Response to Referee #2

This is an interesting paper describing an extension of the earlier inventory published in 2012 (Huang et al. in Global Biogeochemical Cycles) towards a longer time period and with a few modifications of the emission factors used. The approach uses gridded emission factors on a monthly basis, and that is an important improvement compared to annual emissions. This way atmospheric chemistry-transport models can better simulate the fate of the atmospheric ammonia and aerosols. Hence, I think this paper is an important contribution. However, there are a few issues that need to be improved in terms of method description. The paper as it is now does not provide sufficient information to understand how the inventory has been constructed, and only by going back to the 2012 publication readers can understand how the monthly emission factor shave been calculated.

Response: We would appreciate the referee very much for providing the insightful comments, which help us to improve the manuscript greatly.

For example, it is not clear how the emission factor for synthetic fertilizer use has been calculated. My guess is that the authors have a crop calendar with data on timing of fertilizer application. The 2012 paper mentions that a range of different crops are considered, but this information is missing in the current text. Also, it is not clear how then, for fertilizer use in a specific month, emissions are calculated; is it a flush, or is the emission extended over a longer period (the same comment is for all other sources)?

Response: Accepted. It was reworded in Lines 163−181. A crop calendar covering timing of the growth of various crops in each province was used to derive the monthly changes of fertilizer application. 16 kinds of crops cultivated were involved in the activity data and their fertilization timings are generally fixed. Consequently, monthly condition-dependent emission factors were calculated based on timing of crop growth and practice of fertilizer application. In this work, the monthly emissions for all sources were the product of monthly activity data and corresponding EFs. We supposed that these emissions were produced instantaneously from related activities in current month and couldn't extend to next month. More descriptions about the calculation of monthly emissions are listed in Lines 271−282.

Revision in Lines 163–181 on Page 7: "A crop calendar, which involves the type of crop cultivated at specific region and corresponding fertilization timing was used to derive the monthly fertilizer application. We considered 16 kinds of crops in the activity data that are wildly cultivated in different seasons in China, including early rice, semi-late rice, late rice, non-glutinous rice, wheat, maize, bean, potato, peanut, oil crop, cotton, beet, sugarcane, tobacco, vegetables and fruits. We derived monthly condition-specific EFs for synthetic fertilizer volatilization by introducing several influencing factors like the type of fertilizers, soil pH, ambient temperature, fertilization method, and application rate (see Table 2 in (Huang et al., 2012b)). Briefly, EFs are characterized by fertilizer types with ABC and urea more volatile than the other fertilizers. Liner relationships between the volatilization of mineral fertilizers and soil pH were developed to correct EFs (Fan et al., 2005; Bouwman et al., 2002). A threshold of 200 kg N ha^{-1} was defined as the high fertilization rate and when the local fertilization rate exceeded this value, we multiplied EFs by 1.18 (Fan et al., 2006). We derived the relationship between the emission rate and temperature for various fertilizers from EEA (2009) and Lv et al. (1980). Compared to Huang et al. (2012b), the effects of wind speed and in-situ measurements of NH_3 flux conducted by our research group in a typical cropland were involved to further refine the EFs for synthetic fertilizer emissions in this study."

<u>Revision in Lines 271–282 on Page 11:</u> "The seasonal NH_3 emission estimation for fertilizer application could be calculated as the product of condition-specific EFs derived from meteorological factors (average monthly temperature and wind speed) and monthly fertilizer consumption associated with agricultural timing. For livestock emissions, we assumed that the number of each livestock category per month remains constant, because the monthly fluctuation in the production of meat, eggs and milk is very small (http://www.caaa.cn/). The monthly EFs were distinguished by average monthly temperature and wind speed from NCEP. Besides, the emission from biomass burning also shows a temporal fluctuation. MCD45A1 (monthly burned area product), MOD14A2 and MYD14A2 products (8-day thermal anomalies/fire products) were utilized to ascertain the timing of different kinds of biomass. For other minor sources, the emissions were equally divided into 12 months."

In section 2.2 on livestock waste there is the same problem of lack of information to understand the approach. In addition, I wonder if wind speed is also used for this source, since it is also soil-borne for grazing and spreading –related emissions. I also wonder how the authors can assume that the parameters used to compute TAN have not changed. I guess that the feeding situation has changed in the inventory period, so that the composition and amount of manure or N excretion per kg of product or per animal probably has changed significantly. So a brief discussion on the impact of this assumption is needed.

