Response to Reviewer #1:

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.

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?

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 $\approx$ 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-hour 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 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 the supplement.

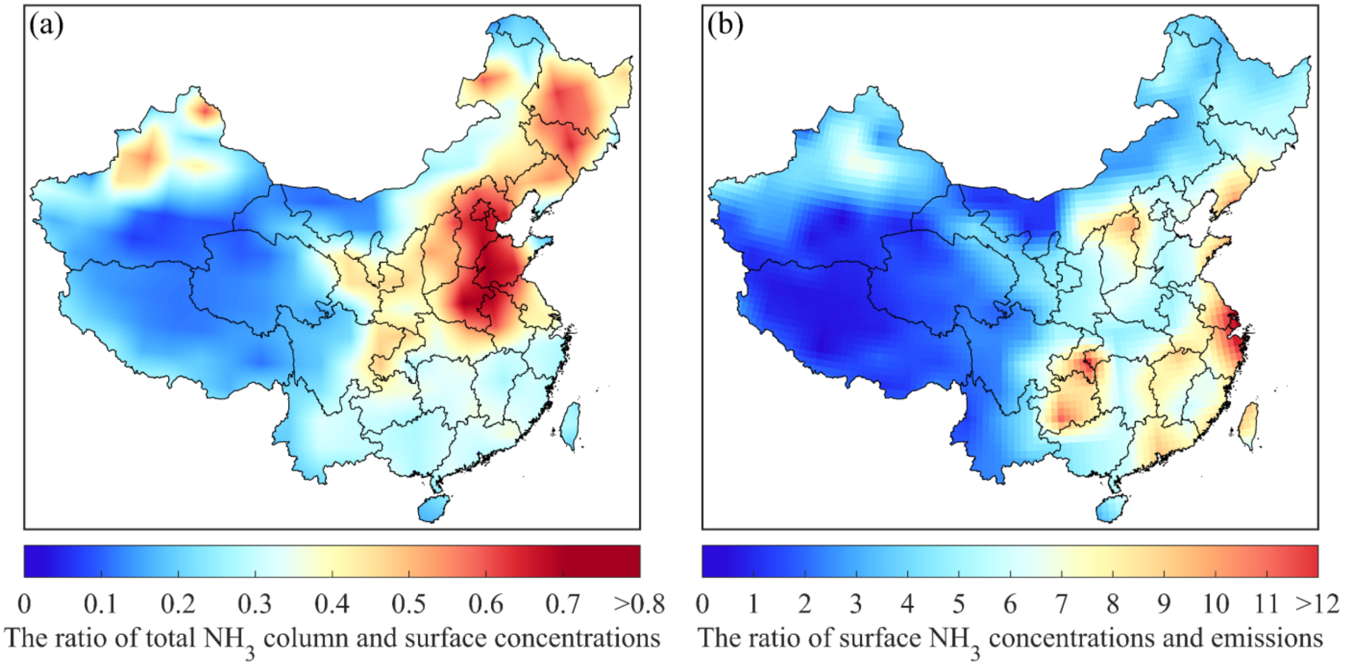

Figure S2. Conversion ratios from GEOS-Chem simulations. (a) The conversion ratio of total NH_3 concentrations and surface NH_3 concentrations. (b) The feedback ratio of surface NH_3 concentrations and NH_3 emissions.

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.”

