Comments:

In this manuscript, the authors report on a study aimed at analyzing the changes of hourly NH_3 concentrations and estimating surface NH_3 concentrations and NH_3 emissions in China with top-down method. The manuscript fits into the scope of ACP and the results presented are very interesting to their readers. Overall the paper is clearly structured and generally well written. I have the following comments of the paper that should be addressed.

We thank the reviewer for your time and helpful comments. Our point-by-point response is enclosed.

General comments:

1. Although the sources of uncertainty in the experiments covered are described in the limitations and outlook section, a quantitative analysis is lacking and should be added. How did you solve the problem of missing GIIRS data in the Yangtze River Basin mentioned in the constraints?

We thank the reviewer for the comment. Surface NH_3 concentration is the key variable in NH_3 emission calculation. In this paper, the NH_3 measurements from the NNDMN in China were collected and compared with IASI-derived surface NH_3 concentrations. The regression R_2 between measured results and satellite-estimated annual means was 0.72 and the RMSE was 2.24 μg N m^{-3}. The coefficient of the fitted line was 1.03 ≈ 1, with the bias of 2.59%. The regression R_2 between monthly average IASI-derived NH_3 concentrations and measured NH_3 by month ranged from 0.38-0.84, and the RMSE ranged from 2.29-3.36 μg N m^{-3}, with the biases less than 30% for all months. Overall, the calculated annual and monthly average IASI-derived surface NH_3 concentrations showed good agreement with the

measurements of sites, and generally indicated the level of error in the surface NH_3 concentration estimates.

For the missing GIIRS NH_3 observations in the Yangtze River Basin, we used GIIRS NH_3 observations to analyze the regional daily variation of NH_3 concentrations in China, and to estimate monthly average surface NH_3 concentrations and study the spatial and temporal distribution. The absence of NH_3 column in the Yangtze River basin can be filled by spatial interpolation. We did not interpolate the GIIRS NH_3 column, as it weakly affected the analysis of the daily cycle of NH_3 concentrations in China. We had averaged the observations for the same period (5 hours interval), and the spatial missing values were greatly reduced. With the exception of the Yangtze River Basin, the distribution of NH_3 concentrations was relatively complete in other regions. In addition, the main missing fraction of monthly mean surface NH_3 concentrations from GIIRS was also found in a small part of the Tibetan Plateau, and interpolation was not carried out as it would introduce additional errors.

We added the following sentences into our manuscript.

"Third, the spatial resolution of the NH_3 vertical profile simulated by the atmospheric model is relatively coarse (0.5 degrees). In order to make it consistent with the spatial resolution of the remote sensing data, the outputs of GEOS-Chem (vertical profiles and feedback ratio between emissions and surface NH_3 concentrations) were interpolated through resampling methods. Owing to the resolution limit, the ratio-based mass balance approach to estimate NH_3 emissions neglected the effects of internal transport of NH_3 and displacement of emission sources within the fine grid.

Finally, there are some uncertainties and biases in the observed NH_3 column by satellite. Earlier versions of the IASI NH_3 column product were 25-50% lower than ground-based measurements (Whitburn et al., 2016; Dammers et al., 2017). However, the new version of IASI v3 lacks a comprehensive ground-based measurement assessment, which has only been compared with limited aircraft observations(Guo et al., 2021). Comparing IASI-derived surface NH_3 concentrations with measurements of ground sites (NNDMN) generally shows consistency in this study. The further work is needed for the complete assessment and error analysis."

2. I am confused about the treatment of the feedback ratio of surface NH_3 concentrations and emissions mentioned in the methodology. Is it the calculation done on an annual scale or on a monthly scale? Is it a variable value over time or a constant value? The feedback ratio should also be included as an element in the uncertainty and limitation analysis.

