the revised

version, we defined “RDs” in caption in Figure.

Lines 800-804 on Page 31 in the revised version: “ $RD = (E_1 - E_2) / ((E_1 + E_2)/2)$, where E_1 and E_2 are the E_{total} for agricultural fertilizer of the counties for CAF-NH₃ and for PRO-NH₃ for each county, respectively. A negative (positive) RD suggests an overestimation (underestimation) of a county’s emissions by utilizing the provincial disaggregation approach (PRO-NH₃).”

3. Figure 4. "Monthly NH₃ emissions in 2008 as compared with previous inventories"? Are these field studies or previous inventories for China? In caption, please include the date for the reference. There are many Zhang et al’s and Huang et al’s in your references.

Response: We thank the reviewer for the comment. We apologize that the date description about this reference is not clear here. These are previous inventories for China. In Figure 4, we added the date for the reference in caption. Details as follows Figure 4:

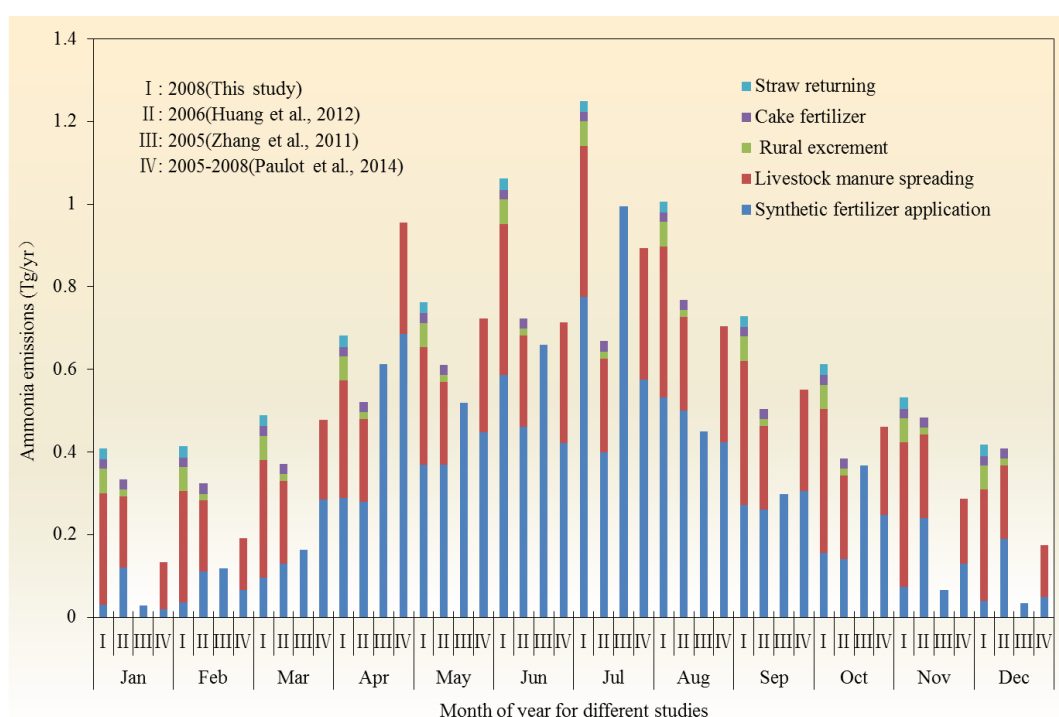


Figure.4 Monthly NH₃ emissions in 2008 and compared with earlier studies.

4. Figure 5. Please define R₅₀ either in the text or in the caption.

Response: We thank the reviewer for the comment. We added the definition R₅₀ in the text (See Lines 228-230 on Page 9 in the revised version).

Lines 228-230 on Page 9 in the revised version: “Medians and the R₅₀ (difference between the 75th and 25th quartiles) were aimed at estimating the emissions and representing the uncertainties.”

Reference:

Amann, M., Klimont, Z., and Wagner, F.: Regional and Global Emissions of Air Pollutants: Recent Trends and Future Scenarios, *Annu. Rev. Env. Resour.*, 38, 31-55, 10.1146/annurev-environ-052912-173303, 2013.

Bash, J. O., Cooter, E. J., Dennis, R. L., Walker, J. T., and Pleim, J. E.: Evaluation of a regional air-quality model with bidirectional NH₃ exchange coupled to an agroecosystem model, *Biogeosciences*, 10, 1635-1645, 10.5194/bg-10-1635-2013, 2013.

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Clarisse, L., Clerbaux, C., Dentener, F., Hurtmans, D., and Coheur, P. F.: Global ammonia distribution derived from infrared satellite observations, *Nat. Geosci.*, 2, 479-483, 2009.