<u>Response:</u> Accepted. We add more detail description. Please see Lines 218−246. Actually, wind speed was not involved for grazing and manure spreading in the previous manuscript. In this revised version, we recalculated the livestock emissions within the grazing and spreading stages by multiplying an exponent term with original EFs, similar with the modification in the EFs for mineral fertilizers (please see Lines 250−255). Finally, the livestock emissions have been updated in the revised text.
The feed situation in Chinese agricultural has been changed, e.g. animal housing conditions or feedstuff types, and how to parameterize these changes is difficult. However, based on the statistics year book for husbandry, we still introduced an interannual ratio between the free-range system and the intensive one to reflect the change of animal rearing types in the inventory periods. It could represent the changes

of Chinese livestock practice in the inventory period to some degree. They were discussed in the manuscript (Lines 619−632).

<u>Revision in Lines 218–246 on Pages 9−10:</u> "In this study, TAN inputted into manure management was the product of the daily amount of urine and faece produced (kg/[day*capita]), N content (%), and TAN content (%) (see Table 3 in (Huang et al., 2012b)). We assumed that these parameters have not changed during the 30-year period and some uncertainties from this assumption would be discussed in Sect. 3.5. We estimated livestock emissions by multiplying TAN at four different stages of manure management: outdoor, housing, manure storage, and manure spreading onto farmland (Pain et al., 1998) with corresponding EFs. In the outdoor stage, the excreta were directly deposited in the open air without any treatment after that while animals' excrete inside buildings would release emissions during housing, storage and spreading stages. The periods spent in buildings in a year for different livestock classes were used to determine the portion of excrement indoors or outdoors. After a proportion of TAN was depleted through some processes like immobilization, discharge of NH_3, N_2O and N_2, or the leaching loss of nitrogen, the rest TAN would flow into next stage (EEA, 2013). In addition, we also considered three main animal-rearing systems in China: free-range, intensive, and grazing. The first two systems are extensively implemented in most rural areas of the country. The free-range system is characterized by small-scale rearing belonging to individual families and has been rapidly developed over recent decades (http://www.caaa.cn/). Based on animal husbandry yearbooks, we defined an intensive rearing system as that where the number of a single livestock class on a single farm (except grazing) was larger than a certain value (Table S1). Under this definition, an interannual ratio between the free-range system and the intensive one was introduced to reflect the change of animal rearing types in the inventory periods. It could represent the changes of Chinese livestock practice in the inventory period to some degree."

<u>Revision in Lines 250–255 on Page 11:</u> "Temperature-dependent volatilization rates were considered by using specific EFs at different temperature intervals in the manure housing stage (Koerkamp et al., 1998). We also implemented wind speed and temperature adjustment in the stages of manure spreading and grazing, based on model results reported by Gyldenkaerme et al. (2005). Ambient temperature and wind speed data were also extracted from NCEP final analysis dataset."

<u>Revision in Lines 617–630 on Page 25:</u> "For example, the feed situation in Chinese agricultural has been changed, e.g. animal horsing conditions, feedstuff types or feeding periods. Zhou et al. (2003) conducted rural household surveys on the Chinese household animal raising practices. They found that in some provinces like Zhejiang, industrial processed feed had become a major animal feed. The industry processed feed is easy to digest and absorb, showing more use efficiency than traditional farm-produced forage. Therefore, the amount of N excreta per animal feed by industry forage should be less than that by farm forage. But Li et al. (2009) investigated that compared with 1990s, the average N content in manure from pig, chicken, beef and sheep has little change in recent years according to nationwide 170 samples analysis. On the other hand, rearing periods for animals like poultry were significantly reduced during recent years along with the development of breeding technology, that is, manure excreted per animal per year was supposed to be declining. However, this change was not considered in this study and it may result in overestimation of livestock emissions in recent years."

Finally, I wonder how the authors can use monthly temperatures and monthly wind speed as a factor in the calculation of the emission factors. How representative are monthly mean wind speed and temperature, while perhaps maximum day temperature and variability of wind speed are better predictors of NH3 emissions. In addition, there may be an interaction between temperature and wind speed that is not represented in the emission factor approach.