We thank the reviewer for the comment. We obtained the feedback ratio between surface NH_3 concentrations and NH_3 emissions using the mass balance method with GEOS-Chem simulation. In the study, the REAS emission inventory was used as China's anthropogenic emissions into GEOS-chem. However, as the time series range of REAS only corresponded to the IASI observations during 2008-2015, the feedback ratios for 2016-2019 were not obtained. Therefore, we used the fixed monthly average feedback ratio (Fig. S2b) for the calculation of NH_3 emissions.

We added the following figures in supplement.

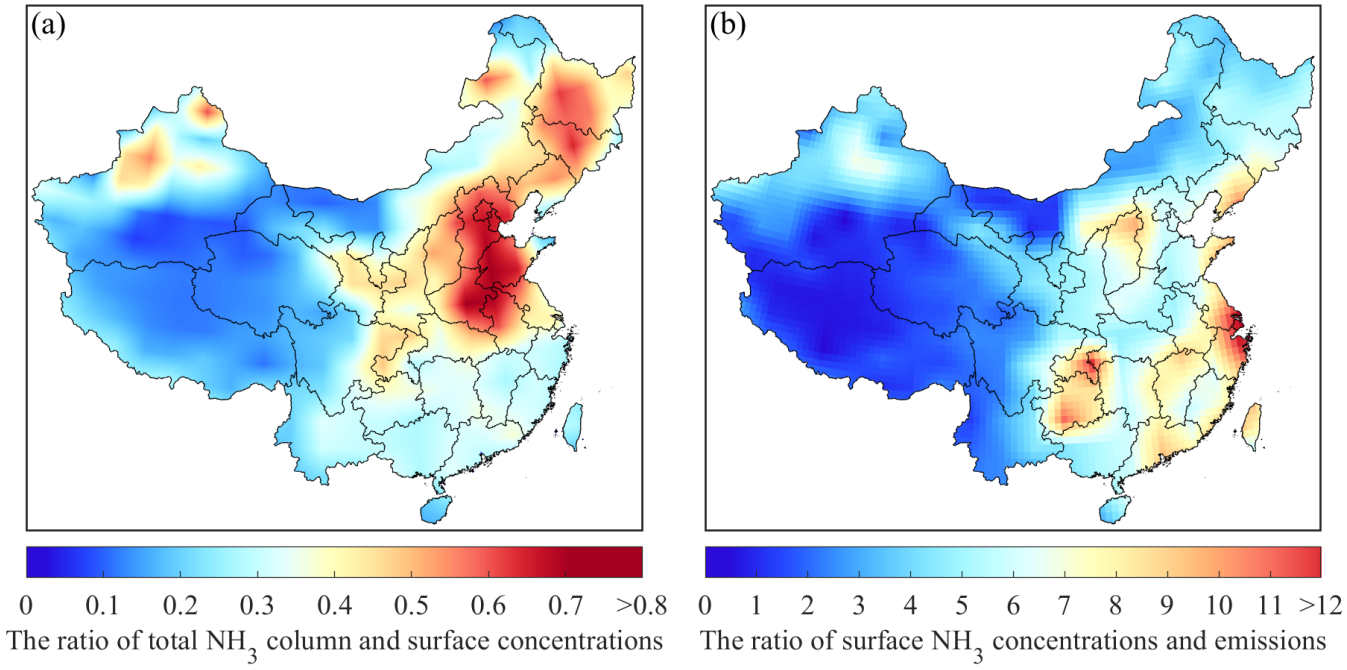

Figure S2. Conversion ratios from GEOS-Chem simulations. (a) The conversion ratio of total NH_3 concentrations and surface NH_3 concentrations. (b) The feedback ratio of surface NH_3 concentrations and NH_3 emissions.

Minor comments:

line 247: check and modify the content in the Figure 5

Figure 5 showed satellite-derived surface NH_3 concentrations compared to ground-based measurements from 2008-2015. There were some errors in the description and we have revised them.

"Monthly regression R^2 between the satellite-derived NH_3 concentration and the measured NH_3 was 0.38-0.84. The regression R^2 reached the higher value (>0.80) in July and August. The RMSE ranged from 2.29- 3.36 μg N m^{-3}, which reached the maximum value of 3.36 μg N m^{-3} in July, and reached the smallest in March (2.29 μg N m^{-3}). The bias is basically less than 31% for all months, and reached the minimum

value of -0.67% in February, indicating that the monthly IASI-derived surface concentration obtained are consistent with measurements."