Cooter, E. J., Bash, J. O., Walker, J. T., Jones, M. R., and Robarge, W.: Estimation of NH₃ bi-directional flux from managed agricultural soils, *Atmos. Environ.*, 44, 2107-2115, 10.1016/j.atmosenv.2010.02.044, 2010.

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Kanter, D., Mauzerall, D. L., Ravishankara, A. R., Daniel, J. S., Portmann, R. W., Grabel, P. M., Moomaw, W. R., and Galloway, J. N.: A post-Kyoto partner: Considering the stratospheric ozone regime as a tool to manage nitrous oxide, *P. Natl. Acad. Sci. USA.*, 110, 4451-4457, 10.1073/pnas.1222231110, 2013.

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Comments from Anonymous Referee #2

General Comments:

This is a very interesting manuscript that develops a new fertilizer NH₃ emission inventory of China (CAF_NH₃) and also reports the temporal trends for 1978-2008. The authors explain a detail of the relationship between the NH₃ emission patterns and government policies, which is an important factor of developing a practical and precise NH₃ emission inventory of China. I really like this section.

Response: We highly appreciated the referee's positive comments, and the recognition of the importance of our work.

I have three main comments. 1) A high resolution NH₃ emission inventory based on the county-level activity data is developed for 2008. While the NH₃ emission inventory for 1978-2007 is based on the province-level activity data. In the manuscript, a sensitivity test of NH₃ emission spatial patterns to input of activity data demonstrates that the province-level activity data could bias the total emission and the spatial variation. Then, how much confidence could we have on the results of 1978-2007 that based on the province-level activity data? This point should be well explained in the text.

Response: We thank the reviewer for the suggestion. In this study, the NH₃ emissions based on province-level activity data from 1978 to 2007 were estimated and the uncertainties were quantified for the inventories by using the Monte Carlo simulation. Medians and the R₅₀ (difference between the 75th and 25th quartiles) were aimed at estimating the emissions and representing the uncertainties. In addition, we added an explanation that why we choose the province-level activity data from 1978 to 2007 to develop this inventories (See Lines 302-314 on Page 11 in the revised version).

Lines 302-314 on Page 11 in the revised version: “By comparing nitrogen fertilizer, compound fertilizer, rural population, rice, wheat, maize, cattle, sheep and pigs activity data (1978-2007) which are the major NH₃ emission sources in this study from NBSC provincial statistics (sums of the provincial data), IFA and FAO (national data), it was found that 64.8% IFA and FAO statistics underestimated the above activity data from 1978 to 2007 because of the difference statistical criteria, especially rural population and sheep attained 100% and 80.0% respectively (Fig. S3). The possible underestimation of national emission statistics has been demonstrated by NH₃ emission trends based on the per capita livestock that can cover all the NH₃ emission during the whole lifespan of livestock in Gu et al. (2012), and this fact may support our conclusion. Considering the information presented here and the limit of county-level activity data availability in 1978-2007, province-level activity data from 1978 to 2007 was used in our study in order to develop high resolution inventory.”

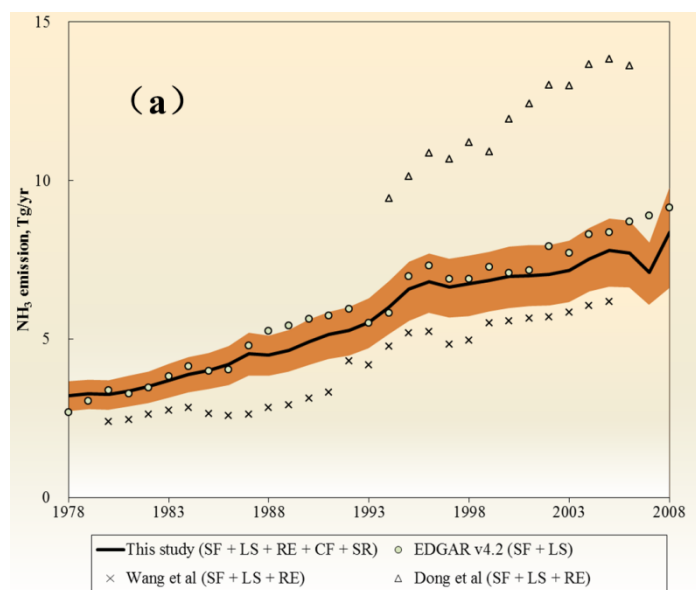


Figure.5 Temporal trend for agricultural fertilizer NH_3 emissions for China (a). The emission estimate and the uncertainty are provided as a median value (black curve) and the R_{50} (shaded area, for total emissions) derived from a Monte Carlo simulation. Note: SF = Synthetic fertilizer application; LS = Livestock manure spreading; RE = Rural excrement; CF = Cake fertilizer; SR = Straw returning.