Response: Yes. It could be better to use daily temperature and wind speed for estimation. However, in this study, the daily activity data is not available (we could not quantify the synthetic fertilizer use each day), or we could not identify the exact date of fertilizer application and the timing varies annually. We adopted monthly mean temperature to parameterize the emission factor according to EEA (2009). Despite the uncertainties, we still use the monthly mean temperature and wind speed to produce monthly emission inventories. Please see Lines 208−215 in the revised text. We admit there might be some interactions between temperature and wind speed. Some discussion on this uncertainty in Section 3.5 in the revised manuscript (Please see Lines 599−608).

Revision in Lines 208–215 on Page 9: "It should be note that in this study, we used monthly weather values in the adjustment of EFs rather than the daily maximum since the daily activity data was not available (we could not quantify the synthetic fertilizer use each day) or we could not identify the exact date of fertilizer application and the timing varies annually. On the other hand, we adopted the parameterization of temperature adjustment provided by EEA (2009), which is also based on mean temperature. Despite the uncertainties, we still used the monthly mean temperature and wind speed to produce monthly emission inventories."

Revision in Lines 597–606 on Pages 24−25: "In this study, the impacts of wind speed and ambient temperature on the EFs in agricultural ammonia emissions were isolated but in real condition, there might be some interactions between temperature and wind speed. Ogejo et al. (2010) indicated that parameter interactions may play a significant role in emission estimation with a process-based model for ammonia emissions but they also didn't consider the interaction between temperature and wind velocity. Actually, previous studies generally examined the respective effect of wind speed and temperature on ammonia volatilization according to controlled experiments (Sommer et al., 1991) and we expect more experimental evidences for the interaction effect."

In relation to this I wonder if this inventory is better than the one published earlier. Have the authors tested this claim. I am asking this, because the emission estimates are quite close, and given the uncertainties I wonder if the modifications are really improvements. Also the claim that the approach of this paper is better or more realistic than emission factors that are based on less factors needs some more thinking. I wonder if the authors can show that this is the case. Does the approach of this paper result in a better comparison with Paulot et al. (2014) and Van Damme et al. (2014) than the previous version of Huang et al. (2012) and of less sophisticated emission factor approaches? In my opinion the claim that a model is better needs to be supported by evidence.

Response: Accepted. Indeed, the emission amounts are close between the two results though some modifications have been done in the present study. It is not surprising because we mostly followed Huang et al.'s method. It should be noted that in our study, we mainly focused on compiling a long-term emission inventory rather than developing a new methodology for NH_3 emission. It covered more than 30 years. These comprehensive multi-year inventories are very necessary for global and regional air quality modeling, especially for studying the impacts of NH_3 emissions on air quality.

Moreover, some modifications are still needed to improve the emissions as some potential problems existed in Huang et al.'s method (listed in Lines 124−129 in the introduction part). First, we used wind speed in the adjustment of emission factors. Secondly, we applied the NH_3 EF which was measured by using micrometeorological method for whole a year in a typical farmland in North China Plain by our research group (Huo et al., 2014, 2015). The in-situ results could represent better than those used in Huang et al. which were derived from studies in early years.

Paulot et al. (2014) estimated the average annual ammonia emission of 10.4 Tg in the period of 2005−2008. For the same period, our result 10.2 Tg, and Huang et al. (2012b) estimated 9.8 Tg in 2006. The three results are close. The excellent qualitative agreement for spatial distributions could be found between our estimations and IASI sensor (Van Damme et al., 2014). The comparisons have been discussed in the revised manuscript (please see Lines 583−590).

Revision in Lines 124–129 on Pages 5−6: "However, there were still some problems in this method; for example, Huang et al. generally adopted EFs reported in early years and up-to-date in-situ measurement was needed; moreover, wind speed that could be of importance in emission estimation was not considered in that study. Though Huang et al. have involved as many NH_3 emitters as possible, some minor sources may be neglected like fertilization in orchard, NH_3 escape from thermal power plants."