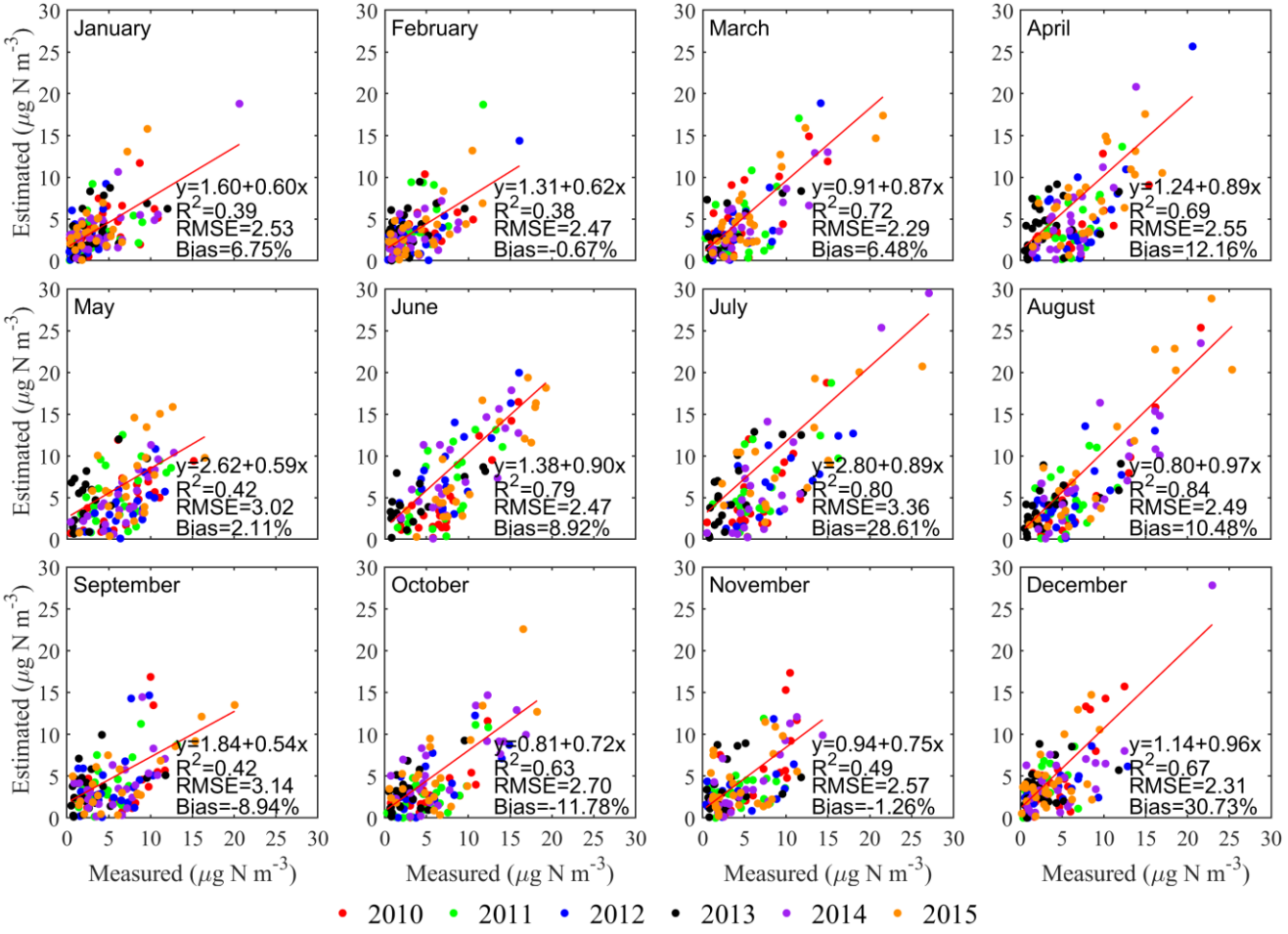

Figure 5. Comparison of monthly average values of IASI-derived and observed NH_3 surface concentrations in 2010-2015.

