Reply to Reviewer's Comments

We would like to first thank the editor and four 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

1. Application of EPIC model to China

The authors explain the lower emissions total from this study as being explained by US and UK emissions factors being applied to China (Sect. 3.2.2). In this study, EPIC is described as simulating "a wide range of vegetative systems, tillage systems, and other crop management practices" (p.750, l.25-6). To what extent are these methods specifically reflective of practices in China (versus the US) since the model was developed by US researchers for application in the US (originally)? Are there sensitivity tests that could be conducted to examine how much parameters that are known to differ between cultures influence the fertilizer applied? What extensions might be added in the future to more accurately represent farming practices in China?

Response: Thank you for comments. This is the first try to apply this model system to China and the first step to build the model system to estimate agricultural emissions. In this study, we didn't revise the algorithm in the EPIC model, but we used the Chinese input data, e.g. landscape, land use, crop, soil distribution, weather etc. In addition, this study focuses on the agriculture NH_3 emission, so the fertilizer use is the most important influencing factor among the crop management practices. In the US case (Cooter et al., 2012; Bash et al., 2013), they used the fertilizer application rates simulated by EPIC. However, the test results showed that the fertilizer application rates would be underestimated in China if the simulated values were used directly. This is because the Chinese farmers are used to applying more fertilizer. Therefore, in this study, the cultural fertilizer application rates from the Chinese statistics were used. Uncertainties indeed exist and more work should be done in the future in order to more accurately estimate the agriculture emissions in China. For example, more research should be done to capture the farmer's logic to use fertilizer and design the automatic fertilizer application algorithm in the EPIC model for China. We are trying to cooperate with the agricultural experts in China and the further work is going-on.

In order to make the readers to understand this research better, we have added more uncertainty analysis in section 3.4 and also give more advice about future work in the conclusion part.

2. Soil characteristics

The pH of the soil will have a significant impact on the partitioning of ammonium to ammonia. Since the cited website and associated data manual are in Chinese, the reader will be helped by an explanation in English in the paper of the method of estimating the pH of the soil across the country. 1 of 4 It is mentioned that some soil data are from the US soil profile. Which soil parameters are from this database? Why is it reasonable to use the US soil characteristics in these cases? How might these gaps in the Chinese database motivate future research in China? Also, does the 25-year spin up period in EPIC alter soil pH and other soil characteristics from the input parameters?

Response: Thank you for comments. We apologize that the description about soil characteristics is not clear. In this study, the dominant soil type in each grid is taken from the Harmonized World Soil Database, which is based on Chinese research, but the soil characteristics data is from the US soil profile data (Cooter et al., 2012; http://www.nrcs.usda.gov/wps/portal/nrcs/main/national/technical/nra/nri/). We matched the soil in each grid with a specific soil profile data based on soil type, ecological region and latitude. The soil characteristics of the matched soil in the soil profile dataset are used in the corresponding grid. The assumption is that in China and US, the soil characteristics of same soil types in the similar eco-region and latitude are similar. The major reasons why this US soil profile data was used in this study are as follows. Firstly, the Chinese soil profile data is very difficult to obtain. In the soil characteristics dataset of HWSD, some important soil characteristics input for the EPIC model are missing, e.g. soil albedo, initial soil water storage. Most importantly, this soil characteristics data is just an initial input for general soil, not specially for agriculture soil. The spin-up run will allow soil characteristics to adjust to the agriculture management. For example, EPIC is set up to apply lime to maintain the soil pH at levels that reduce crop stress due to low pH. For soil pH, the normal growth pH range of three dominant crops (rice, corn and wheat) is 6.0-7.0 (http://njzx.mianxian.gov.cn/xxgk/ccpf/20804.htm; http://nmsp.cals.cornell.edu/publications/factsheets/factsheet5.pdf). The 95% confidence interval of EPIC simulated values is 6.3-7.6, which is reasonable and acceptable although uncertainties still exist. Besides, the soil characteristics are also updated with CMAQ running.

This is a pilot study to apply this model system to China and it's the first step to build the model system to estimate agricultural emissions. Some uncertainties indeed exist and further improvement work is going-on. We are trying to cooperate with the soil experts in China to build the soil initial input file for EPIC based on Chinese soil profile data, which is a big work.

In order to make the readers to understand this research better, we revised the description about soil processing, added more uncertainty analysis in section 3.4 and also gave advice about future work in the conclusion part. Please see the revised manuscript.

3. Evaluation of model coupling

The authors state that two simulations were conducted "to evaluate the performance of this NH3 emission, fate and transport model", but the description of the distinctions of these two modeling scenarios is incomplete, which leaves confusion about the intention of the comparison as well as the utility of it.

The base case is indicated to use the Zhao et al. (2013) emissions inventory. Does it include the bi-directional flux algorithm in CMAQ? If not, the authors would ammonia emissions to influence atmospheric concentrations differently from the second model run simply because the ammonia can be re-emitted once deposited.

The bi-directional case is described as using ammonia emissions from fertilizer that were calculated online CMAQ. Given the name of the case, it is assumed that this includes the bidirectional treatment, but clarification would be helpful for the reader. If the distinction between the two scenarios is not whether the bi-directional algorithm is included but rather the method of estimating agricultural ammonia emissions, this case should be renamed to indicate that distinction.

In addition to clarifying the distinctions, it would be helpful to explain the purpose behind the choice of model configurations in the two cases. Is the base case designed to reflect what others might model without the capabilities that these authors have added to the CMAQ framework?

Response: Thank you for your comments. I am sorry that the description is not clear. In this study, the distinction between the two cases is the method of estimating ammonia emissions from fertilizer use. In the Base-case, the emissions from Zhao et al. (2013) was used, which was estimated by the traditional "emission-factor" method. The bi-directional flux algorithm in CMAQ was not used. In the Bidi-case, the emission was estimated online by the bi-directional module in the CMAQ. The bi-directional flux algorithm is a major part of this method. In order to make it more clearer for the readers, we revised the last paragraph the section 2.3:

"In order to evaluate the performance of this method, two simulations are conducted in this study, including Base-case and Bidi-case. The difference between these two cases is the method of estimating ammonia emissions from fertilizer use. For Base-case, the emission inventory from Zhao et al. (2013) is used, which is estimated by the traditional "emission-factor" method. This case does not include the bi-directional flux algorithm in CMAQ. For Bidi-case, NH_3 emission is estimated online by the bi-directional module in the CMAQ. The emissions of ammonia from other sectors and the emissions of other pollutants are both from Zhao et al. (2013) in these two cases."

The locations at which aerosol were collected are, presumably, urban. Were both anions and cations observed by ion chromatograph? If so, were their relative abundances indicative of the sulfate being fully neutralized by ammonium such that the authors would expect ammonium nitrate to be the primary component controlling nitrate presence? Was sodium or another cation present in the samples sufficiently to suggest that nitrate may partition apart from the contribution of ammonium?

If it is not possible to evaluate whether sulfate would be fully neutralized in these locations through observations, this information should be available in the CMAQ grid cells representative of the observation locations, which would provide some indication of the relevance of these measurements to evaluating ammonia emissions.

Response: Thank you for the comments. In addition to NH_4^+, some other anions and cations were also observed by ion chromatograph, such as SO_4^{2+}, NO_3^-, Ca^{2+}, K^+, Mg^{2+}, Na^+. In order to answer this question, three indicators in Fountoukis et al., (2007)were used:

$$R_1 = \frac{[NH_4^+] + 2[Ca^{2+}] + [K^+] + 2[Mg^{2+}] + [Na^+]}{[SO_4^{2-}]}$$

$$R_2 = \frac{2[Ca^{2+}] + [K^+] + 2[Mg^{2+}] + [Na^+]}{[SO_4^{2-}]}$$

$$R_3 = \frac{2[Ca^{2+}] + [K^+] + 2[Mg^{2+}]}{[SO_4^{2-}]}$$

Based on their values, different aerosol composition regimes are defined and the different possible species exist for each regime, as shown in **Table.R1**(Fountoukis et al., 2007):

Table.R1. Potential species for different aerosol composition regimes

Regime Number	R1	R2	R3	Aerosol type	Solid phase
1	R1<1	any value	any value	Sulfate Rich	$NaHSO_4$, NH_4HSO_4, $KHSO_4$, $CaSO_4$
2	1⩽R1<2	any value	any value	Sulfate Rich	$NaHSO_4$, NH_4HSO_4,Na_2SO_4, $(NH_4)_2SO_4$,$(NH_4)_3H(SO_4)_2$, $CaSO_4$, $KHSO_4$,K_2SO_4, $MgSO_4$
3	R1⩾2	R2<2	any value	Sulfate Poor, Crustal & Sodium Poor	Na_2SO_4, $(NH_4)_2SO_4$, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$
4	R1⩾2	R2⩾2	R3<2	Sulfate Poor, Crustal & Sodium Rich, Crustal Poor	Na_2SO_4, $NaNO_3$, NaCl, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$
5	R1⩾2	R2⩾2	R3>2	Sulfate Poor, Crustal & Sodium Rich, Crustal Rich	$NaNO_3$, NaCl, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $Mg(NO_3)_2$, $MgCl_2$, KNO_3, KCl

The observed R values for the three months at three monitoring stations were shown in **Table.R2**. It can be seen that R_1 are all greater than 2, implying that sulfate would be fully neutralized. R2 are smaller than 2 or approximately equal to 2, implying that NH_4NO_3 is dominant for nitrate.

Table.R2. The R values at three monitoring stations

	Shanghai			Suzhou			Nanjing		
	R1	R2	R3	R1	R2	R3	R1	R2	R3
June (2011.6.1-6.30)	6.5	0.8	0.6	3.7	1.3	0.8	4.7	0.7	0.7
August (2011.7.20-8.20)	3.4	0.9	0.5	2.7	1.0	0.4	2.8	0.2	0.2
Nov (2011.11.1-11.30)	4.7	0.9	0.5	7.8	2.1	0.5	6.8	0.8	0.7

4. Comparison with other emissions estimates

The other studies to which the ammonia emissions estimates of this work are compared do not include the bi-directional flux of ammonia. Could the authors include an estimate (perhaps based on the two studies conducted in this work) how different they might anticipate the estimates of ammonia emissions in the other studies to be if they were calculated in accordance with the method used in this work (i.e., bi-directional flux of ammonia)? Perhaps the change would be negligible, but even this information would be worth including in Section 3.2.2.

Response: Thank you for the comments. I am sorry for confusing you here. Similar with the reply to the question 3, the distinction between this study and the other studies is the method of estimating ammonia emissions from fertilizer use. In the other studies, the ammonia emissions were estimated offline using "emission factor" method. In this study, the emission was estimated online by the bi-directional module in the CMAQ. The bi-directional flux algorithm is a major part of this method.

5. Uncertainty analysis

The authors note that previous studies (e.g., the national statistical database)likely has uncertainties. Had those authors provided confidence intervals on their estimates, error bars in the ammonia emissions estimates might be included in Figures5 and 7. Similarly, when future studies cite this work, they would be helped by having estimates of the uncertainty due to select parameters (e.g., parameters in the bidirectional flux model mentioned in Section 3.4). If it is feasible for the authors to provide quantification of uncertainty in ammonia emissions by propagating uncertainty in some parameters, future research would certainly benefit from such an estimate.