Revision in Lines 581–588 on Page 24: "Paulot et al., (2014) estimated the annual NH_3 emissions of 10.4 Tg using a global 3D chemical transport model averagely in 2005−2008 while our result was 10.2 Tg for the same period and Huang et al. (2012b) estimated 9.8 Tg in 2006. The three results are quite close. We also found excellent qualitative agreement for spatial distribution between our estimation and the global NH_3 column retrieved by IASI sensor (Van Damme et al., 2014). Several emissions hotspots shown in this study, including the North China Plain, Sichuan and Xinjiang provinces (near Ürümqi and in Dzungaria), and the region around the Tarim Basin were also detected by the IASI sensor."

Finally, I wonder why the authors have tried to generate monthly emissions, but nowhere discuss the temporal variation (likewise, the 2012 Huang et al. paper also lacks such a discussion). It would also be interesting to test if the temporal patterns are changing with the shifts in the different sources?

Response: Accepted. A subsection discussing the temporal variation has been added in Sect. 3.3 (please see Lines 468−531).

Revision in Lines 468–531 on Pages 19−22: "The temporal patterns of NH_3 emissions in 1980, 1990, 2000, and 2012 are clearly presented in Fig. 6 (a). The emissions were primary concentrated during April to September due to the intensive agricultural activities and higher temperatures. Specifically, the different sources showed the diverse distribution characteristics.

Figure 6 (b) describes the temporal distributions of NH_3 emissions from synthetic fertilizer application in 1980, 1990, 2000, and 2012, respectively. It is obvious that the monthly emissions from fertilizer exhibited similar seasonal distribution among different years. Generally, the largest emissions were occurred in summer (June to August), accounting for 44.8–47.7% of annual emissions from synthetic fertilizer, which are attributed to denser fertilization and higher temperatures during this time. Conversely, because of the less NH_3 volatilization related to lower temperatures and relatively rare cultivation during the winter (December to February), the NH_3 emissions reduced to 7.7–11.1% of annual fertilizer emissions. In China, the new spring seeding begins in April and is accompanied by corresponding fertilizer application. In the following 1–2 months, due to application of top fertilizer and warming temperatures, particularly in eastern and central provinces such as Jiangsu, Anhui and Henan, the NH_3 emissions continuous increase to August. In the North China Plain, the winter wheat-summer maize rotation system has practiced as a characteristic farming practice. The high emission rates in June and August could be attributed to the basal dressing and top dressing of summer plants, such as maize. From autumn on, most of the crops begin to harvest, which lead to the decline of emissions during this time. In particular, winter wheat is usually seeded in September with the application of basal dressing, and the top dressing is applied two months later, which could be responsible for the peak emissions occurred during September and November. Besides, owing to more temperature fluctuations and fertilizer application, the monthly distribution of emissions in the northern regions was more remarkable than that in the southern.

The significant seasonal dependence of NH_3 emissions from livestock wastes in different years can be clearly seen in Fig. 6 (c). The monthly distributions of NH_3 emissions were highly consistent with the variations in temperature under the premise of the constant animal population among the different months we assumed above. The major emissions occurred in warmer months (May to September), and more than 45% of the annual livestock emissions, which could be explained by more NH_3 volatilization related to substantial increase of temperature. In contrast, the lowest NH_3 emissions from livestock wastes were estimated in winter (December to February), and this is attributed to relatively smaller EFs linked to lower temperatures. Apart from the two major sources, the NH_3 emissions from biomass burning also had distinctly temporal disparities in spite of the relatively small contribution of total emissions.

The temporal variation of emissions from crop burning in fields from 2003 to 2012 (when the annual MODIS thermal anomalies/fire products (MOD/MYD14A1)) were available) is described in Fig. 6 (d). The occurrences of crop burning in fields were concentrated in March to June with another smaller peak in October, which are consistent with local sowing and harvest times (Huang et al., 2012a). The highest emissions rates occurred in June are mostly attributed to the burning of winter wheat straw that fertilizes the soil after the harvest (in the end of May) in the North China Plain. The peak in October can be partly explained by the burning of maize straw after the harvest (in the end September) in the North China Plain. In addition, the south China including Guangdong and Guangxi provinces have two or three harvest times every year. The sowing time for crops here begins in March, when crop residues would be burned to increase the soil fertility. Simultaneously, in northeast China, there is the local farming practice of clearing the farmland before sowing in April, which may emit corresponding NH_3 during the spring. In winter, the mature period of late rice in south China lead to a certain amount of NH_3.