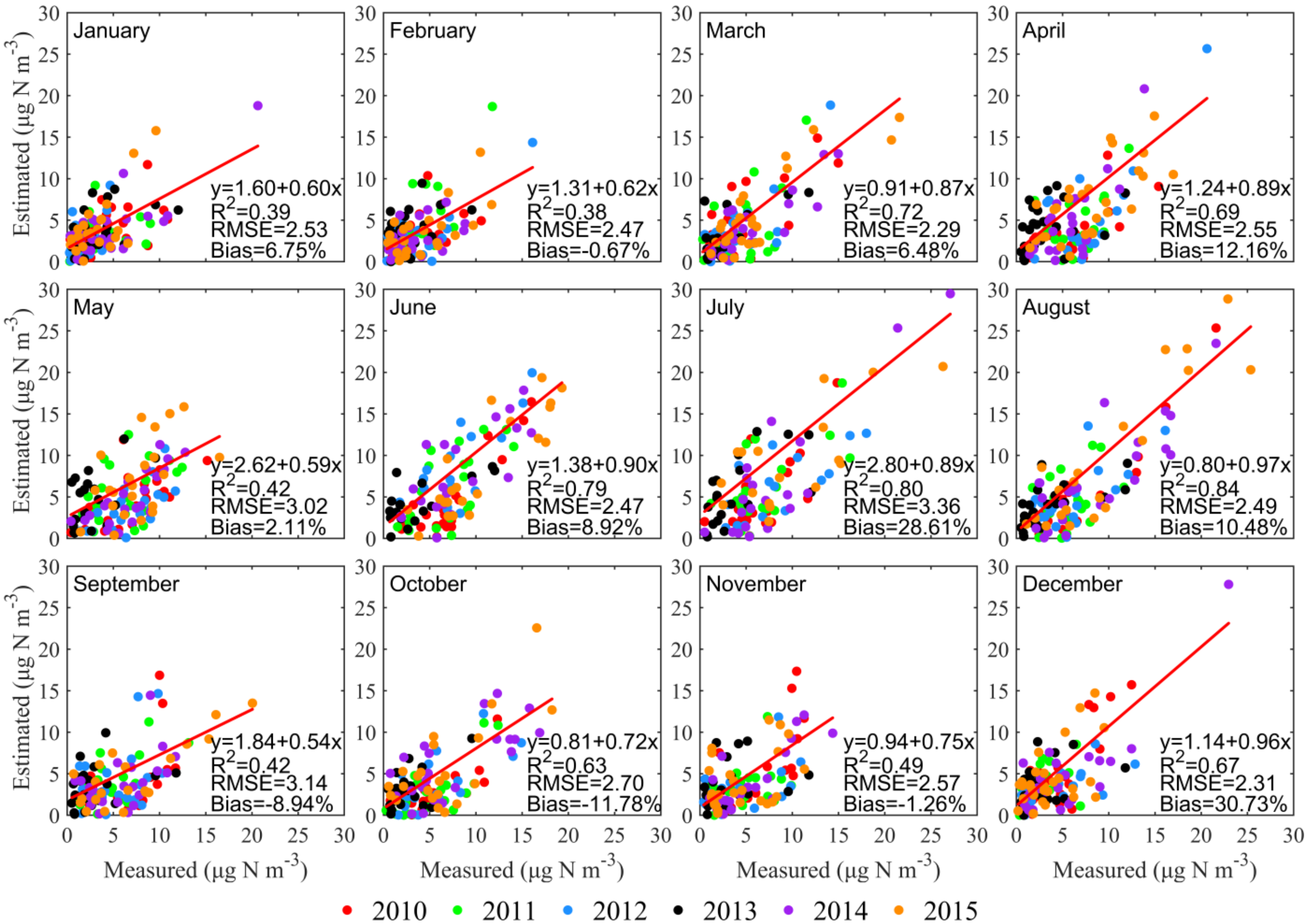

Figure 5. Comparison of monthly average values of IASI-derived and observed NH_3 surface concentrations in 2010-2015.

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). ”

