Reply to Referee #1'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. 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.S1**(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.

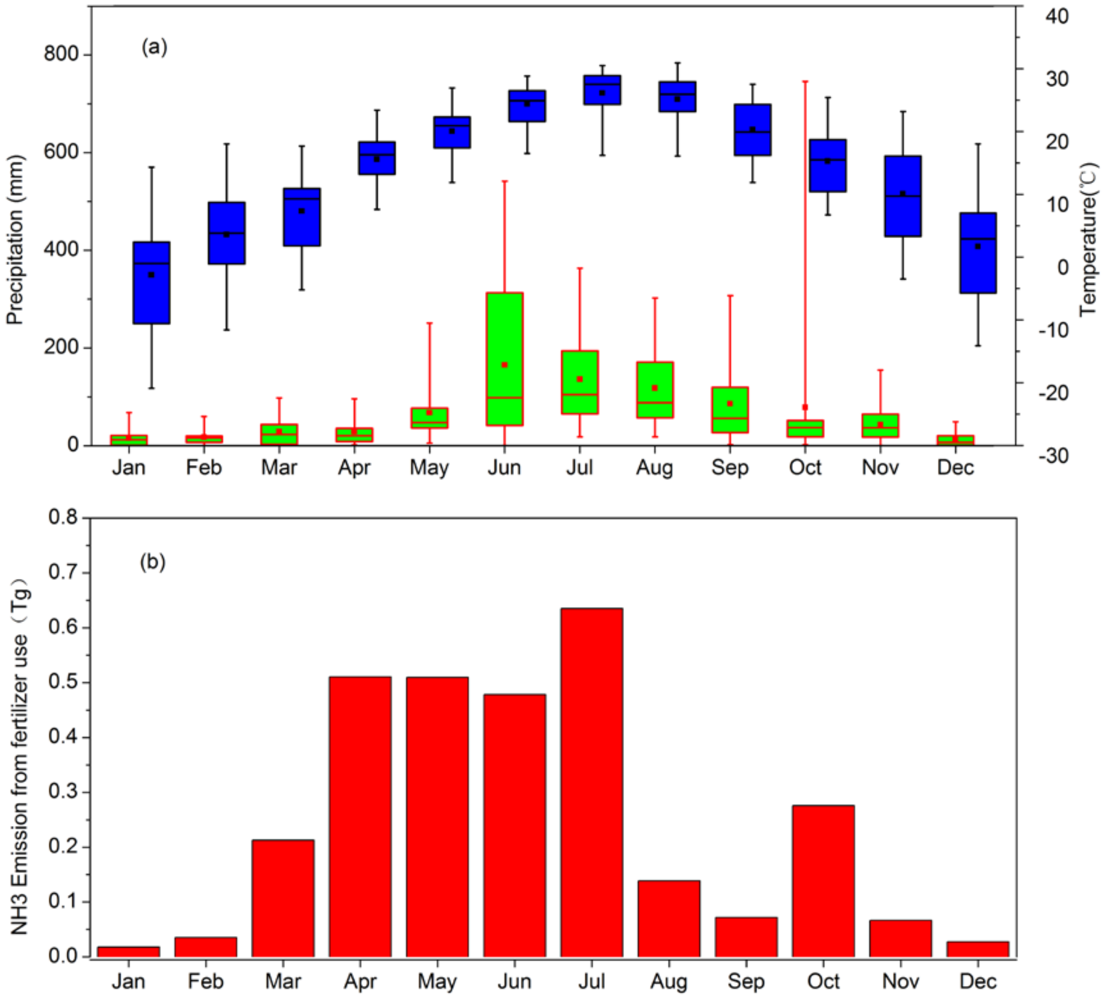

Fig.R2.(a)The variation of monthly precipitation (green) and temperature (blue) in 31 provinces. In the box-whisker plots, the boxes and whiskers indicate the 100th (max), 75th, 50th (median), 25th and 0th(min) percentiles, respectively. The point represents the average value. (b) Monthly NH_3 emissions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

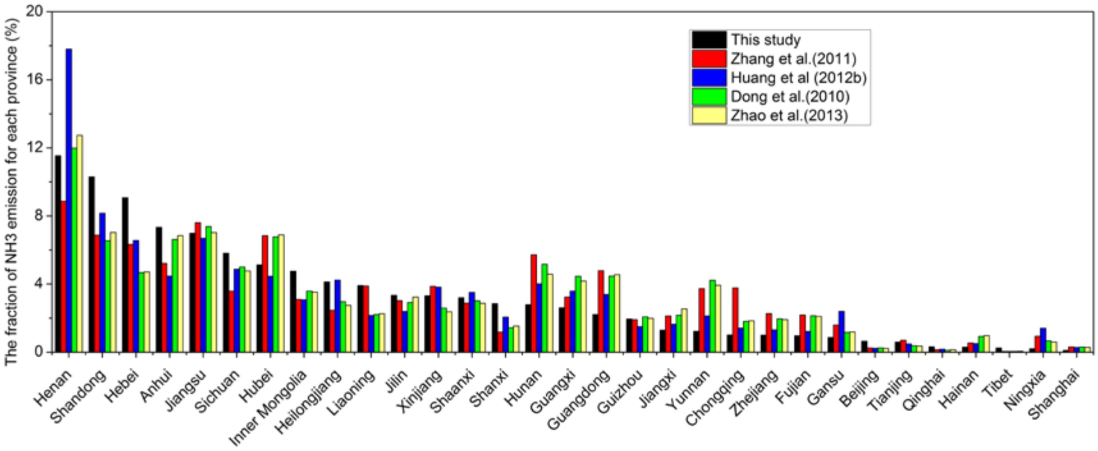

Fig.R3. Comparison of provincial NH_3 emissions from N fertilizer use in different studies

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