Response: Thank you for comments. We agree that uncertainty analysis is important and beneficial. For the previous studies mentioned in this study, Streets et al. (2003) and Huang et al. (2012) gave an estimated uncertainty of ±53% and -34%~28% for the total NH_3 emission, rather than only from fertilizer use, based on quantitative analysis. Zhang et al. (2011) considered an uncertainty of ±50% was appropriate for NH_3 emission from fertilizer use based on semi-quantitative analysis. Dong et al. (2010) and Zhao et al. (2013) didn't represent uncertainty estimation in the papers. Different from the traditional emission-factor method, NH_3 emission was calculated online by the model directly, the uncertainties of which were associated with quality of the input data, and the mathematical algorithm and a large amount of parameters applied in the EPIC and bi-directional model. Therefore, it's difficult to provide uncertainty intervals accurately for the estimated NH_3 emissions. Nevertheless, more detailed uncertainty analysis for the major impact factors has been done in this study. We have revised the discussion about uncertainty in section 3.4, which is as follows.

" This is a pilot study to apply this model system to estimate the NH_3 emission in China and large uncertainties still exist for this method at some aspects. Quality of input data, mathematical algorithm, and parameters applied in EPIC and the bi-directional model may be associated with uncertainties in the model output.

Fertilizer application rates for each crop are important input data for the estimation of NH_3 emissions from agricultural fertilizers. They are obtained from the agricultural statistics. These statistical data should have some level of uncertainty, because the amounts of samples in the census are limited. Beusen et al. (2008) has employed an uncertainty of ±10% for the statistical data of fertilizer use based on expert judgments when estimating the global NH_3 emission. A June

2006 sensitivity run of this bi-directional model in US shows that a 50% increase of crop fertilizer use would result in a 31% increase in NH_3 emission (Denniset al., 2013). In addition, the spatial distribution of NH_3 emissions from agricultural fertilizer is strongly related to cropland area and its distribution, which are achieved from the MODIS data. Friedl et al. (2010) mentions that the producer's and user's accuracies are 83.3%/92.8% for MODIS class 12 (cropland) and 60.5%/27.5% for class 14 (Cropland/Natural Vegetation Mosaic) in MODIS Collection 5 product. This would lead to the uncertainties of spatial distribution. Additionally, due to the limit of data availability, the initial characteristics of the dominant soil in each grid are gotten from the US dataset. Although we have matched the soil based on soil type, eco-region, and latitude, uncertainties still existed due to different long-term agriculture management.

Seeing from the algorithm described in section 2.3, the EPIC outputs, including soil NH_4^+ concentration, soil volumetric water content (θ_s) and soil pH, are important inputs of the bidirectional module. EPIC has been used and evaluated world widely to simulate nitrogen cycle and soil water. Some validation studies have found favorable results for soil nitrogen or/and crop nitrogen uptake levels (Cavero et al., 1998 and 1999; Wang et al., 2014). However, less accurate simulation results are also reported (Chung et al., 2002). For soil volumetric water content, Li et al. (2004) found that EPIC model could catch the variation of soil water in different years well with the relative bias of 11.7%, and the research conducted by Huang et al. (2006) also showed that the EPIC-simulated long-term average θ_s values were not significantly different from the measured values in the Loess Plateau of China. For soil pH, the normal growth pH range of three dominant crops (rice, corn and wheat) is 6.0-7.0 (http://njzx.mianxian.gov.cn/xxgk/ccpf/20804.htm; http://nmsp.cals.cornell.edu/publications/factsheets/factsheet5.pdf). The 95% confidence interval of EPIC simulated values is 6.3-7.6, which is reasonable and acceptable although uncertainties still exist.

The bi-directional ammonia flux module in the CMAQ is the core of this model system. The uncertainties of the bidirectional exchange parameterization would bring uncertainties to NH_3 emission estimates. Pleim et al. (2013) has compared the simulated NH_3 flux from the box model of this ammonia bi-directional flux algorithm with observations in three periods. The results showed that the model generally reproduced the observed series and significantly correlated with the observations (p<0.001). The mean normalized biases were 78.6%, -49% and 1% for soybeans (18 June-24 August, 2002), corn (21-29 June, 2007) and corn (11-19 July, 2007), respectively. The soil gamma (Γ_g) and appoplast gamma (Γ_s) are two important parameters in this ammonia bi-directional flux algorithm (Bash et al., 2013) and their parameterization remains uncertain (Massad et al., 2010). The field measurements of Γ_g and Γ_s are limited, and measured values are scattered owing to complex impact factors (Massad et al., 2010 and reference therein). Dennis et al.(2013) assessed the effects of these uncertainties. A 50% increase of Γ_g would result in a 42.3% increase in NH_3 emission. Two different parameterization methods of Bash et al.(2013) and Massad et al. (2010) could lead to a 17% change in NH_3 emission.

It's very difficult to give an uncertainty interval accurately for this method, because there are many factors contributing to this model system. Here, an uncertainty of about ±50% is considered appropriate based on the above analysis, which is also the upper limit of uncertainty in previous studies (Bouwman et al., 1997; Zhang et al., 2011; Zheng et al., 2012). Therefore, the NH_3 emission from agricultural fertilizer application in China of 2011 is in the range of 1.5-4.5Tg. In order to reduce the uncertainty, much work still need to do. In addition to improve the quality of input data, additional local measurements of soil and vegetation chemistry, ambient NH_3 concentration and flux data are needed to enhance and evaluate the parameterizations of EPIC model and bi-directional module."

Specific Comments

A. Abstract

Lines Comment

20Add space before “Compared”; “researches” to “research”

B. Text

Page | Lines Comment

748 | 5 “aerosol and nitric acid (HNO3) to generate” to “and nitrate (NO3-)aerosol, adding to the concentration of”

750 | 9,14 “agriculture” to “agricultural”

750 | 21 “modeled 36 km CMAQ” to “CMAQ”

750 | 24 “agriculture” to “agricultural”

751 | 2 “it’s” to “it is” (also on p.759 at line 16)

751 | 5 “next” to “next section”

753 | 3 Please provide a citation of personal communication.

755 | 2 “fraction of the crop” to “fraction of cell used for crop”

756 | 8 “kg grid−1 cell” to “kg grid cell−1”

758 | 15 “alkaline gas in the atmosphere, NH3” to “positive ion in the atmosphere, NH4+”

Response: Thank you for your comments. The above editorial mistakes have been amended.

759 | 1 Why were July 1-19 not included in the observations? Is November selected to evaluate the performance at lower temperatures?

Response: Thank you. In China, the observation data for chemical components of fine particulates was very spare and not publicly available. The reason that July 1-19 was not included is that there were no field measurements in these days.

760 | 5 “researches” to “research”

760 | 19 “human activity has on food production with air-quality” to “human activity has on air quality through food production”

760 | 19 “with climate model” to “with climate models”

Response: Thank you for your comments. The above editorial mistakes have been amended.

764 | 18 The Williams et al. (2008) citation is for APEX, not EPIC, even though in the text EPIC is the model mentioned. Please correct the reference.

Response: Thank you for your comments. The reference has been changed to

"Williams, J. R., Jones, C. A., and Dyke, P. T.: A modeling approach to determining the relationship between erosion and soil productivity., Trans. ASAE, 27, 129–144, 1984"

C. Figures

Figure 2. Please add the locations of the nitrate observations to the map.

Response: Thank you. The locations of the nitrate observations have been added to Fig.2.

Fig.R1. The modeling domain and the black points represent the locations of the nitrate observations

Figure 5. It is nice that the authors mention uncertainty in the statistical database on p. 759, l. 17. Does the statistical database include any confidence interval estimates that could be included as error bars?

Response: Thank you. The statistical data in Fig.5 was obtained from the investigation of Zhang et al. (2008). Unfortunately no confidence interval estimates were included in that research.

Figure 7. Given the importance of temperature and precipitation to the emissions rate as noted in the text, could an indicator of these variables be provided alongside the current results? One option would be to produce a single box-and-whisker plot as Figure 7a with temperature on the left y-axis and precipitation on the right y-axis against the months of the year on the x-axis so that the median, quartiles, and extremes of these important driving parameters would be evident as readers evaluate the ammonia emissions (perhaps as Figure 7b).

Figure 7. In addition to the suggested addition above, making the units on the y-axis Tg (consistent with Table 3) would assist the reader in reading this absolute scale.

Response: Thank you. Figure 7 has been revised based on the comments, as shown in the following figure.

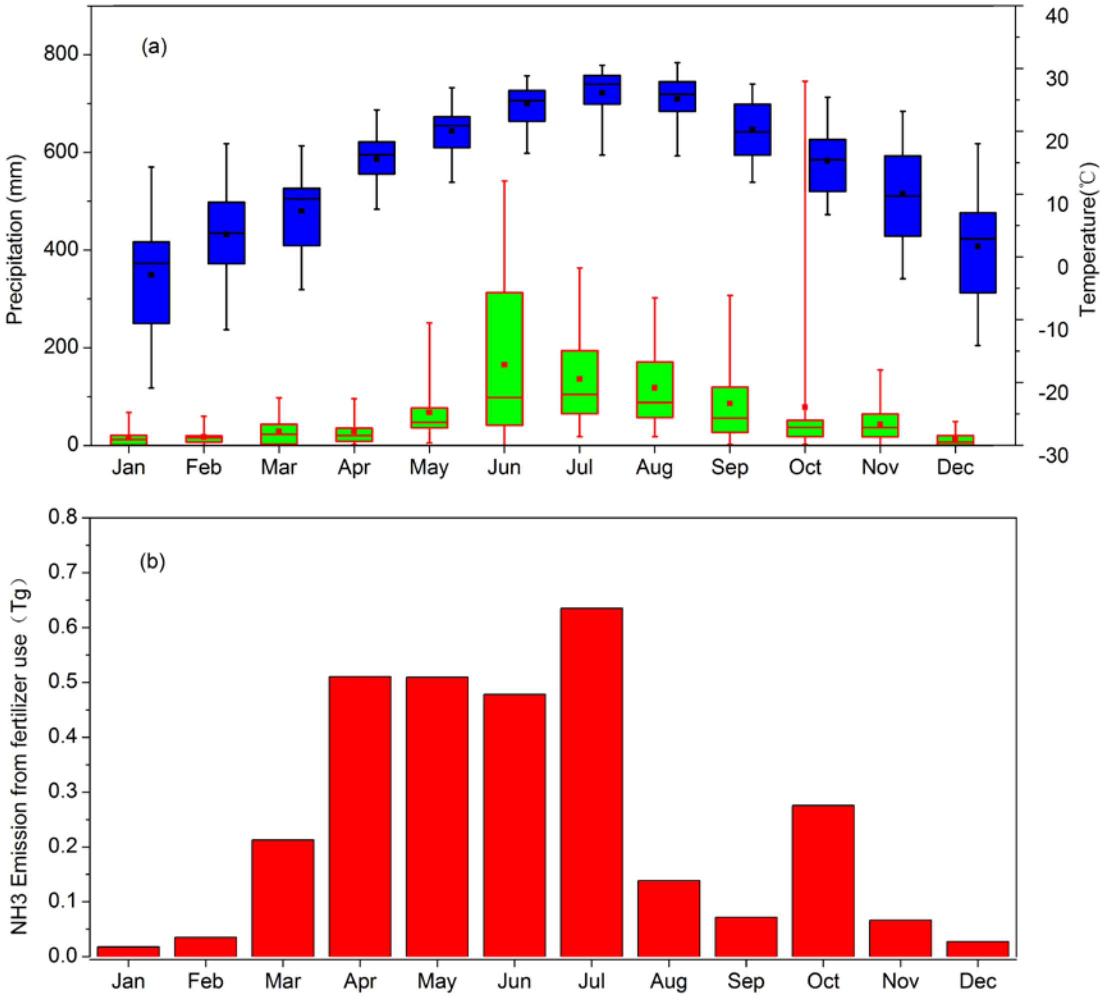

Fig.R2.(a)The variation of monthly precipitation (green) and temperature (blue) in 31 provinces. In the box-whisker plots, the boxes and whiskers indicate the 100th (max), 75th, 50th (median), 25th and 0th(min) percentiles, respectively. The point represents the average value. (b) Monthly NH_3 emissions from N fertilizer use