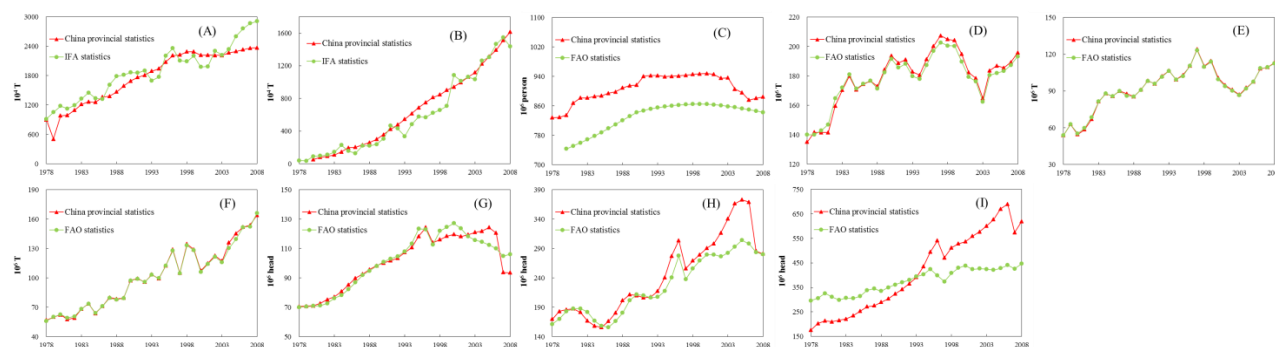


Figure S3 Comparison of nitrogen fertilizer (A), compound fertilizer (B), rural population (C), rice (D), wheat (E), maize (F), cattle (G), sheep (H) and pigs (I) activity data for a period from 1978 to 2007 which are the major NH_3 emission sources in this study from NBSC (2009a and 2009b) provincial statistics (sums of the provincial data), IFA and FAO (national data) (<http://www.fertilizer.org/> and <http://faostat.fao.org/>).

2) The bi-directional flux of NH_3 hasn't been discussed in the manuscript. Zhu et al. (2015, acpd) shows there are large impacts on NH_3 emissions in eastern China when including the bi-directional exchange processes in the model. I think including the bi-directional flux in this emission inventory could be a great improvement. Although the authors may not be able to finish the work in this manuscript, it should be discussed at least.

<http://www.atmoschem-phys-discuss.net/15/4823/2015/acpd-15-4823-2015.html>

Response: We thank the reviewer for the suggestion. We totally agreed with referee's suggestion that it is important to include some discussion of the bi-directional flux of NH_3 estimation method as it compares to the one in this paper. However, our study does not include bi-directional

exchange, and more work should be done in the future. In order to make the readers to understand this research better, in the revised version, we added the description for bidirectional exchange of NH_3 and the comparison with Fu et al. (2015) and Zhu et al. (2015) (See Lines 86-111 on Page 4 in the revised version), and the suggestion of our inventory for the future work (See Lines 497-500 on Page 18 in the revised version).

Lines 86-111 on Page 4 in the revised version: “In addition, studies have attempted to focus on implementing the bidirectional exchange of NH_3 in many air quality models, e.g., the Community Multi-scale Air Quality (CMAQ) model (Cooter et al., 2012; Bash et al., 2013; Pleim et al., 2013; Fu et al., 2015), and the GEOS-Chem global chemical transport model (Zhu et al., 2015). NH_3 deposition, emission, reemission, and atmospheric lifetime can be affected by rigorous treatment of the bidirectional flux of NH_3 , and vegetation and soil can be either a sink or a source of atmospheric NH_3 (Sutton et al., 2007). Fu et al. (2015) provides the first online estimate of NH_3 emissions from agricultural fertilizer use China, based on coupling the CMAQ model with a bi-directional NH_3 exchange module and the Environmental Policy Integrated Climate (EPIC) model, this method considers an increased number of influencing factors, such as meteorological fields, soil and fertilizer application, and provides improved NH_3 emissions with higher spatial and temporal resolution, whereas, gaps still exist for this method owing to the uncertainties of more model parameterization and input data. Zhu et al. (2015) developed the adjoint of bidirectional exchange in GEOS-Chem model which suggests that although the implementing bidirectional exchange greatly extends the lifetime of NH_3 in the atmosphere via deposition and reemission processes and conducts a better fundamental description of NH_3 emissions from fertilizers, whereas, it does not uniformly ameliorate estimation of NH_3 concentrations, NH_4^+ wet deposition and nitrate aerosol concentrations due to the NH_3 re-emissions from the ammonium soil pool that accumulates ammonium from previous months or the ammonium soil pool preserves ammonia/ammonium in the soil rather than emitting it directly after fertilizer application during the growing seasons (e.g., bidirectional exchange significant decreases NH_3 gross emissions in southeastern China and NH_3 concentrations in China in April of 2008, but changes in NH_4^+ wet deposition are not very large in April).”