Figure 6 (e) displays the seasonal distribution of NH_3 emissions from forest and grass fires from 2001 to 2012 (when the annual MODIS burned area product (MCD45A1) was available) in China. The weather and vegetation conditions are regard as dominate factors that regulate the fire activity (Perry et al., 2011). The fire emissions were primarily concentrated in February to April and August to October, because of scarce precipitation, high wind speed and gradually rising temperature during early spring and late winter, especially in the southwestern regions. Simultaneously, the lower moisture content of vegetation is favor of burning. In addition, the abundant fallen leaves and crop residues in autumn could make contributions to the fires dramatically. It is worth noting that as the global warming, the number and magnitude of forest and grass fires are increasing and the seasonal feature of fires emissions would weaken in the future."

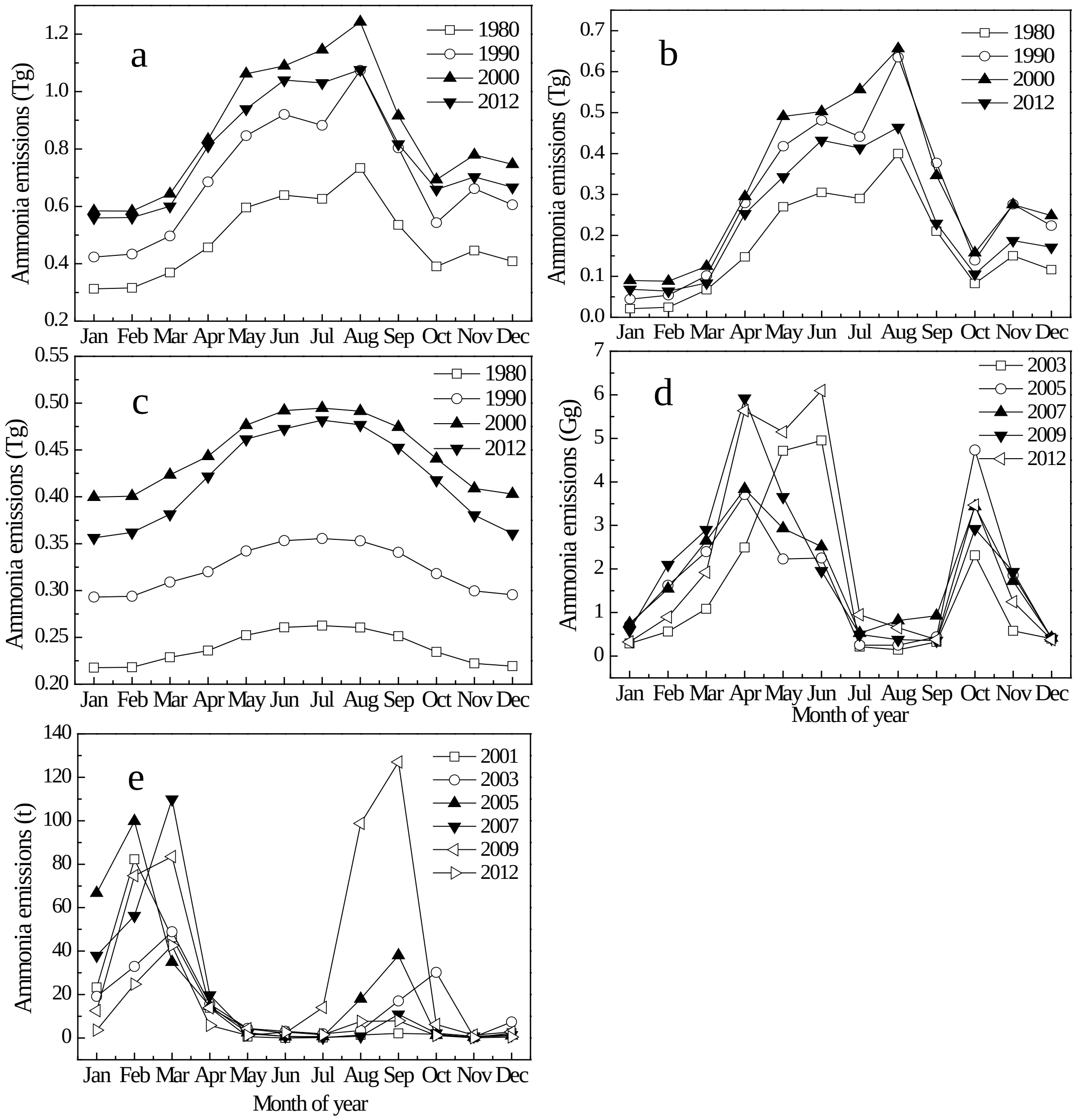

Figure 6. Monthly distribution of NH_3 emissions from different sources in China: (a) all the sources; (b) synthetic fertilizer; (c) livestock wastes; (d) crop burning in fields; (e) forest and grass fires.

Minor comments

-Table S1 is copied from the Huang et al. (2012) paper except for the EEA reference which is now more recent. To avoid problems, this needs to be made clear.

Response: Accepted. We thank the reviewer for the suggestion. Actually, there are almost no differences between recent released EEA inventory guide and the early version for the parameters we used. So we removed this table in the revised manuscript and the detailed values can be looked up in Table 3 in Huang et al.'s paper. We have added more information about estimation method to livestock emissions. Please see Section 2.2.

-Header of section 2.1.2: the soil pH cannot be a source of ammonia.

Response: Accepted. We changed it to "In-situ measurement" in Section 2.1.1 (Line 183).

-The section 2.1.2 on improvement of the EF for soil pH is not clear.

Response: Accepted. We reworded it in Section 2.1.1 (please see Lines 184–195).

Revision in Lines 182–196 on Page 8: "For acquiring up-to-date EFs that could reflect NH_3 volatilization from synthetic fertilizer application in present Chinese agricultural practice, we measured NH_3 EF by using micrometeorological method for a whole year in a typical farmland in the North China Plain and an inverse dispersion model was also used to derive the ammonia EFs (Huo et al., 2014, 2015). The in-situ results could represent better than those used in Huang et al. which were derived