Figure 9. Please consider replacing this column chart with five pie charts that show the fraction of NH_3 emissions from each province for each of the five studies being evaluated. These could be ordered as the province contributing the most to the least for each study (i.e., for each pie). As it is, the results are very hard to compare from each study. If the authors have a special purpose behind using the bar chart, please at least order the provinces according to the most to least fractional contribution according to this study.

Response: Thank you for the comments. The reason we use the bar chart is that there are too many provinces so that it is not easy to distinct using the pie charts. Here, we revised the figure by ordering the provinces according to the most to least fractional contribution in this study, as shown in the following figure. In addition, we have moved this figure to the supplementary materials.

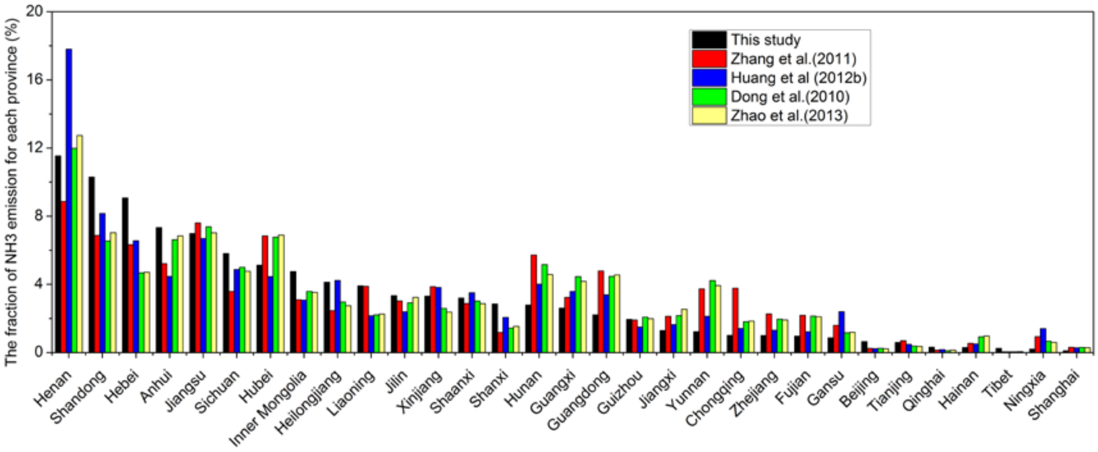

Fig.R3. Comparison of provincial NH_3 emissions from N fertilizer use in different studies

Comments from Anonymous Referee #2

General Comments

Accurate ammonia emissions are crucial for correctly simulating aerosol concentrations and in developing aerosol control strategies using regional air quality models. Developing and evaluating models to estimate ammonia emissions falls within the scope of ACP and is of interest to the readers. However, this manuscript is lacking in two areas. First, there is insufficient observational data available to evaluate the developed model system. Second, the comparison with previously published studies seems tenuous since different years are being compared without some type of normalization. Is one year higher then another simply because more fertilizer was applied? What role do economic factors play in determining amount of fertilizer applied and techniques used? Without some way of normalizing between years, evaluating models by comparing total estimated ammonia emissions for different years is difficult. Similarly, lack of independent observational data to evaluate any of the models, makes it very difficult to conclude any one model better represents the actual ammonia emissions. Obviously, further observations are beyond the scope of this manuscript. However, the authors may consider running this model system using input data from one of the years previously reported to make a more valid comparison on the two models. Nevertheless, the manuscript represents an advancement in modeling agricultural emissions and could be published in ACP after minor changes and more explicitly addressing the need for more thorough model evaluation with observational data.

Response: Thank you for comments. In order to make the inventories more comparable, we updated the emissions in different years to the year of 2011 based on the changes of fertilizer use, temperature and precipitation.

(1) fertilizer use

The basic emission factors in each research remain same. First of all, the NH_3 emissions are affected by the amount of fertilizer used. The amount of different fertilizer types used in each province from 2000 to 2011 were obtained from the Chinese statistics. The values for the whole country were shown in the following figure. We firstly updated the NH_3 emissions in these researches according to the changes of fertilizer use.

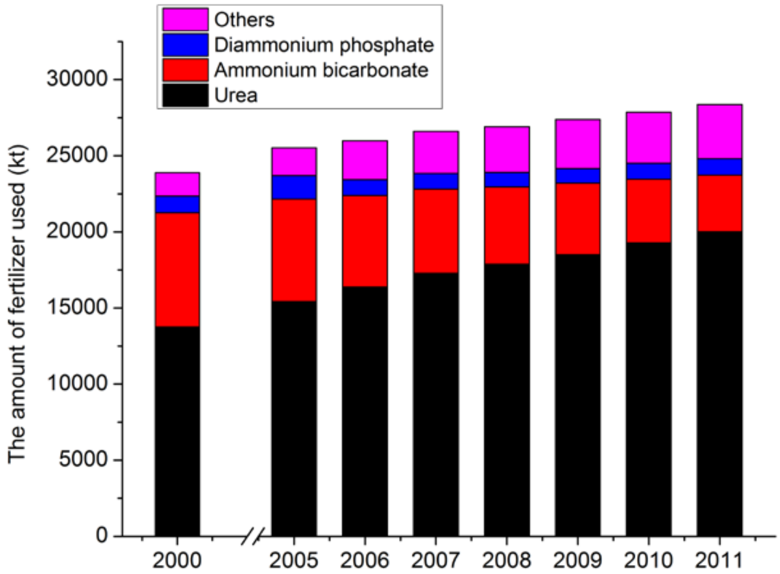

Fig.R4. The amount of different fertilizer types used in China from 2000 to 2011

(2) temperature

Zhang et al. (2011) and Huang et al. (2012) considered the impacts of temperature on emission factors. The averaged temperatures in major cities for each province and each month in the year of 2005, 2006 and 2011 were obtained from the China statistical yearbook. The annual averaged temperatures were shown in the following figure:

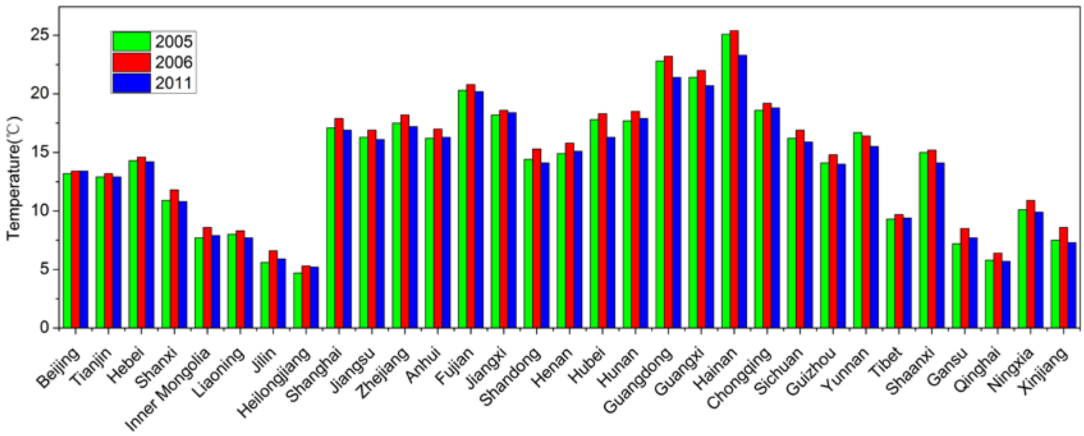

Fig.R5. The provincial temperatures in the year of 2005, 2006 and 2011

Huang et al. (2012) set four temperature intervals: <10℃, 10-20℃, 20-30℃ and >30℃. In each temperature interval, specific emission factor was used. The interval width is 10℃, but the temperature change between these years, so we don't consider the impacts of temperature on the result of Huang et al. (2012).

In the research of Zhang et al. (2011), the impact factor of temperature $RF_{temperature}$ is determined by equation:

$$RF_{temp} = e^{\left(0.1386\times\left(T_{month}-T_{year}\right)/3\right)} / 2$$

Here, T_{month} is the monthly averaged temperature and T_{year} is the annual averaged temperature. We adjusted the NH_3 emission in Zhang et al. (2011)from 2005 to 2011 according to the change of RF_{temp}.

(3) precipitation

Zhang et al. (2011) considered the impacts of precipitation on emission factors. The precipitations in major cities for each province and each month in the year of 2005 and 2011 were obtained from the China statistical yearbook. The total precipitations were shown in the following figure:

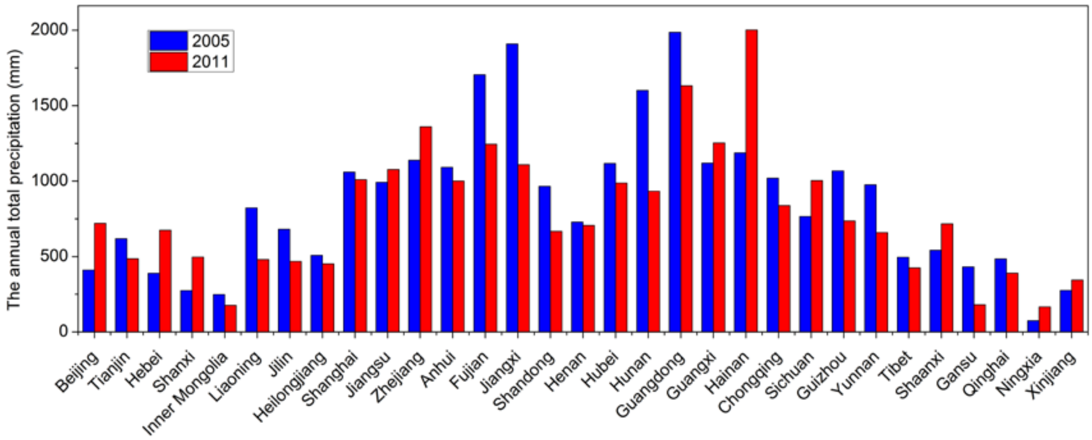

Fig.R6. The provincial precipitations in the year of 2005, 2006 and 2011

In the research of Zhang et al. (2011), the impact factor of precipitation $RF_{precipitation}$ is set as 0.75, 0.80, 0.85,0.90, 0.95 and 1.0 for significant rainfall events (>5 mm in 24 h)within 24h, 24-48h, 48-72h, 72-96h, 96-120h and >120h. We adjusted the NH_3 emission in Zhang et al. (2011)from 2005 to 2011 according to the change of days with significant rainfall events (>5 mm in 24 h).

