Reply to Referee #3'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

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.

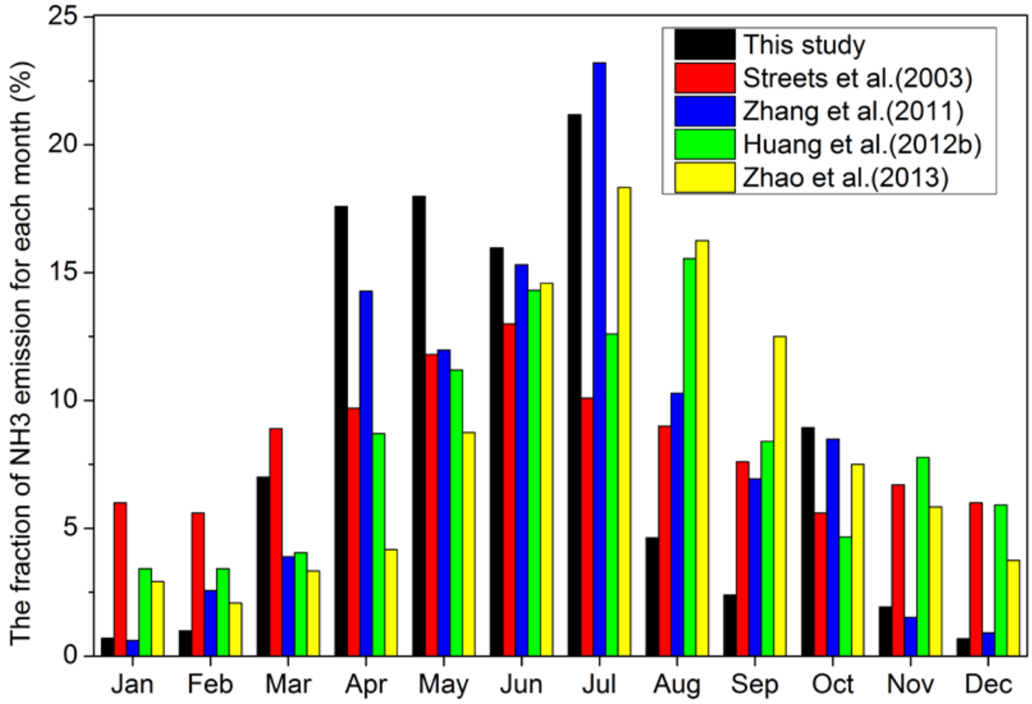

Fig.R1. Comparison of monthly NH_3 emissions from N fertilizer use in different studies

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.

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