Lines 497-500 on Page 18 in the revised version: “Our high resolution inventory can be applied to simulate atmospheric aerosol formation in air quality models with a bidirectional NH_3 exchange module, and then explore the effects of NH_3 emissions on China’s urban air pollution.”

3) Beside the previous studies of estimating emission inventories, satellite measurement is a good resource to evaluate the spatial distribution of NH_3 emissions inventory. The comparison of the satellite data and this study is not shown. There are many satellite data can be used, such as TES, IASI. I’m interested to see more results and discussions according to the comparisons to satellite data.

Response: We thank the reviewer for the comment. In the revised version, we added the temporal distribution comparison between observation and our study (Lines 318-319 on Page 12 in the revised version) (Ianniello et al., 2010; Meng et al., 2011). In the revised version, we also did some further discussions based upon other studies (Clarisse et al., 2009; Van Damme et al., 2014; Shephard et al., 2011; Van Damme et al., 2015) about the satellite measurement in order to evaluate the spatial and temporal distribution of NH_3 emissions inventory, such as the

Tropospheric Emission Spectrometer (TES), Infrared Atmospheric Sounding Interferometer (IASI), and have revised the manuscript by referring to the results (See Lines 264-281 on Page 10 and Lines 319-323 on Page 12 in the revised version).

Lines 264-281 on Page 10 in the revised version: “We compared our results with the global NH₃ column distribution using satellite monitoring from the Infrared Atmospheric Sounding Interferometer (IASI) (Clarisse et al., 2009; Van Damme et al., 2014). Several emissions hotspots are observed in the Tarim basin, the North China Plain and western Heilongjiang province and Jilin province by the IASI sensor, emission density is 4.2, 7.4, 5.2 and 9.6 tNH₃ km⁻² yr⁻¹, respectively. This result demonstrated excellent qualitative consistent with our estimated emissions. However, the higher emission areas were not observed by satellite monitoring because of clouds, water vapor, the surface temperature, high SO₂ emissions (Kharol et al., 2013; Wang et al., 2013; Garcia et al., 2008), land surface variation and the retrieval methods of NH₃ total columns (Xu et al., 2015). Higher plenty of cloud cover and precipitation could generate some uncertainties in the Sichuan Basin and Lianghu Plain. Additionally, NH₃ concentration distribution might not be always agreement with emission pattern due to its high reactive ability, solubility and short-lived in the atmosphere (Huang et al., 2012); Higher surface temperature and humidity could speed up NH₃ consumption simultaneously. And Sichuan Basin is high SO₂ pollution district where NH₃ gas could easily react with SO₂ (Zhang et al., 2009). Therefore, these factors lead the inconsistent between satellite monitoring and our inventory.”

Lines 318-319 on Page 12 in the revised version: “These higher emissions are virtually identical with several in situ data sets (Ianniello et al., 2010; Meng et al., 2011).”

Lines 319-323 on Page 12 in the revised version: “In addition, the seasonality of emissions from IASI and Tropospheric Emission Spectrometer (TES) satellite observations demonstrated excellent consistent with the temporal distribution of our inventory which is a summer maximum of NH₃ emissions in China (Shephard et al., 2011; Van Damme et al., 2015).”

Overall, I suggest publishing this manuscript after revision based on the comments above and below.

Response: We thank the referee for favorable considerations of our manuscript. We have made major revisions as suggested by the referee, and details are described in our responses to the Specific Comments and Technical Comments below.

Specific comments:

Page 25300, line 10. You don't have the spatial pattern for 1978-2007.

Response: We thank the reviewer for the comment. We apologize for our inaccurate description. We made the modification (See Lines 22-23 on Page 1 in the revised version).

Lines 22-23 on Page 1 in the revised version: “the temporal patterns of historical time trends for 1978-2008 were estimated and the uncertainties were quantified for the inventories;”

Page 25301, line 14. IPCC, 2006 is not the right way to cite.

Response: We thank the reviewer for the suggestion. We made the modification.

Page 25301, line 26. I'm not clear about the logic. Do you mean to achieve the balance of food demand and environment effects?

Response: We thank the reviewer for the comment. We are sorry that the description is not clear. We revised this sentence (See Lines 67-69 on Page 3 in the revised version).

Lines 67-69 on Page 3 in the revised version: “Therefore, to achieve the balance of food demand and environment effects, NH₃ emissions must be accurately estimated in a manner that reflects the spatial and temporal pattern of their sources.”

Page 25302, line 12. You have to list these “specific sectors”.

Response: We thank the reviewer for the suggestion. We have listed these “specific sectors” (See Lines 81-82 on Page 3 in the revised version).

Lines 81-82 on Page 3 in the revised version: “which is still limited to specific sectors, such as precipitation and the monitoring networks provide high-density data (Paulot et al., 2014).”