In summary, the updated results of comparison for the total emission were shown in the following table.

Table.R3. Comparison of the NH3 Emissions from fertilizer use in our study with other published results

Reference	Year	Original NH_3 Emission (Tg/yr)	Revised to 2011(Tg/yr)
Streets et al. (2003)	2000	6.7	7.0
Zhang et al. (2011)	2005	3.6	3.8
Huang et al.(2012b)	2006	3.2	3.2
Dong et al. (2010)	2006	8.7	8.9
Zhao et al.(2013)	2010	9.8	9.8
This study	2011	3	3

Methodology and inputs

1. In section 2.2.2 Soil Information, both the China Soil Scientific Database and the US soil profile data are used. Given the different agricultural practices and history of each country, the

authors should address the appropriateness of combine the two databases when calculating soil pH. How does each compare with actually soil pH measurements in the respective countries?

Response: Thank you for comments. We apologize that the description about soil characteristics is not clear. In this study, the dominant soil type in each grid is taken from the Harmonized World Soil Database, which is based on Chinese research, but the soil characteristics data is from the US soil profile data (Cooter et al., 2012; http://www.nrcs.usda.gov/wps/portal/nrcs/main/national/technical/nra/nri/). We matched the soil in each grid with a specific soil profile data based on soil type, ecological region and latitude. The soil characteristics of the matched soil in the soil profile dataset are used in the corresponding grid. The assumption is that in China and US, the soil characteristics of same soil types in the similar eco-region and latitude are similar. The major reasons why this US soil profile data was used in this study are as follows. Firstly, the Chinese soil profile data is very difficult to obtain. In the soil characteristics dataset of HWSD, some important soil characteristics input for the EPIC model are missing, e.g. soil albedo, initial soil water storage. Most importantly, this soil characteristics data is just an initial input for general soil, not specially for agriculture soil. The spin-up run will allow soil characteristics to adjust to the agriculture management. For example, EPIC is set up to apply lime to maintain the soil pH at levels that reduce crop stress due to low pH. For soil pH, the normal growth pH range of three dominant crops (rice, corn and wheat) is 6.0-7.0 (http://njzx.mianxian.gov.cn/xxgk/ccpf/20804.htm; http://nmsp.cals.cornell.edu/publications/factsheets/factsheet5.pdf). The 95% confidence interval of EPIC simulated values is 6.3-7.6, which is reasonable and acceptable although uncertainties still exist. Besides, the soil characteristics are also updated with CMAQ running.

This is a pilot study to apply this model system to China and it's the first step to build the model system to estimate agricultural emissions. Some uncertainties indeed exist and further improvement work is going-on. We are trying to cooperate with the soil experts in China to build the soil initial input file for EPIC based on Chinese soil profile data, which is a big work.

In order to make the readers to understand this research better, we revised the description about soil processing, added more uncertainty analysis in section 3.4 and also gave advice about future work in the conclusion part. Please see the revised manuscript.

2. The terms basal and topdressing fertilizer should be defined and explained in section2.2.4. What is the differences between the two? For example, is one applied before the other, type of fertilizer used, method of application?

Response: Thank you for comments. The difference between the basal and topdressing fertilizer is the time when the fertilizer is used. Basal fertilizer is used before crops are planted and topdressing fertilizer is used during crops are growing. We added the description to section 2.2.4.

3. The term bi-directional is not defined or explained anywhere in the text. It should be further explained in section 2.3 and why it could be important to include in estimating ammonia emissions.

Response: Thank you for comments. We added the additional description to section 2.3:

"Direct flux measurements have shown that the air–surface flux of NH_3 is bidirectional, and vegetation and soil can be a sink or a source of atmospheric NH_3 (Fowler et al., 2009; Sutton et al.,1995). The direction and magnitude of the flux depend on the concentration gradient between the canopy or soil and the atmosphere. Bash et al. (2013) implemented a bi-directional ammonia flux module in CMAQv5.0.1 to represent this process. This module is based on the two-layer (soil and vegetation canopy) resistance model described by Pleim et al. (2013), which is similar to the model presented by Nemitz et al., (2001). The NH_3 air–surface flux (F_t)is calculated by the following formula:

$$F_t = \frac{1}{R_a + 0.5R_{inc}}\left(C_c - C_a\right)$$

where the aerodynamic resistance (R_a) and the in-canopy aerodynamic resistance (R_{inc}) are calculated following Pleim et al. (2013). C_a is the atmospheric NH_3 concentration. C_c is a function of C_a, the soil compensation point (C_g) and the stomatal compensation point (C_{st}).

$$C_c = \frac{\dfrac{C_a}{R_a + 0.5R_{inc}} + \dfrac{C_{st}}{R_b + R_{st}} + \dfrac{C_a}{0.5R_{inc} + R_{bg} + R_{soil}}}{\left(R_a + 0.5R_{inc}\right)^{-1} + \left(R_b + R_{st}\right)^{-1} + \left(R_b + R_w\right)^{-1} + \left(0.5R_{inc} + R_{bg} + R_{soil}\right)^{-1}}$$

where the quasi laminar boundary layer resistance of leaf surface (R_b), the stomatal resistance (R_{st}) and the quasi laminar boundary layer resistance of ground surface (R_{bg}) are calculated following Pleim et al. (2013). The cuticular resistance (R_w) is a function of C_c similar to Jones et al. (2007). C_{st} and C_g are calculated as follows:

$$C_{st} = M_n / V_m \frac{161500}{T_c} e^{\left(-\frac{10380}{T_c}\right)} \Gamma_s$$

$$C_g = M_n / V_m \frac{161500}{T_s} e^{\left(-\frac{10380}{T_s}\right)} \Gamma_g$$

where M_n is the molar mass of NH_3, V_m is the conversion factor of L to m^3, T_s and T_c are the soil and canopy temperature in K. The appoplast gamma (Γ_s) is modeled with a function similar to Zhang et al. (2010). The soil gamma (Γ_g) is defined as soil $[NH_4^+]/[H^+]$, and the soil NH_4^+ budget in CMAQ was parameterized following the method in EPIC (Williams et al., 1984). When fertilizer is used, Γ_g is calculated by the following function:

$$\Gamma_g = \frac{N_{app} / \left(\theta_s M_N d_s\right)}{10^{-pH}}$$

where N_{app} is the fertilizer application rate (g N/m^2), θ_s is the soil volumetric water content (m^3/m^3), M_N is the molar mass of nitrogen (14 g/mol), d_s is the depth of soil layer (m), and pH is the soil pH. The initial soil NH_4^+,θ_s and pH are all from the EPIC output and then calculated in CMAQ hourly. "

Results and Discussion

4. Two conditions are necessary for the formation of ammonium nitrate particles(NH4NO3). First, there has to be enough gas phase ammonia to partition to the particle phase and neutralize all the sulfate before it can react with nitrate. Second, the partial pressure product of gas phase ammonia and nitric acid has to be sufficient to create thermodynamically favor conditions for NH4NO3

formation. Since the molar ratio of NH3:HNO3 in ammonium nitrate is 1:1, it is not necessarily true that aerosol nitrate is only sensitive to gas phase ammonia. Even in agricultural areas with high ammonia emissions, aerosol nitrate could be low if there is no source of nitric acid. Using CMAQ modeled aerosol nitrate to evaluate the ammonia emissions assumes that CMAQ is correctly modeling gas phase nitric acid. How valid is this assumption? Does CMAQ simulate nitric acid correctly? What is the uncertainty of CMAQ modeled photochemical oxidation products, such as nitric acid? Also, what other aerosol components were measured with the IC system? Was the observed sulfate neutralized? Was there evidence of any other cations indicating the presence of other nitrates in the aerosol? Further and more comprehensive field measurements are necessary to fully evaluate this model system.

Response: Thank you for the comments. In China, the observation data for HNO_3 concentration was very spare and not publicly available. Therefore, the comparison between observation and simulation can't be done in this case and few evaluations in China can be found at the same time. But some researches for other countries can be used as a reference. Zhang et al.(2006) and Shimadera et al.(2014) used CMAQ model to simulate the HNO_3 concentrations in US and Japan, respectively. The comparison results are shown in **Fig.R7** and **Table R4**, respectively. It can be seen that the model performance for HNO_3 is acceptable.

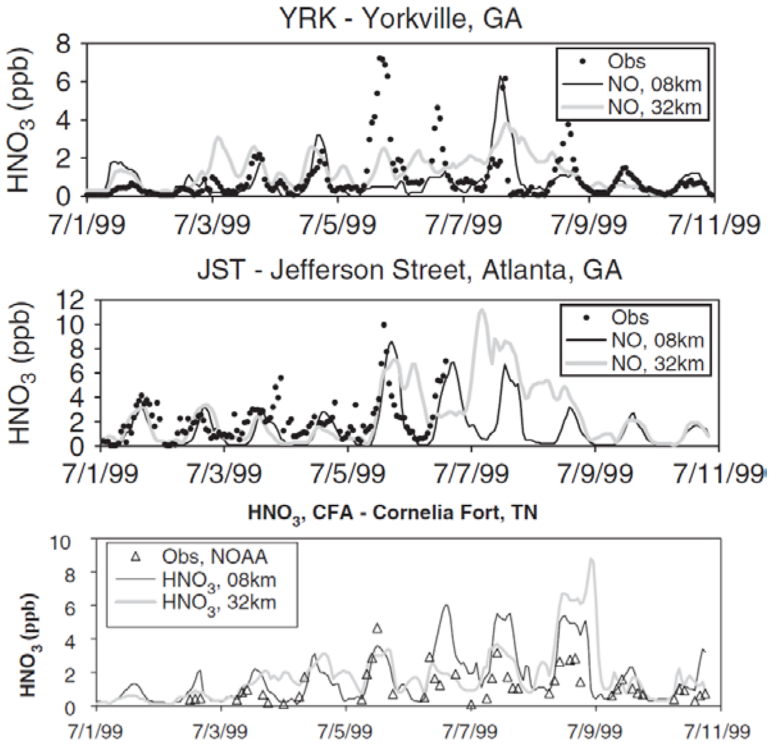

Fig.R7. The comparisons of observed and predicted HNO3 in Zhang et al.(2006)

