Reply to Referee #2's Comments

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

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.

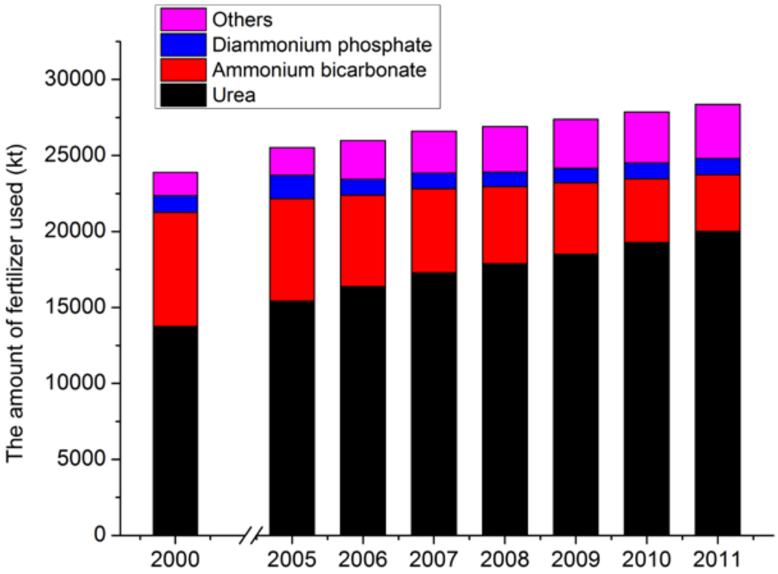

Fig.R1. The amount of different fertilizer types used in China from 2000 to 2011

(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:

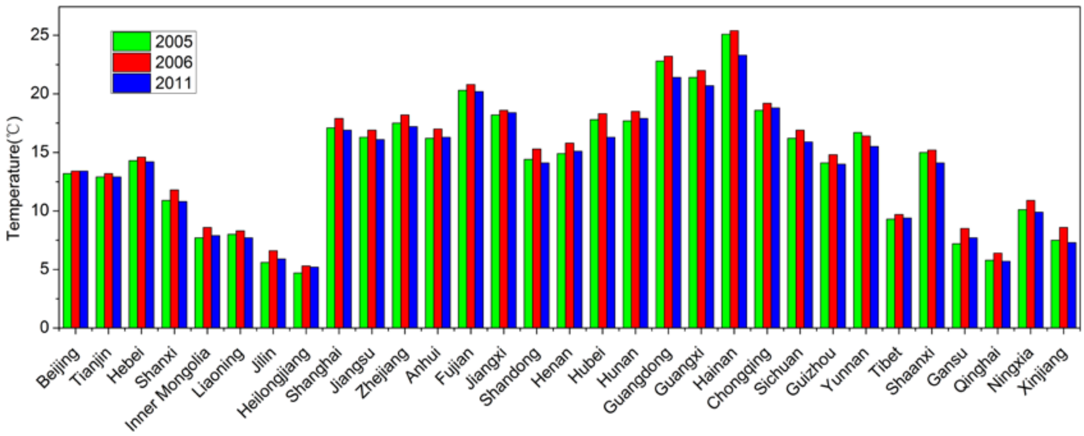

Fig.R2. The provincial temperatures in the year of 2005, 2006 and 2011

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:

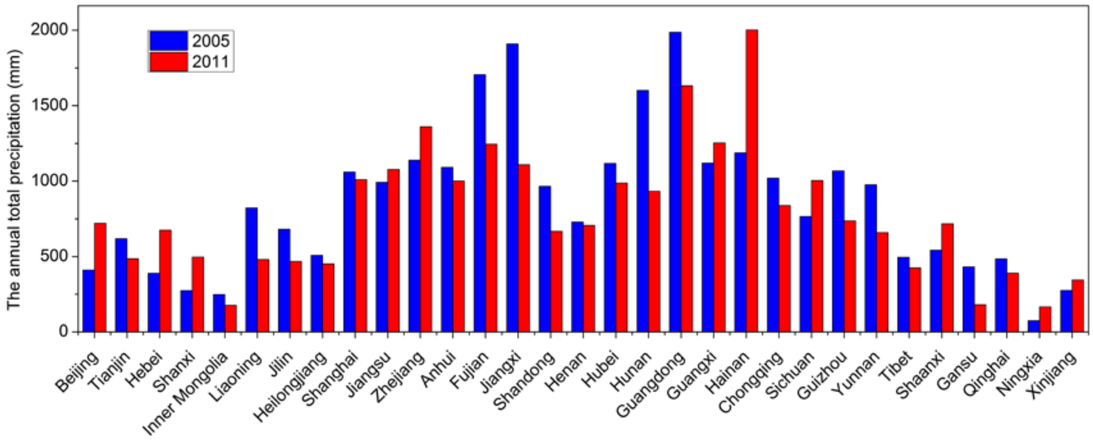

Fig.R3. The provincial precipitations in the year of 2005, 2006 and 2011

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.R1. 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.1 and Table 1, respectively. It can be seen that the model performance for HNO_3 is acceptable.

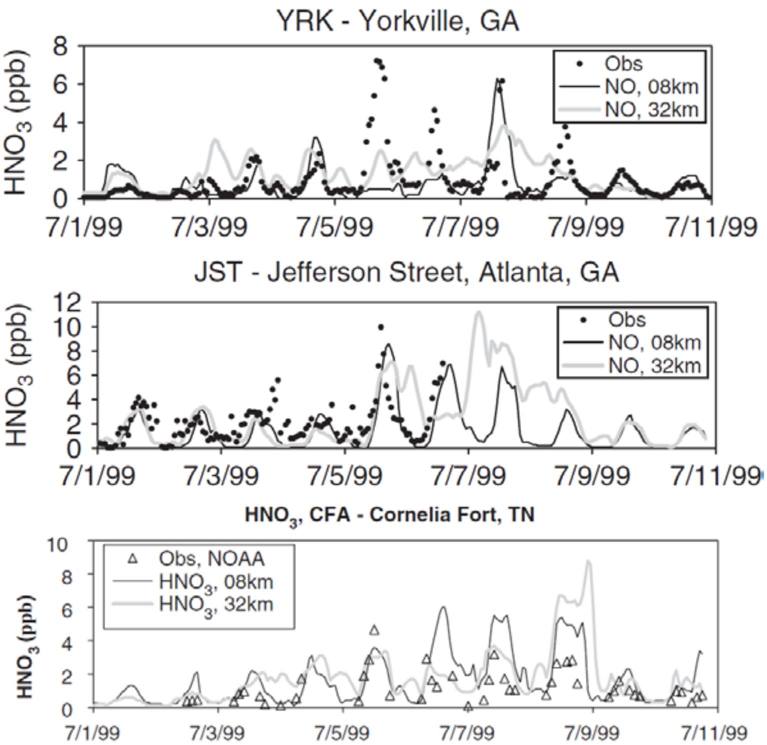

Fig.R4. The comparisons of observed and predicted HNO3 in Zhang et al.(2006)

Table R2. 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.R3. 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.R4. 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.R5. 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.

References cited in this response:

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