Page 25302, line 12-13. I guess you miss “in the year” after “temporal distributions”.

Response: We thank the reviewer for the suggestion. We made the modification.

Page 25303, line 21-23. So have you used data for Hong Kong and Macau or not? Please rephrase the sentence.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. In this study, Hong Kong and Macau primarily have no agriculture, and thus all activity data of them equal zero (See Lines 153-154 on Page 6 in the revised version).

Lines 153-154 on Page 6 in the revised version: “Hong Kong and Macau primarily have no agriculture, and thus all activity data of them equal zero”.

Page 25305, line 6-7. Please list these “specific parameters”.

Response: We thank the reviewer for the suggestion. We have listed these “specific parameters” (See Lines 192-193 on Page 7 in the revised version).

Lines 192-193 on Page 7 in the revised version: “this effect is only reflected by specific parameters (i.e., some form of dietary manipulation) on the farm scale (Ross et al., 2002).”

Page 25305, line 17. So, in this study, did you use the same EFs for all seasons or not? Please rephrase the sentence.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. The EFs for different seasons are different, and for the same season of different months are equivalent. We revised this sentence (See Lines 203-205 on Page 8 in the revised version).

Lines 203-205 on Page 8 in the revised version: “the EFs for the same season of different months are equivalent and the different seasons are different (Huang et al., 2012; Hutchings et al., 2001).”

Page 25306, line 5-6. Please explain the reason or list the reference for the assumption of “0.2”

right after this sentence.

Response: We thank the reviewer for the suggestion. We added the reference for the assumption of “0.2” (Zhou et al., 2014).

Page 25307, line 16. Add “from Infrared atmospheric sounding interferometer (IASI)” after “monitoring”.

Response: We thank the reviewer for the suggestion. We made the modification.

Page 25307, line 18. What “those” stand for? Please clarify here.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. We revised this sentence (See Lines 267-270 on Page 10 in the revised version).

Lines 267-270 on Page 10 in the revised version: “Several emissions hotspots are observed in the Tarim basin, the North China Plain and western Heilongjiang province and Jilin province by the IASI sensor, emission density is 4.2, 7.4, 5.2 and 9.6 tNH₃ km⁻² yr⁻¹, respectively. This result demonstrated excellent qualitative consistent with our estimated emissions.”

Page 25307, line 20. Do you want to say, “not all the higher emission areas were observed by satellite because of cloud . . .”? Actually, satellite is more able to observe high emissions than low emissions. The factors you mention here all could lead the inconsistent between satellite monitoring and your inventory, but do you know which is the main possible reason? It may vary for different regions, but you should at least explain more for one typical region for an example.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. We added explain of inconsistent for the Sichuan Basin and Lianghu Plain between satellite monitoring and our inventory, and revised this sentence (See Lines 274-281 on Page 10 in the revised version).

Lines 274-281 on Page 10 in the revised version: “Higher plenty of cloud cover and precipitation could generate some uncertainties in the Sichuan Basin and Lianghu Plain. Additionally, NH₃ concentration distribution might not be always agreement with emission pattern due to its high reactive ability, solubility and short-lived in the atmosphere (Huang et al., 2012); Higher surface temperature and humidity could speed up NH₃ consumption simultaneously. And Sichuan Basin is high SO₂ pollution district where NH₃ gas could easily react with SO₂ (Zhang et al., 2009). Therefore, these factors lead the inconsistent between satellite monitoring and our inventory.”

Page 25307, line 23-25. Are you still talking about findings from satellite observations? What is “emission density”? Do you mean high NH₃ emissions or concentrate areas with high NH₃ emissions? It is hard to image without a figure.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. We adjusted the location of this sentence (See Lines 262-264 on Page 10 in the revised version). This sentence is talking about findings from our inventory. In this study, “emission density” represents “the per-unit cultivated area NH₃ emission”. In figure 2(a), the resolution of each grid is 1 km × 1 km, and the unit of emission density is tNH₃ km⁻² yr⁻¹, so each grid represents either NH₃ emissions or concentrate areas with NH₃ emissions. We have used the figure 2(a) to display this mean and added the further explain for the implication of emission density (See Lines 251-252 on