Table R4. Comparisons of observed and simulated HNO3 for three sites of Japan in Shimadera et al.(2014)

	Winter 2010			Summer 2011		
	Komae	Kisai	Maebashi	Komae	Kisai	Maebashi
Sample number	-	42	-	30	30	30

Mean Obs.(ug/m3)	-	0.3	-	3.3	2.8	1.5
Mean Sim.(ug/m3)	1	0.7	0.6	2	2.9	1.1
r	-	0.89	-	0.25	0.75	0.79
NMB(%)	-	169	-	-40	2	16

In addition to NH_4^+, some other anions and cations were also observed by ion chromatograph, such as SO_4^{2+}, NO_3^-, Ca^{2+}, K^+, Mg^{2+}, Na^+. In order to know whether the sulfate was neutralized and aerosol types, three indicators in Fountoukis et al., (2007) were used:

$$R_1 = \frac{[NH_4^+] + 2[Ca^{2+}] + [K^+] + 2[Mg^{2+}] + [Na^+]}{[SO_4^{2-}]}$$

$$R_2 = \frac{2[Ca^{2+}] + [K^+] + 2[Mg^{2+}] + [Na^+]}{[SO_4^{2-}]}$$

$$R_3 = \frac{2[Ca^{2+}] + [K^+] + 2[Mg^{2+}]}{[SO_4^{2-}]}$$

Based on their values, different aerosol composition regimes are defined and the different possible species exist for each regime, as shown in the following table (Fountoukis et al., 2007):

Table.R5. Potential species for different aerosol composition regimes

Regime Number	R1	R2	R3	Aerosol type	Solid phase
1	R1<1	any value	any value	Sulfate Rich	$NaHSO_4$, NH_4HSO_4, $KHSO_4$, $CaSO_4$
2	1⩽R1<2	any value	any value	Sulfate Rich	$NaHSO_4$, NH_4HSO_4,Na_2SO_4, $(NH_4)_2SO_4$,$(NH_4)_3H(SO_4)_2$, $CaSO_4$, $KHSO_4$,K_2SO_4, $MgSO_4$
3	R1⩾2	R2<2	any value	Sulfate Poor, Crustal & Sodium Poor	Na_2SO_4, $(NH_4)_2SO_4$, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$
4	R1⩾2	R2⩾2	R3<2	Sulfate Poor, Crustal & Sodium Rich, Crustal Poor	Na_2SO_4, $NaNO_3$, NaCl, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$
5	R1⩾2	R2⩾2	R3>2	Sulfate Poor, Crustal & Sodium Rich, Crustal Rich	$NaNO_3$, NaCl, NH_4NO_3, NH_4Cl, $CaSO_4$, K_2SO_4, $MgSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $Mg(NO_3)_2$, $MgCl_2$, KNO_3, KCl

The observed R values for the three months at three monitoring stations were shown in the following table. It can be seen that R_1 are all greater than 2, implying that sulfate would be fully neutralized. R2 are smaller than 2 or approximately equal to 2, implying that NH_4NO_3 is dominant for nitrate.

Table.R6. The R values at three monitoring stations

	Shanghai			Suzhou			Nanjing		
	R1	R2	R3	R1	R2	R3	R1	R2	R3
June (2011.6.1-6.30)	6.5	0.8	0.6	3.7	1.3	0.8	4.7	0.7	0.7
August (2011.7.20-8.20)	3.4	0.9	0.5	2.7	1.0	0.4	2.8	0.2	0.2
Nov (2011.11.1-11.30)	4.7	0.9	0.5	7.8	2.1	0.5	6.8	0.8	0.7

We agree that further and more comprehensive field measurements are necessary to fully evaluate this model system. We have added the advice about future work in the conclusion part.

5. The authors pass on including or performing any uncertainty analysis. This is disappointing. While it may be difficult to estimate the uncertainty in some of the model input parameters, what uncertainty analysis has been done for previous CMAQ model studies? What is the uncertainty of the bi-directional ammonia flux module? In the end it is difficult for the reader to determine whether the differences between the observations and the two model runs or the differences between the two model runs are significant. From Table 4, it is not clear to me that the coupled modeling system improved the nitrate aerosol simulation at a significant level in all cases. For example, in June the bias in the bidi case is larger than the base case for all three stations.

Response: Thank you for comments. We agree that uncertainty analysis is important and beneficial. More detailed uncertainty analysis for the major impact factors were added to section 3.4, which is as follows.

" This is a pilot study to apply this model system to estimate the NH_3 emission in China and large uncertainties still exist for this method at some aspects. Quality of input data, mathematical algorithm, and parameters applied in EPIC and the bi-directional model may be associated with uncertainties in the model output.

Fertilizer application rates for each crop are important input data for the estimation of NH_3 emissions from agricultural fertilizers. They are obtained from the agricultural statistics. These statistical data should have some level of uncertainty, because the amounts of samples in the census are limited. Beusen et al. (2008) has employed an uncertainty of ±10% for the statistical data of fertilizer use based on expert judgments when estimating the global NH_3 emission. A June 2006 sensitivity run of this bi-directional model in US shows that a 50% increase of crop fertilizer use would result in a 31% increase in NH_3 emission (Denniset al., 2013). In addition, the spatial distribution of NH_3 emissions from agricultural fertilizer is strongly related to cropland area and its distribution, which are achieved from the MODIS data. Friedl et al. (2010) mentions that the producer's and user's accuracies are 83.3%/92.8% for MODIS class 12 (cropland) and 60.5%/27.5% for class 14 (Cropland/Natural Vegetation Mosaic) in MODIS Collection 5 product. This would lead to the uncertainties of spatial distribution. Additionally, due to the limit of data availability, the initial characteristics of the dominant soil in each grid are gotten from the US dataset. Although we have matched the soil based on soil type, eco-region, and latitude, uncertainties still existed due to different long-term agriculture management.

Seeing from the algorithm described in section 2.3, the EPIC outputs, including soil NH_4^+ concentration, soil volumetric water content (θ_s) and soil pH, are important inputs of the bidirectional module. EPIC has been used and evaluated world widely to simulate nitrogen cycle and soil water. Some validation studies have found favorable results for soil nitrogen or/and crop nitrogen uptake levels (Cavero et al., 1998 and 1999; Wang et al., 2014). However, less accurate simulation results are also reported (Chung et al., 2002). For soil volumetric water content, Li et al. (2004) found that EPIC model could catch the variation of soil water in different years well with the relative bias of 11.7%, and the research conducted by Huang et al. (2006) also showed that the EPIC-simulated long-term average θ_s values were not significantly different from the measured values in the Loess Plateau of China. For soil pH, the normal growth pH range of three dominant crops (rice, corn and wheat) is 6.0-7.0 (http://njzx.mianxian.gov.cn/xxgk/ccpf/20804.htm; http://nmsp.cals.cornell.edu/publications/factsheets/factsheet5.pdf). The 95% confidence interval of EPIC simulated values is 6.3-7.6, which is reasonable and acceptable although uncertainties still exist.

The bi-directional ammonia flux module in the CMAQ is the core of this model system. The uncertainties of the bidirectional exchange parameterization would bring uncertainties to NH_3 emission estimates. Pleim et al. (2013) has compared the simulated NH_3 flux from the box model of this ammonia bi-directional flux algorithm with observations in three periods. The results showed that the model generally reproduced the observed series and significantly correlated with the observations (p<0.001). The mean normalized biases were 78.6%, -49% and 1% for soybeans (18 June-24 August, 2002), corn (21-29 June, 2007) and corn (11-19 July, 2007), respectively. The soil gamma (Γ_g) and appoplast gamma (Γ_s) are two important parameters in this ammonia bi-directional flux algorithm (Bash et al., 2013) and their parameterization remains uncertain (Massad et al., 2010). The field measurements of Γ_g and Γ_s are limited, and measured values are scattered owing to complex impact factors (Massad et al., 2010 and reference therein). Dennis et al.(2013) assessed the effects of these uncertainties. A 50% increase of Γ_g would result in a 42.3% increase in NH_3 emission. Two different parameterization methods of Bash et al.(2013) and Massad et al. (2010) could lead to a 17% change in NH_3 emission.

It's very difficult to give an uncertainty interval accurately for this method, because there are many factors contributing to this model system. Here, an uncertainty of about ±50% is considered appropriate based on the above analysis, which is also the upper limit of uncertainty in previous studies (Bouwman et al., 1997; Zhang et al., 2011; Zheng et al., 2012). Therefore, the NH_3 emission from agricultural fertilizer application in China of 2011 is in the range of 1.5-4.5Tg. In order to reduce the uncertainty, much work still need to do. In addition to improve the quality of input data, additional local measurements of soil and vegetation chemistry, ambient NH_3 concentration and flux data are needed to enhance and evaluate the parameterizations of EPIC model and bi-directional module."

Specific Comments

Page 747, line 4 add “husbandry” or “production” after “livestock”
Page 747, line 20 add space before “Compared” and change “researches” to research
Page 748, lines4 and 5 This sentence is awkward and incorrect. NH3 does not partition to nitric acid.
Page 748, line 12 NH3 was previously defined as ammonia in line.
Page 750, lines 9, 14, 24 change “agriculture” to “agricultural”
Page 750, line 21 remove “36 km”
Page 751, line 2 change “it’s” to “it is”
Page 751, line 5 add “section” after “next”
Page 753, line 4 change “accurate” to “accurately”
Page 754, line 3 add space between “Nemitz” and “et”
Page 756, line 19 change “consumption” to “usage”
Response: Thank you for your comments. These editorial mistakes have been amended.

Figure 2. Add the locations of the nitrate observations to the map.
Response: Thank you. The locations of the nitrate observations have been added to Fig.2.

Fig.R8. The modeling domain and the black points represent the locations of the nitrate observations

Figure 3. What does the small insert represent?
Response: It represents the south China sea and its islands. We have added this clarification.

Figure 4. Again, what are the small inserts on the left for?

Response: It represents the south China sea and its islands. We have added this clarification.

Figure 5. Use month name on the x-axis instead of number

Response: Thank you. We have revised the figure.

Figure 6. As with Figs. 3 and 4, what is shown in the small insert?

Response: It represents the south China sea and its islands. We have added this clarification.

Figure 7. Change the y-axis units to Tg or kg for consistency with other tables. Use the month name on the x-axis.

Response: Thank you. We have revised the figure.

Figure 8. What are the small inserts for? This is a difficult figure to read because the panels are so small. Consider putting each months map into paper supplemental

Response: Thank you. We have put each months map into paper supplemental.

Comments from Anonymous Referee #3

General Comments

1. Compared with the US case in the research of Cooter et al.,2012 and Bash et al., 2013, what are the differences and difficulties when this method is used in China?