line 292: The data of Figure. 8 doesn't match the data described in the article

We thank the reviewer for pointing this out. Figure 8 showed the yearly change in IASI-derived NH_3 emissions over China from 2008-2019, the monthly change in NH_3 emissions in 2019 and the spatial

distribution of NH_3 emissions in January, April, July and October 2019. There were some errors in the description and we have revised them.

"Based on the top-down estimates, China's NH_3 emissions ranged from 12.17-17.77 Tg N yr^{-1} during 2008-2019. From 2008 to 2015, NH_3 emissions increased from 13.00 Tg N yr^{-1} to 17.06 Tg N yr^{-1}. Since 2008, the temperature in China has risen steadily (Ding et al., 2007), which promotes the volatilization of NH_3, which partly explains the increase in NH_3 emissions from 2008 to 2015. After 2015, NH_3 emissions fluctuated and changed slightly (16.08-17.77 Tg N yr^{-1}). Compared with other studies, the change in NH_3 emissions from 2008 to 2015 is consistent with previous estimates, and the overall NH_3 emissions show an upward trend (Kang et al., 2016; Zhang et al., 2018; Ma, 2020; Fu et al., 2020; Zhang et al., 2021). Our estimates are on the rise as a whole, but the calculated values are generally lower than those by (Fu et al., 2020) (around 15 Tg N yr^{-1}), but larger than those by EDGAR and Kang et al. (2016). "