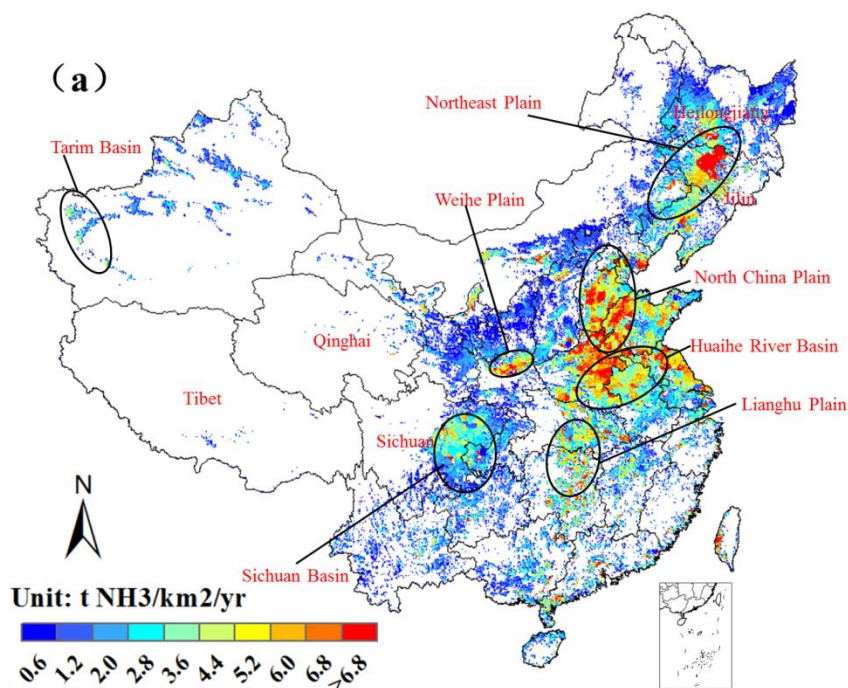


Figure.2 NH₃ emission map of China's agricultural fertilizer at 1 km × 1 km (a) for 2008. Major emission areas are circled.

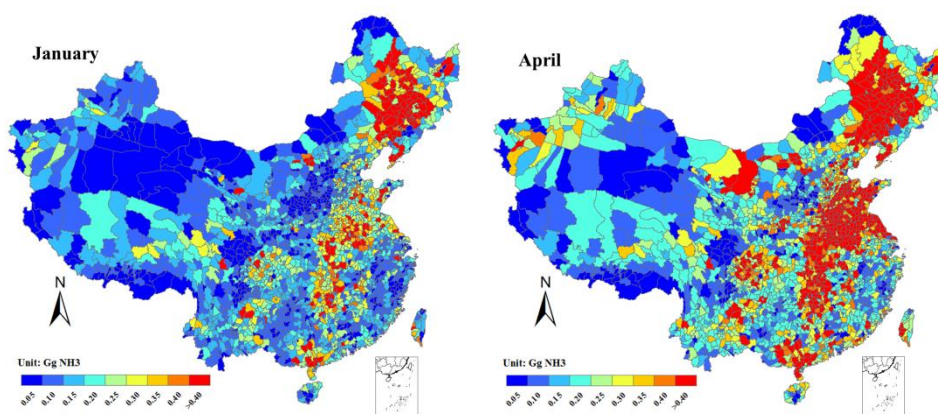
Lines 251-252 on Page 9 in the revised version: “The average emission density (per-unit cultivated area NH₃ emission) over western, central and eastern China is 4.7, 6.4 and 6.5 tNH₃ km⁻² yr⁻¹, respectively.”

Page 25308, line 21. Add “of the annual total emissions” after “39.7%”.

Response: We thank the reviewer for the suggestion. We made the modification.

Page 25309, line 9. “Fig. S3”: It is better to use the same color scale for all plots.

Response: We thank the reviewer for the suggestion. We made the modification.



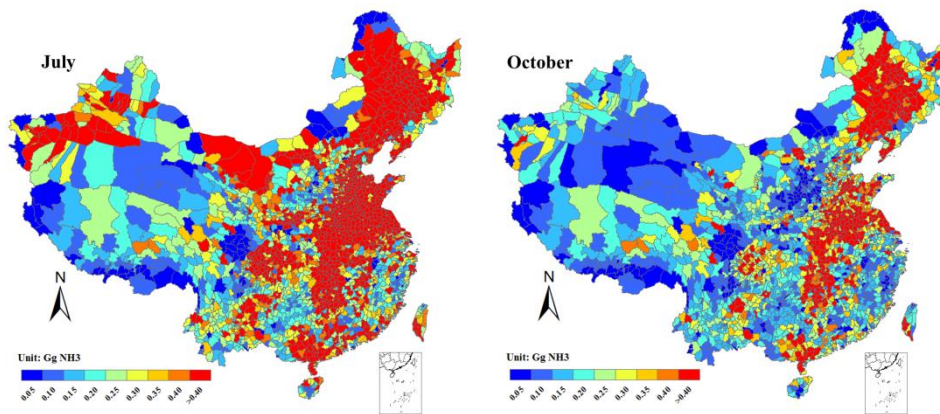


Figure S3 The spatial distributions of the agricultural fertilizer NH_3 emissions for January, April, July and October.

Page 25309, line 12-13. What kind of “temperature variations”?