Response: Thank you. This is a pilot study to apply this model system to estimate the NH_3 emission in China. One of the major differences and difficulties is the collecting and processing Chinese local input data, e.g. landscape, land use, crop area by county, soil type distribution, weather characteristics and fertilizer application characteristics etc. In addition, based on Chinese agriculture, we added new crop types into the system, like early-rice and late-rice. Besides, this study focuses on the agriculture NH_3 emission, so the fertilizer use is the most important influencing factor among the crop management practices. In the US case (Cooter et al., 2012; Bash et al., 2013), they used the fertilizer application rates simulated by EPIC. However, the test results showed that the fertilizer application rates would be underestimated much in China if the simulated values were used directly, because the Chinese farmers are used to applying much fertilizer. Therefore, in this study, the cultural fertilizer application rates from the Chinese statistic materials were used.

2. It is nice to see the authors compared the results of this study with others in 3.2.2.In addition to the current comparison, I wonder if the authors can also compare the seasonal variations of different NH_3 emission inventories.

Response: Thank you for comments. We have added the comparison in section 3.2.2. Compared with provincial distributions, the difference of seasonal variations among these studies is larger, as shown in **Fig.R1**. The seasonal profile in Zhao et al. (2013) was based on temperature variations. In addition to temperature, others also considered the impacts of fertilizer application timing. It is indeed difficult to capture entirely the exact date of fertilizing for the whole China, which may bring this large diversity. For example, Huang et al. (2012) thought that the basal-dressing and top-dressing fertilizer of winter wheat were conducted in September and November, respectively. However, the basal-dressing fertilizer was applied in October in this study and Zhang et al. (2011), and the top-dressing fertilizer was mainly used in March in the next year. The diversity of seasonal variations among different studies reflects that large uncertainties still exist for temporal distribution of NH_3 emissions and much local research work is still needed.

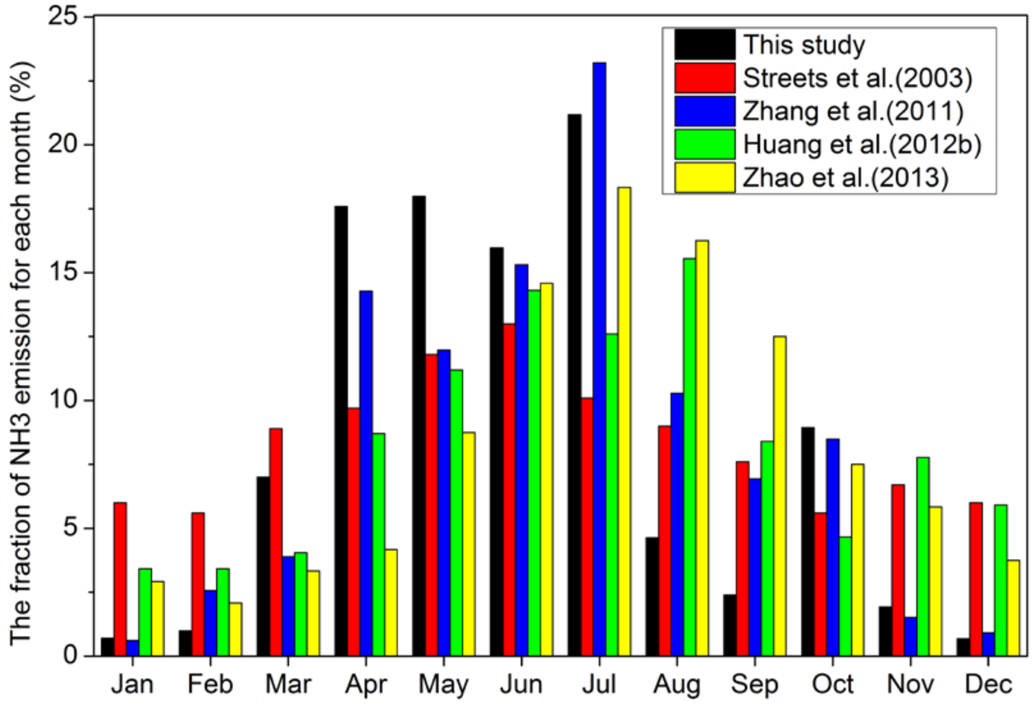

Fig.R9. Comparison of monthly NH_3 emissions from N fertilizer use in different studies

3. The discussions on the uncertainties of NH_3 emissions are simple and not very clear, is it possible to have more details in conducting or estimating the uncertainties of emission inventories in this study?

Response: Thank you for comments. We agree that uncertainty analysis is important and beneficial. More detailed uncertainty analysis for the major impact factors were added to section 3.4, which is as follows.

" This is a pilot study to apply this model system to estimate the NH_3 emission in China and large uncertainties still exist for this method at some aspects. Quality of input data, mathematical algorithm, and parameters applied in EPIC and the bi-directional model may be associated with uncertainties in the model output.

Fertilizer application rates for each crop are important input data for the estimation of NH_3 emissions from agricultural fertilizers. They are obtained from the agricultural statistics. These statistical data should have some level of uncertainty, because the amounts of samples in the census are limited. Beusen et al. (2008) has employed an uncertainty of ±10% for the statistical data of fertilizer use based on expert judgments when estimating the global NH_3 emission. A June 2006 sensitivity run of this bi-directional model in US showed that a 50% increase of crop fertilizer use would result in a 31% increase in NH_3 emission (Denniset al., 2013). In addition, the spatial distribution of NH_3 emissions from agricultural fertilizer is strongly related to cropland area and its distribution, which are achieved from the MODIS data. Friedl et al. (2010) mentions that the producer's and user's accuracies are 83.3%/92.8% for MODIS class 12 (cropland) and

60.5%/27.5% for class 14 (Cropland/Natural Vegetation Mosaic) in MODIS Collection 5 product. This would lead to the uncertainties of spatial distribution. Additionally, due to the limit of data availability, the initial characteristics of the dominant soil in each grid are gotten from the US dataset. Although we have matched the soil based on soil type, eco-region, and latitude, uncertainties still existed due to different long-term agriculture management.

Seeing from the algorithm described in section 2.3, the EPIC outputs, including soil NH_4^+ concentration, soil volumetric water content (θ_s) and soil pH, are important inputs of the bidirectional module. EPIC has been used and evaluated world widely to simulate nitrogen cycle and soil water. Some validation studies have found favorable results for soil nitrogen or/and crop nitrogen uptake levels (Cavero et al., 1998 and 1999; Wang et al., 2014). However, less accurate simulation results were also reported (Chung et al., 2002). For soil volumetric water content, Li et al. (2004) found that EPIC model could catch the variation of soil water in different years well with the relative bias of 11.7%, and the research conducted by Huang et al. (2006) also showed that the EPIC-simulated long-term average θ_s values were not significantly different from the measured values in the Loess Plateau of China. For soil pH, the normal growth pH range of three dominant crops (rice, corn and wheat) is 6.0-7.0 (http://njzx.mianxian.gov.cn/xxgk/ccpf/20804.htm; http://nmsp.cals.cornell.edu/publications/factsheets/factsheet5.pdf). The 95% confidence interval of EPIC simulated values is 6.3-7.6, which is reasonable and acceptable although uncertainties still exist.

The bi-directional ammonia flux module in the CMAQ is the core of this model system. The uncertainties of the bidirectional exchange parameterization would bring uncertainties to NH_3 emission estimates. Pleim et al. (2013) compared the simulated NH_3 flux from the box model of this ammonia bi-directional flux algorithm with observations in three periods. The results showed that the model generally reproduced the observed series and significantly correlated with the observations (p<0.001). The mean normalized biases were 78.6%, -49% and 1% for soybeans (18 June-24 August, 2002), corn (21-29 June, 2007) and corn (11-19 July, 2007), respectively. The soil gamma (Γ_g) and appoplast gamma (Γ_s) are two important parameters in this ammonia bi-directional flux algorithm (Bash et al., 2013) and their parameterization remains uncertain (Massad et al., 2010). The field measurements of Γ_g and Γ_s are limited, and measured values are scattered owing to complex impact factors (Massad et al., 2010 and reference therein). Dennis et al.(2013) assessed the effects of these uncertainties. A 50% increase of Γ_g would result in a 42.3% increase in NH_3 emission. Two different parameterization methods of Bash et al.(2013) and Massad et al. (2010) could lead to a 17% change in NH_3 emission.

It's very difficult to give an uncertainty interval accurately for this method, because there are many factors contributing to this model system. Here, an uncertainty of about ±50% is considered appropriate based on the above analysis, which is also the upper limit of uncertainty in previous studies (Bouwman et al., 1997; Zhang et al., 2011; Zheng et al., 2012). Therefore, the NH_3 emission from agricultural fertilizer application in China of 2011 is in the range of 1.5-4.5Tg. In order to reduce the uncertainty, much work still need to do. In addition to improve the quality of input data, additional local measurements of soil and vegetation chemistry, ambient NH_3 concentration and flux data are needed to enhance and evaluate the parameterizations of EPIC model and bi-directional module."

4. As mentioned in section 3.4, some uncertainties still exist for this approach. I would like to suggest that in the conclusion part, authors may need to add some discussions about the possible improvements for this model when it is applied in China in the future.

Response: Thank you for comments. We have added some discussions about the possible improvements for this model applying to China in the conclusion section, which is as follows.

"This is a pilot study to apply this model system to estimate the NH_3 emission in China and uncertainties still exist for this method due to the uncertainties of model parameterization and input data. Much work is still needed to improve this model system when it is applied in China in the future. For example, it is important to build the soil initial input file for EPIC based on Chinese soil profile data. In addition, Chinese farmers' logic of agriculture management shall be explored and the automatic management algorithm in the EPIC model for China shall be designed. This model system also can likely be improved with additional local measurements of soil and vegetation chemistry, ambient NH_3 concentration and flux data to enhance and evaluate the parameterizations of EPIC model and bi-directional module."

Specific Comments

1. P751, Line11: In the year of 2010, there are 2856 official counties in China, not 2710. Please check.

Response: Thank you. I am sorry that the description here is confusing. Due to the limit of data availability, we just collected the crop area data in 2710 counties. We have revised the description to "Data of cropland area for each crop in 2710 counties was collected and processed based on each province-level or city-level statistical yearbook."

2. Page 753, Line 1-2:It would be nice to have some sort of citation for the "unpublished materials"

Response: Thank you. We have provided a note of personal communication for the unpublished materials in the revised manuscript.

3. P756, Line 7-8, kg grid-1seems not a good unit. It might be better to use kg ha-1.

Response: Thank you. We have revised the unit in the revised manuscript.

4. Page 772: Please clarify what the thin black line represents in Figure 3.

Response: Thank you. The thin black line in figure 3 represents the county boundary. We have added this clarification.

5. Page 778: The green colors in Fig. 8 are not easy to distinct. Please make the figure more readable.

Response: Thank you. Maybe the panels are too small to distinct the green colors. We have put each months map into paper supplemental.

6. The language shall be improved. For example, the tenses in some sentences are confusing in Line 1-3, Page 751 & Line 4-7,Page 756. Please double check the languages of the whole paper carefully.

Response: Thank you. We have double checked the languages of the whole paper carefully.