from studies in early years. The soil pH and mean air temperature in this farmland was 8.2 and 15°C, respectively. The measurement yielded an NH_3 EF for urea of 12%±3% in this case. Huang et al. (2012b) develop a linear relationship between NH_3 volatilization and soil pH to involve the impact of soil acidity on EFs according to Cai et al. (1986) and Zhu et al. (1989). We applied the condition-specific EF we measured recently to refine this relationship with linear regression analysis."

-A description of the approach for the Monte Carlo analysis is missing.

Response: Accepted. We add a brief description about the Monte Carlo method in Lines 634−640.

Revision in Lines 634–640 on Page 26: "Monte Carlo is an effective method to evaluate the uncertainties in various issues including an emission inventory. In Monte Carlo simulation, random numbers are selected from each distribution (normal or uniform) of input variables and the output uncertainty of an emission inventory is based on the input uncertainties from activity data and emission factors. In this study, we ran 20,000 Monte Carlo simulations to estimate the range of NH_3 emissions with a 95% confidence interval for 1980, 1990, 2000, and 2012. The estimated emission ranges were 4.5–7.4 Tg/yr, 6.3–11.1 Tg/yr, 8.0–13.4 Tg/yr, and 7.5–12.1 Tg/yr, respectively."

-It is not clear how the temporal distribution of the other sources has been done.

Response: Accepted. We add a brief description about the method of the monthly NH_3 emissions from the other sources in Section 2.4 (please see Lines 271−282).

<u>Revision in Lines 271–282 on Pages 11−12:</u> "The seasonal NH_3 emission estimation for fertilizer application could be calculated as the product of condition-specific EFs derived from meteorological factors (average monthly temperature and wind speed) and monthly fertilizer consumption associated with agricultural timing. For livestock emissions, we assumed that the number of each livestock category per month remains constant, because the monthly fluctuation in the production of meat, eggs and milk is very small (http://www.caaa.cn/). The monthly EFs were distinguished by average monthly temperature and wind speed from NCEP. Besides, the emission from biomass burning also shows a temporal fluctuation. MCD45A1 (monthly burned area product), MOD14A2 and MYD14A2 products (8-day thermal anomalies/fire products) were utilized to ascertain the timing of different kinds of biomass. For other minor sources, the emissions were equally divided into 12 months."

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