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. In this study, “temperature variations” represents temperature rebounded significantly from March to April in central China, and from April to May in eastern and western China. We revised this sentence (See Lines 343-346 on Page 13 in the revised version).

Lines 343-346 on Page 13 in the revised version: “In central China, synthetic fertilizer’s monthly contribution proportions began to exceed livestock manure in April (Fig. S5), however, this condition occurred in May in eastern and western China because of temperature rebounded significantly.”

Page 25310, line 13. What does “CAY” stand for?

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. “CAY” stands for the China Agriculture Yearbook. We changed “CAY” to “NBSC” (See Line 375 on Page 14 and Line 478 on Page 17 in the revised version).

Page 25310, line 24. Change “first decreased” to “decreased in 2007”.

Response: We thank the reviewer for the suggestion. We made the modification.

Page 25311, line 9. What are the “others”?

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. “others” represent cattle, sheep, horses, donkeys and mules. We added this description in Lines 403-404 on Page 15 in the revised version.

Lines 403-404 on Page 15 in the revised version: “others (from cattle, sheep, horses, donkeys and mules) have observed the opposite trend.”

Page 25311, line 11-13. Please rephrase the sentence.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear.

We revised this sentence (See Lines 405-407 on Page 15 in the revised version).

Lines 405-407 on Page 15 in the revised version: “The average contribution of synthetic fertilizer

to E_{total} is approximately 38.3% during the past 31 years, and the minimum and maximum is 33.4% and 42.7% respectively.”

Page 25312, line 22-23. “Higher”? I see lower.

Response: We thank the reviewer for the corrections. We apologize for our inaccurate description. We made the modification (See Lines 447-448 on Page 16 in the revised version).

Lines 447-448 on Page 16 in the revised version: “Our estimates for 1994 to 2006 are approximately 1.8 times lower than those of Dong et al. (2010) for each year.”

Page 25312, line 23. Change “temporal distribution for” to “monthly variation of”.

Response: We thank the reviewer for the suggestion. We made the modification.

Page 25312, line 25. Add “(Figure 4)” after “Zhang et al. (2011)”.

Response: We thank the reviewer for the comment. We apologize that the date description about this reference is not clear here. In Figure 4, we added the date for the reference in caption. Details as follows Figure 4:

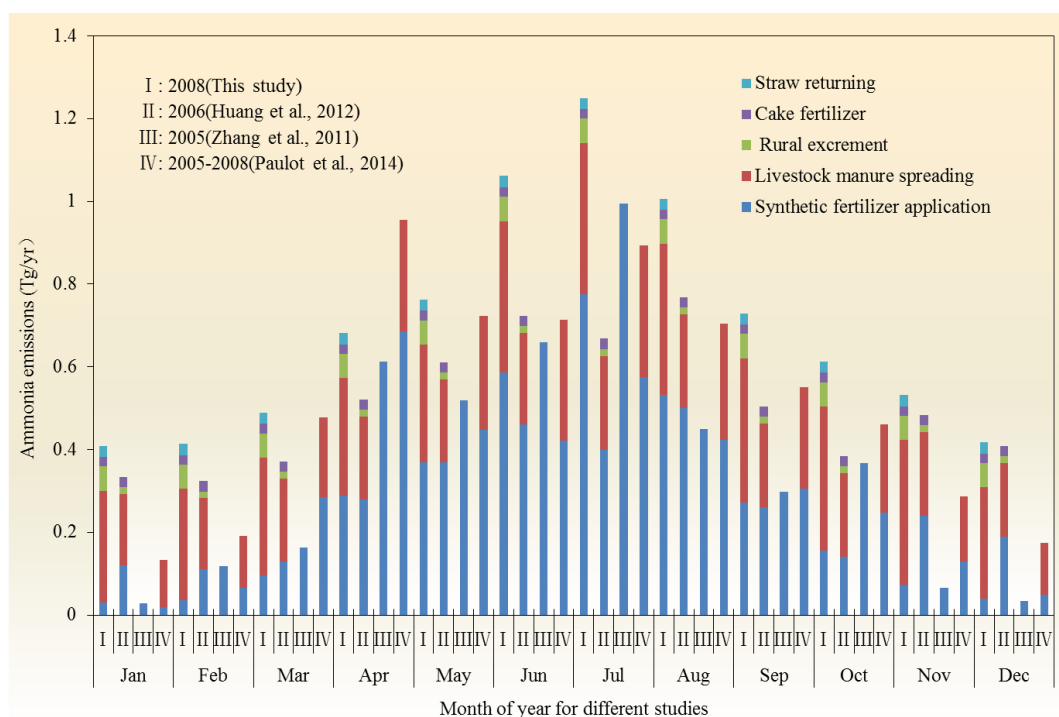


Figure.4 Monthly NH₃ emissions in 2008 and compared with earlier studies.