Comments from Anonymous Referee #4

General Comments

Clearly, the manuscript is on a subject matter appropriate for and of interest to ACP. However, I struggle to understand the significance and context of the results in this study. For example, despite the huge uncertainties in the various input data for the models, there is no estimate of the corresponding uncertainties in the overall emission rate. Sensitivity studies are needed to show how uncertainties in various quantities would result in changing the total emission rate. While I recognize that the input data also likely doesn't have intrinsic uncertainties, the authors need to provide uncertainty estimates of these input variables or at least show how the final emissions would change for a given range of uncertainty. What parameters are most sensitive to the final number and to what extent?

Response: Thank you for comments. The significance of this research includes the following aspects:

(1) For the first time, the NH_3 emissions from fertilizer using in China are estimated using a recently-developed modeling framework in which agricultural activity can be parameterized by meteorological, crop, and soil data. The NH_3 emissions can be calculated as the air quality model progresses through time and the temporal resolution is improved to hourly.

(2) This model system tightly links agricultural, weather and atmospheric processes, and allows for some interesting future research. For example, the model makes it possible to reflect the impacts of long-term climate and agricultural management changes on the NH_3 emission and atmospheric aerosol. If linking it to a water quality and transport model, the impacts of atmospheric nitrogen deposition from CMAQ and nutrient run off from EPIC on the water eutrophication can be estimated. This is the first try to apply this model system to China, and it's also the foundation to explore more scientific researches in the future.

(3) To drive the models, we aggregated and built some important national datasets, such as fertilizer application rate per crop and region, and crop area by county, which are also important contributions to the scientific literature.

We agree with you that uncertainty analysis is important. This is a pilot study to apply this model system to China and it's the first step to build the big system to accurately and reasonably model agricultural emissions. Some uncertainties indeed exist for now and uncertainty analysis for the major impact factors has been added to section 3.4, which is also given as follows.

" This is a pilot study to apply this model system to estimate the NH_3 emission in China and large uncertainties still exist for this method at some aspects. Quality of input data, mathematical algorithm, and parameters applied in EPIC and the bi-directional model may be associated with uncertainties in the model output.

Fertilizer application rates for each crop are important input data for the estimation of NH_3 emissions from agricultural fertilizers. They are obtained from the agricultural statistics. These statistical data should have some level of uncertainty, because the amounts of samples in the census are limited. Beusen et al. (2008) has employed an uncertainty of ±10% for the statistical data of fertilizer use based on expert judgments when estimating the global NH_3 emission. A June 2006 sensitivity run of this bi-directional model in US showed that a 50% increase of crop fertilizer use would result in a 31% increase in NH_3 emission (Denniset al., 2013). In addition, the spatial distribution of NH_3 emissions from agricultural fertilizer is strongly related to cropland area and its distribution, which are achieved from the MODIS data. Friedl et al. (2010) mentions that the producer's and user's accuracies are 83.3%/92.8% for MODIS class 12 (cropland) and 60.5%/27.5% for class 14 (Cropland/Natural Vegetation Mosaic) in MODIS Collection 5 product. This would lead to the uncertainties of spatial distribution. Additionally, due to the limit of data availability, the initial characteristics of the dominant soil in each grid are gotten from the US dataset. Although we have matched the soil based on soil type, eco-region, and latitude, uncertainties still existed due to different long-term agriculture management.

Seeing from the algorithm described in section 2.3, the EPIC outputs, including soil NH_4^+ concentration, soil volumetric water content (θ_s) and soil pH, are important inputs of the bidirectional module. EPIC has been used and evaluated world widely to simulate nitrogen cycle and soil water. Some validation studies have found favorable results for soil nitrogen or/and crop nitrogen uptake levels (Cavero et al., 1998 and 1999; Wang et al., 2014). However, less accurate simulation results were also reported (Chung et al., 2002). For soil volumetric water content, Li et al. (2004) found that EPIC model could catch the variation of soil water in different years well with the relative bias of 11.7%, and the research conducted by Huang et al. (2006) also showed that the EPIC-simulated long-term average θ_s values were not significantly different from the measured values in the Loess Plateau of China. For soil pH, the normal growth pH range of three dominant crops (rice, corn and wheat) is 6.0-7.0 (http://njzx.mianxian.gov.cn/xxgk/ccpf/20804.htm; http://nmsp.cals.cornell.edu/publications/factsheets/factsheet5.pdf). The 95% confidence interval of EPIC simulated values is 6.3-7.6, which is reasonable and acceptable although uncertainties still exist.

The bi-directional ammonia flux module in the CMAQ is the core of this model system. The uncertainties of the bidirectional exchange parameterization would bring uncertainties to NH_3 emission estimates. Pleim et al. (2013) compared the simulated NH_3 flux from the box model of this ammonia bi-directional flux algorithm with observations in three periods. The results showed that the model generally reproduced the observed series and significantly correlated with the observations (p<0.001). The mean normalized biases were 78.6%, -49% and 1% for soybeans (18 June-24 August, 2002), corn (21-29 June, 2007) and corn (11-19 July, 2007), respectively. The soil gamma (Γ_g) and appoplast gamma (Γ_s) are two important parameters in this ammonia bi-directional flux algorithm (Bash et al., 2013) and their parameterization remains uncertain (Massad et al., 2010). The field measurements of Γ_g and Γ_s are limited, and measured values are scattered owing to complex impact factors (Massad et al., 2010 and reference therein). Dennis et al.(2013) assessed the effects of these uncertainties. A 50% increase of Γ_g would result in a 42.3% increase in NH_3 emission. Two different parameterization methods of Bash et al.(2013) and Massad et al. (2010) could lead to a 17% change in NH_3 emission.

It's very difficult to give an uncertainty interval accurately for this method, because there are many factors contributing to this model system. Here, an uncertainty of about ±50% is considered appropriate based on the above analysis, which is also the upper limit of uncertainty in previous studies (Bouwman et al., 1997; Zhang et al., 2011; Zheng et al., 2012). Therefore, the NH_3 emission from agricultural fertilizer application in China of 2011 is in the range of 1.5-4.5Tg. In order to reduce the uncertainty, much work still need to do. In addition to improve the quality of input data, additional local measurements of soil and vegetation chemistry, ambient NH_3 concentration and flux data are needed to enhance and evaluate the parameterizations of EPIC model and bi-directional module."

Furthermore, more thorough discussion is needed to compare to prior studies other than a vague statement that emission factors from the US or UK are not applicable to China. While this may be true, what aspects of the emission factors are not relevant? How are management practices so different that the US/UK emission factors would not be representative of those practices in China?

Response: Thank you for comments. We agree with you that it is not appropriate to give a vague statement. After a further analysis, we think that the discrepancies may be mostly caused by the various estimating methods employed and impacting factors considered. We have re-written section 3.2.2, which is shown as follows:

" The ammonia emissions from N fertilizer use in China have been estimated for different base years by different methods. The results of comparisons between this study and some previous studies are listed in **Table 3**. In order to make the inventories comparable, we updated the emissions in different years to the year of 2011 based on the changes of fertilizer use, temperature and precipitation, as described in the supplementary materials. As presented, the results of this study are generally equivalent and comparable to the researches of Zhang et al. (2011) and Huang et al. (2012b), which is 60-70% lower compared with other studies. The discrepancies are mostly caused by the various estimating methods and emission factors employed. Streets et al. (2003), Dong et al. (2010) and Zhao et al. (2013) used averaged emission factors for all agriculture in China and did not consider the impacts of environmental parameters, e.g. soil pH, precipitation, etc. For example, the emission factors for urea used by Streets et al. (2003), Dong et al. (2010) and Zhao et al. (2013) are 15%/20% (temperate and tropical ozone). However, the basic emission factors for urea used by Huang et al. (2012b) are 8.8% for acid soil and 30.1% for alkaline soil. The agricultural regions in China are dominated by acid soil (http://www.soil.csdb.cn/), so this value is lower by nearly 50% compared with the averaged emission factors. In addition to soil pH, precipitation can also decrease NH_3 emissions, because precipitation can increase the water content in soil and fertilizer N can be leached to a deeper soil layer by water (Wang et al., 2004). Zhang et al. (2011) adjusted the emission factors by 0.75, 0.80, 0.85,0.90, 0.95 and 1.0 for significant rainfall events (>5mm in 24h)within 24h, 24-48h, 48-72h, 72-96h, 96-120h and >120h of fertilizer application. In this study, the impacts of soil pH and precipitation on NH_3 emission are considered by impacting soil gamma and resistances, as shown in section 2.3. In addition, our study and Zhang et al. (2011) include the impacts of irrigation. The experiments of Wang et al. (2004) in Beijing for winter wheat-summer maize cycle have shown that NH_3 volatilization is reduced after irrigation and revealed a low emission factor value of 2.1-9.5%.

Figure S4 and S5 represent the comparisons of provincial distributions and seasonal variations of these different NH_3 emission inventories. The provincial distributions are similar, and the emissions from Henan, Shandong, Jiangsu, Hebei and Anhui dominate the country annual total emissions. At the same time, some discrepancy also exists for the specific province between different studies, which may be caused by distinct fertilizer consumptions and emission rates employed. For example, for Henan province, the estimation of Huang et al. (2012b) is the highest among these studies. The possible reason is that alkaline soil is dominant in Henan and Huang et al. (2012b) set a uniform high emission factor for alkaline soil, which is twice as high as that in Dong et al. (2010). Compared with provincial distributions, the difference of seasonal variations among these studies is larger. The seasonal profile in Zhao et al. (2013) is based on temperature variations. In addition to temperature, others also considered the impacts of fertilizer application timing. It is indeed difficult to capture entirely the exact date of fertilizing for the whole China, which may bring this large diversity. For example, Huang et al. (2012) thinks that the basal-dressing and top-dressing fertilizer of winter wheat are conducted in September and November. However, the basal-dressing fertilizer is applied in October in this study and Zhang et al. (2011), and the top-dressing fertilizer is mainly used in March of the next year. The diversity of seasonal variations among different studies reflects that large uncertainties still exist for the temporal distribution of NH_3 emissions and much local research work still need to do."

Regarding data availability, it is unclear to me which datasets on Chinese agricultural practices (fertilizer use, crop use, etc.) are available to the wider public so that future studies may improve upon this study. One of the citations was a thesis study in Chinese(Zhang, 2008) – is this available to the broader community? What about other data sources?

Response: Thank you for comments. In this study, the data of fertilizer use rate and crop is from province-level or city-level statistical yearbooks. These statistical yearbooks are publicly available. We have collected and processed these data to build the dataset. The integrated database crop area by county can be accessed by contacting the corresponding author and we have added a note in the revised manuscript. In addition, we also used some information in the literatures, like a thesis study in Chinese (Zhang, 2008) you mentioned. All these literatures are publicly available (http://www.cnki.net/). The major information used about fertilizer use has been listed in Table 1 and 2 of the manuscript.

Finally, the manuscript needs a thorough read by a native English speaker. There are many cases with extra or missing definite articles (‘the’). Clauses starting with “which” are frequent, sometimes properly and many times improperly. The co-authors from the US should be able to provide and correct these oversights or questionable grammar aspects.