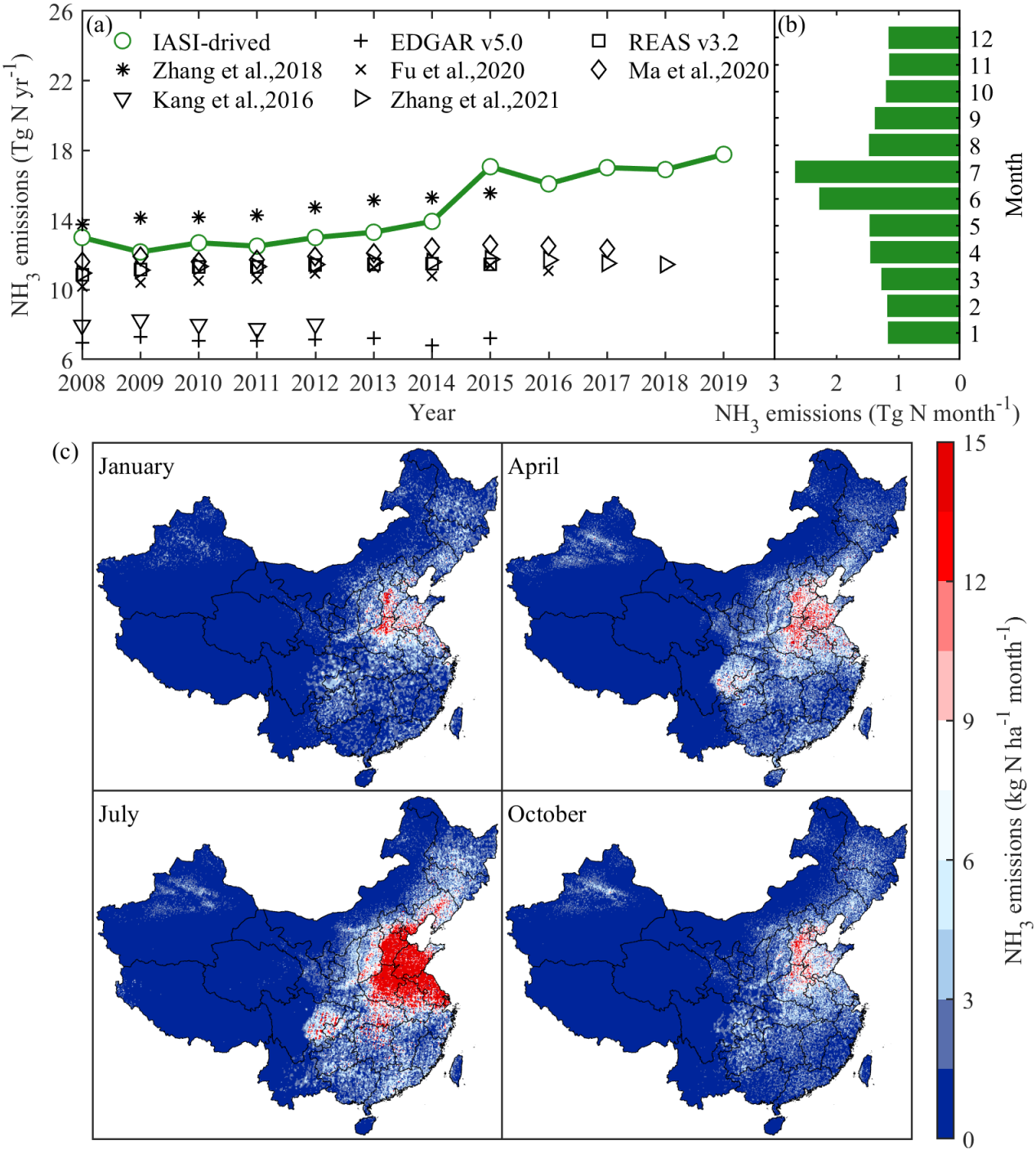

Figure 8. Annual changes of NH_3 emissions (a), monthly changes of NH_3 emissions in 2019 (b) and spatial distribution of NH_3 emissions by month in 2019 (c).

line 13: replace "China has largest NH_3 emissions in the world…" by "China has the largest NH_3 emissions globally…"

We have changed it as suggested. "China has the largest NH_3 emissions globally, mainly associated with agricultural sources including nitrogen fertilizer and livestock."

line 89: replace "method by using" with "method using"

We have changed "by using" to " using".

line 103: Correct "are" to be " is"

It is fixed now.

line 115: replace "at high frequency" with "at high frequencies"

We have changed it as suggested.

line 121: change "The average value of HRI is 0 with the standard deviation as 1" to be "The average value of HRI is 0 with a standard deviation of 1"

We have changed it as suggested. "The average value of HRI is 0 with a standard deviation of 1, and the HRI range is [-1,1]. "

line 124: replace "from November in 2019 to October in 2020" to be "from November 2019 to October 2020"

We have changed it as suggested. " In this study, we used hourly NH_3 concentrations during 2019-2020 (from November 2019 to October 2020) to study NH_3 diel cycle with a resolution of 0.5°."

line 139: change "product of" to be "product of the"

We have changed it as suggested.

line 168: Correct "which is" with "which are"

Corrected.

line 208: replace "while for other time…." with "while NH_3 concentration … at other times"

We have changed it as suggested. "…while NH_3 concentration tends to be stable at other times."

line 208: replace "changes of" to be "changes in"

We have changed it as suggested.

line 214: replace "which may be also related with" by "which may also be related to"

We have changed it as suggested.

line 215: replace "except" by "except for" and replace "have" by "has"

We have changed it as suggested.

line 216: Correct "patterns" by "patterns are"

Corrected.

line 253: delete "during 2010-2015"

We have removed the " during 2010-2015".

line 267: replace "have" by "had"

We have changed it as suggested.

line 276: replace "change of" to be "change in"

It is fixed now.

line 305: Correct "are" to be "is"

Corrected.

line 315: change "estimated" to be "estimate"

We have changed it as suggested.

line 318: change "in" to be "from"

Fixed.

line 320: change "low" to be "the low"

It is fixed now.

line 325: change "occurred" to be "occur"

We have changed it as suggested.

Reference

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