Page 25312, line 25. What does “the other” stand for?

Page 25312, line 28. I’m confused here. Which three? I thought “all three studies” are three previous studies. However, as I read the later sentence “however, Paulot et al. . .”, it seems that “all three studies” are Huang et., Zhang et al., and this study.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. “the other” stands for three research inventories of Paulot et al. (2014), Huang et al. (2012) and

Zhang et al. (2011). And we revised this sentence (See Lines 450-458 on Pages 16-17 in the revised version).

Lines 450-458 on Pages 16-17 in the revised version: “Our estimates agree well with the above three inventories for the monthly variation tendency. However, in our study and that of Zhang et al. (2011), emissions peaked in July, whereas in Huang et al. (2012), the emissions peaked in August, and the maximum emission occurred during summer, this phenomenon could be primarily attributed to the local climate conditions, which affected the EFs for the base year, but in Paulot et al. (2014), the emissions peaked in April because erroneous planting dates were used in the crop model such as the winter wheat-summer corn rotation, corn sown in June instead of April in China (Huang et al. 2012).”

Page 25313, line 9. “Emissions”. Do you mean total emissions? Should you only compare the LS+SF for these two studies since Paulot et al only has LS+SF?

Page 25313, line 12. I don't think the reason has been fully explained. Please finish the explanation.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. “Emissions” represents the emission from livestock manure spreading (LS). We only compare the LS for our inventory and Paulot et al. (2014) here. We revised this sentence and added further the explanation for the reason (See Lines 461-469 on Page 17 in the revised version).

Lines 461-469 on Page 17 in the revised version: “In our study, emissions for livestock manure spreading peaked in June-August, which was similar to the corresponding findings of Paulot et al. (2014), whereas the monthly emission in winter in our study was nearly 2.2-fold higher than in Paulot et al. (2014). The reason is that in Paulot et al. (2014) the timing of livestock manure spreading is presumed to be identical with synthetic fertilizer application and the crops hardly need synthetic fertilizer application in winter, however, in this study the number of livestock is same during each month of the year and the EFs for the same season of different months are equivalent.”

Page 25313, line 18. Urbanization rate is the average rate of change of the size of the urban population over a period of time. If the urbanization rate is 52.5% in 2008, urban population may be 100% in 2009. I guess 17.9% and 52.6% are urban population, not urbanization rate.

Response: We thank the reviewer for the comment. In China, the National Bureau of Statistics defined the urbanization rate is that the urbanization rate equals the proportion of the urban population and the total population (<http://www.stats.gov.cn/>). In fact, these data is both urban population and urbanization rate. We checked this sentence and the urbanization rate is 47.0% in 2008. We revised this sentence (See Lines 476-478 on Page 17 in the revised version).

Lines 476-478 on Page 17 in the revised version: “The nation's urbanization rate (the urbanization rate equals the proportion of the urban population and the total population, <http://www.stats.gov.cn/>) rapidly increased from 17.9% in 1978 to 47.0% in 2008 (NBSC, 2009a).”

Page 25314, line 11-12. Please rephrase the sentence.

Response: We thank the reviewer for the suggestion. We are sorry that the description is not clear. We revised this sentence (See Lines 503-507 on Page 18 in the revised version).

Lines 503-507 on Page 18 in the revised version: “With the local high-resolution data, spatially and temporally precise EF_{NH_3} and related parameters, times series of CAF_ NH_3 emissions were developed, which provide the high-resolution maps of NH_3 emission densities, the source apportionment, and the spatial and temporal pattern for 2008 as well as a historical time trend analysis of total NH_3 emissions from 1978 to 2008.”

Page 25314, line 26. Change “decrease CAF_ NH_3 emissions” to “decrease the NH_3 emissions from agricultural fertilizers”. I think the point is to decrease practical NH_3 emissions, not the NH_3 emission inventory CAF_ NH_3 .

Response: We thank the reviewer for the suggestion. We made the modification.

Technical comments:

Page 25302, line 22. Add “,” after “uncertainty”.

Page 25302, line 23. Delete “the findings of”.

Page 25303, line 19. Change “5” to “5 types”, “livestock types (8)” to “livestock (8 types)”, “crop types (17)” to “crop (17 types)”.

Page 25304, line 3. Add “.” after “2009”.

Page 25305, line 14. Add “the” between “is same”.

Page 25306, line 2. Change “to determine” to “determining”.

Page 25307, line 11. Change “or” to “and”.

Page 25307, line 13. Change “or” to “and”.

Page 25309, line 18. Change “from” to “based on”.

Page 25309, line 25. Add “,” after “In addition”. Add “of” after “because”.

Page 25313, line 19. Add “.” After “2009”.

Response: We thank the reviewer for the comment. The above mistakes have been amended.

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