Response: Thank you for comments. We have asked our co-authors from the US help on the grammar aspects.

Particular details/comments:

1. First sentence of the abstract regarding ammonia’s importance to atmospheric chemistry – I would be more specific here and emphasize its importance in aerosol composition/chemistry instead. Gas phase atmospheric chemists have long ignoredNH3 because it really doesn’t matter. While I agree on balance it is very important, for the first sentence in the abstract I would refine the focus slightly.

Response: Thank you for comments. We have revised the sentence to "Atmospheric ammonia (NH_3) plays an important role in atmospheric aerosol chemistry "

2. p. 748, line 5: inorganic aerosol is more convention, rather than “non-organic”

Response: Thank you for comments. We have revised it in the revised manuscript.

3. p. 749, lines 4-7: “: : :correction factors are empirical and too simple.” More elaboration is needed here – what parts of the emission factors and why are they not appropriate?

Response: Thank you for comments. We agree with you that it is not appropriate to give a vague statement. After a further analysis, we think that the discrepancies may be mostly caused by the various estimating methods employed and impacting factors considered. We have re-written section 3.2.2. Please see the reply for the second general comment or section 3.2.2 in the revised manuscript.

4. Section 2.2.1: Is there a database of the cropland area for 2710 counties? If not available publically, can the authors post their datasets in such a location for others?

Response: Thank you. There was no existing database containing all the counties when we did this work and the information is disperse in each province-level or city-level statistical yearbook. Data of cropland area for each crop in each county was collected and processed based on these statistical yearbooks to build the database. We have added a note in the revised manuscript and the integrated database crop area by county can be accessed by contacting the corresponding author.

5. Section 2.2.2 and 2.2.3: since everything is related to the 36 x 36 km CMAQ grid, also specify in parentheses the scale in km after the native unit resolutions for the soil database (arc-second) and MERRA (0.5 degree x 0.667 degree) to help the reader

Response: Thank you for comments. We have revised the description in the revised manuscript. For soil database, 30 arc-second is about 1km maximally. For MERRA, 0.5 degree x 0.667 degree is about 55 x 75km maximally.

6. Section 2.4.4: It is unclear why the EPIC model uses heat units in some cases and unpublished data in other cases. What is the justification to selectively use on metric over the other? Why not use the best database, which appears to be the unpublished Chinese Academy of Sciences research? Can the authors provide a link to that data used in this study?

Response: Thank you for comments. In this study, the heat-unit scheduled timing method was dominant, because the date of application would vary with crop, local soil and weather conditions leading to more spatially and temporally resolved application estimates. Differently, the unpublished Chinese Academy of Sciences research estimated a fixed time range, lasting several weeks to 1 month. It played a subsidiary role to limit the application date to the fixed range. In addition, we have provided a note of personal communication for the unpublished materials in the revised manuscript.

7. Section 3.2.2 (Comparison to other studies) was lacking, as noted in my most significant

comments above. No detailed discussion of the differences between studies are noted, other than a generalized statement that "the parameters were set based on conditions in UK, which may be very different from China". Likewise, a similar statement is made about US experiments. What happens the parameters were similar to the US, Europe, or UK? Then how can one explain the different results?

Response: Thank you for comments. We agree with you that it is not appropriate to give a vague statement. After a further analysis, we think that the discrepancies may be mostly caused by the various estimating methods employed and impacting factors considered. We have re-write section 3.2.2. Please see the reply for the second general comment or section 3.2.2 in the revised manuscript.

8. Section 3.3, CMAQ and ground observations: Is the assumption that if CMAQ models NO3- correctly that NH4+ would also be OK? This argument is flawed. For example, see Schiferl et al., JGR, 119, 1883-1902, 2014 on the difficulties of modeling NO3- and NH4+ with other constituents and trying to match observations. I found this entire section to be speculative and not add much to the paper. I suggest removing or significantly revising.

Response: Thank you for comments. The assumption here is not that if CMAQ models NO_3^- correctly that NH_4^+ would also be OK. NH_4^+ is the dominant positive ion in the atmosphere and the changes of NH_3 emissions can affect the modeled NO_3^- aerosol concentrations. The assumption is that the more reasonable the estimated NH_3 emission is, the better the NO_3^- aerosol concentrations are modeled. In the research of Schiferl et al. (2014) you mentioned, he has a conclusion that uncertainties in the simulation of the inorganic gas-particle system are dominated by emissions, which is consistent with our point. Therefore, we have kept this part and clarified the purpose of this comparison further in this part.

9. Section 3.4: The uncertainty analysis here is actually just a qualitative discussion of potential biases, and a discussion in the most general of sense. This section needs to be greatly expanded so others can assess how changes in the input parameters will result in changes to the total emissions. A full sensitivity analyses is needed here, particularly if datasets are not readily available. Otherwise, it will be difficult for others to ever compare to this study

Response: Thank you for comments. We agree that uncertainty analysis is important and beneficial. More detailed uncertainty analysis for the major impact factors were added to section 3.4, which is as follows.

" This is a pilot study to apply this model system to estimate the NH_3 emission in China and large uncertainties still exist for this method at some aspects. Quality of input data, mathematical algorithm, and parameters applied in EPIC and the bi-directional model may be associated with uncertainties in the model output.

Fertilizer application rates for each crop are important input data for the estimation of NH_3 emissions from agricultural fertilizers. They are obtained from the agricultural statistics. These statistical data should have some level of uncertainty, because the amounts of samples in the census are limited. Beusen et al. (2008) has employed an uncertainty of ±10% for the statistical data of fertilizer use based on expert judgments when estimating the global NH_3 emission. A June 2006 sensitivity run of this bi-directional model in US showed that a 50% increase of crop fertilizer use would result in a 31% increase in NH_3 emission (Denniset al., 2013). In addition, the spatial distribution of NH_3 emissions from agricultural fertilizer is strongly related to cropland area and its distribution, which are achieved from the MODIS data. Friedl et al. (2010) mentions that the producer's and user's accuracies are 83.3%/92.8% for MODIS class 12 (cropland) and 60.5%/27.5% for class 14 (Cropland/Natural Vegetation Mosaic) in MODIS Collection 5 product. This would lead to the uncertainties of spatial distribution. Additionally, due to the limit of data availability, the initial characteristics of the dominant soil in each grid are gotten from the US dataset. Although we have matched the soil based on soil type, eco-region, and latitude, uncertainties still existed due to different long-term agriculture management.

Seeing from the algorithm described in section 2.3, the EPIC outputs, including soil NH_4^+ concentration, soil volumetric water content (θ_s) and soil pH, are important inputs of the bidirectional module. EPIC has been used and evaluated world widely to simulate nitrogen cycle and soil water. Some validation studies have found favorable results for soil nitrogen or/and crop nitrogen uptake levels (Cavero et al., 1998 and 1999; Wang et al., 2014). However, less accurate simulation results were also reported (Chung et al., 2002). For soil volumetric water content, Li et al. (2004) found that EPIC model could catch the variation of soil water in different years well with the relative bias of 11.7%, and the research conducted by Huang et al. (2006) also showed that the EPIC-simulated long-term average θ_s values were not significantly different from the measured values in the Loess Plateau of China. For soil pH, the normal growth pH range of three dominant crops (rice, corn and wheat) is 6.0-7.0 (http://njzx.mianxian.gov.cn/xxgk/ccpf/20804.htm; http://nmsp.cals.cornell.edu/publications/factsheets/factsheet5.pdf). The 95% confidence interval of EPIC simulated values is 6.3-7.6, which is reasonable and acceptable although uncertainties still exist.

The bi-directional ammonia flux module in the CMAQ is the core of this model system. The uncertainties of the bidirectional exchange parameterization would bring uncertainties to NH_3 emission estimates. Pleim et al. (2013) compared the simulated NH_3 flux from the box model of this ammonia bi-directional flux algorithm with observations in three periods. The results showed that the model generally reproduced the observed series and significantly correlated with the observations (p<0.001). The mean normalized biases were 78.6%, -49% and 1% for soybeans (18 June-24 August, 2002), corn (21-29 June, 2007) and corn (11-19 July, 2007), respectively. The soil gamma (Γ_g) and appoplast gamma (Γ_s) are two important parameters in this ammonia bi-directional flux algorithm (Bash et al., 2013) and their parameterization remains uncertain (Massad et al., 2010). The field measurements of Γ_g and Γ_s are limited, and measured values are scattered owing to complex impact factors (Massad et al., 2010 and reference therein). Dennis et al.(2013) assessed the effects of these uncertainties. A 50% increase of Γ_g would result in a 42.3% increase in NH_3 emission. Two different parameterization methods of Bash et al.(2013) and Massad et al. (2010) could lead to a 17% change in NH_3 emission.

It's very difficult to give an uncertainty interval accurately for this method, because there are many factors contributing to this model system. Here, an uncertainty of about ±50% is considered appropriate based on the above analysis, which is also the upper limit of uncertainty in previous studies (Bouwman et al., 1997; Zhang et al., 2011; Zheng et al., 2012). Therefore, the NH_3

emission from agricultural fertilizer application in China of 2011 is in the range of 1.5-4.5Tg. In order to reduce the uncertainty, much work still need to do. In addition to improve the quality of input data, additional local measurements of soil and vegetation chemistry, ambient NH_3 concentration and flux data are needed to enhance and evaluate the parameterizations of EPIC model and bi-directional module."

10. Conclusions: What are the larger implications of your work? If NH_3 is less from fertilizer than others report, does this mean that animal systems are more important to controlling ammonia? What is the comparable scale between such inventories? Some larger perspectives are needed here.

Response: Thank you for comments. Our study just estimated the NH_3 emission from N fertilizer use, but not including from animal systems. As described in section 3.2.2, the results of this study are generally equivalent and comparable to the research of Zhang et al. (2011) and Huang et al. (2012). In the research of Huang et al. (2012), the NH_3 emission from N fertilizer use accounted for 33% and that from animal systems accounted for 54%. Although the percentage is smaller, controlling the NH_3 emission from N fertilizer use is still important. In addition, we think that the major significance of our work includes the following two parts:

(1) For the first time, the NH_3 emissions from fertilizer using in China are estimated using a recently-developed modeling framework in which agricultural activity can be parameterized by meteorological, crop, and soil data. The NH_3 emissions can be calculated as the air quality model progresses through time and the temporal resolution is improved to hourly. The higher resolution of NH_3 emission is good for modeling and exploring the impacts of NH_3 emission on air quality. In addition, the results can be utilized for a better comparison of novel and traditional method. This is also an important contribution to the scientific literature

(2) This model system tightly links agricultural, weather and atmospheric processes, and allows for some interesting future research. For example, the model makes it possible to reflect the impacts of long-term climate and agricultural management changes on the NH_3 emission and atmospheric aerosol. If linking it to a water quality and transport model, the impacts of atmospheric nitrogen deposition from CMAQ and nutrient run off from EPIC on the water eutrophication can be estimated. This is the first try to apply this model system to China, and it's also the foundation to explore more scientific researches in the future.

We have added the additional description in the conclusion part.

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