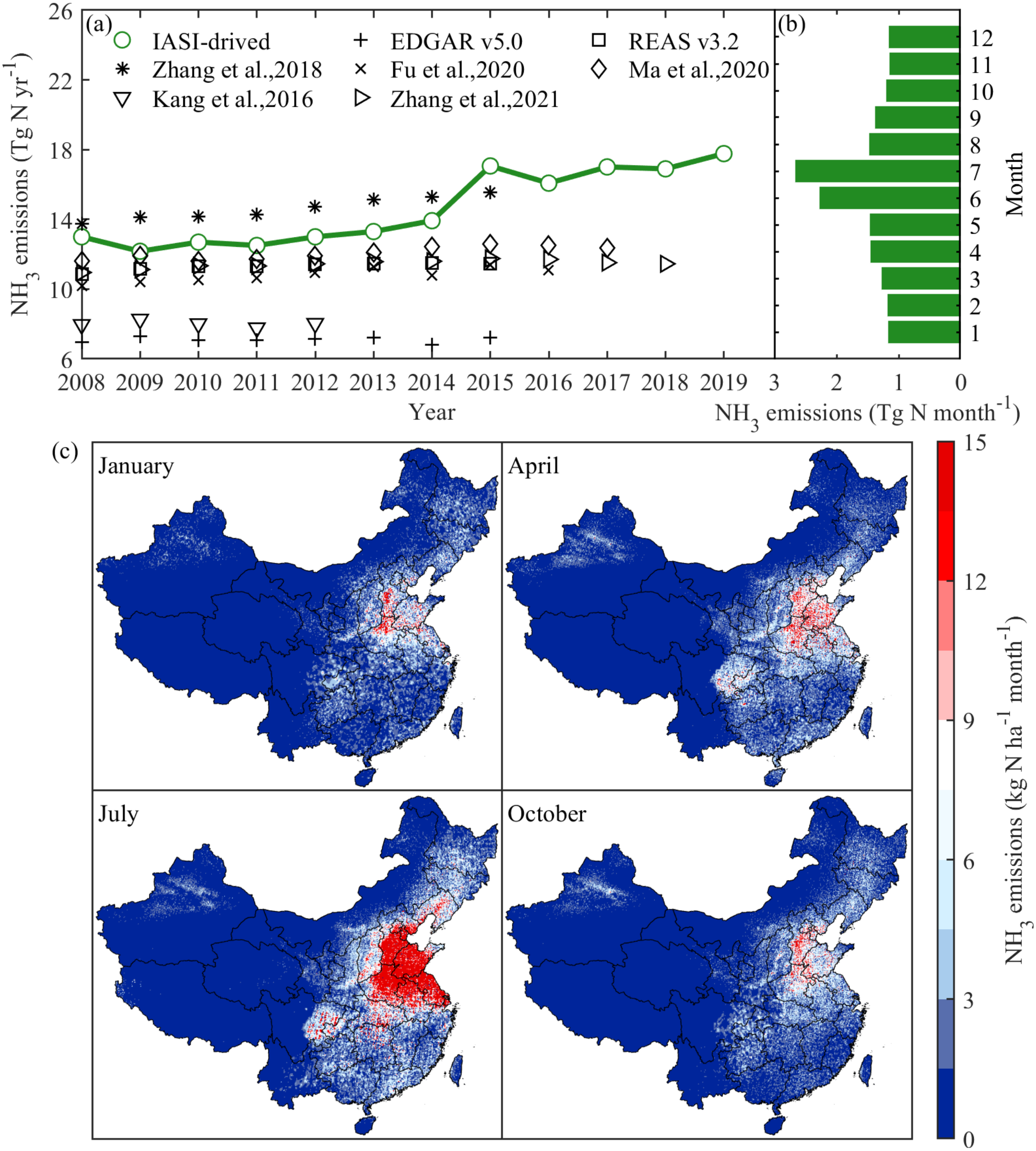

Figure 8. Annual changes of NH_3 emissions (a), monthly changes of NH_3 emissions in 2019 (b) and spatial distribution of NH_3 emissions by month in 2019 (c).

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"

It is fixed now.

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"

It is fixed now.

Response to Reviewer #2:

Comments:

This paper is analyzing hourly variation of NH_3 concentrations and quantifying surface NH_3 concentrations and NH_3 emissions in China, using observations from GIIRS and IASI. A three parameter Gaussian function is used to fit NH_3 vertical profiles from GEOS-Chem and get information of NH_3 concentration at different heights. Surface NH_3 concentrations and total NH_3 emissions are estimated based on the mass balance method and ratio from GEOS-Chem.

It was found that diurnal NH_3 concentrations are larger than nightly NH_3 concentrations. A good agreement is obtained between the ground measurements and the estimated. The NH_3 emissions range from 12.99 to 17.77 Tg N yr^{-1} between 2008 and 2019 in China. The paper also discussed the uncertainties and capabilities of the method. The topics of paper fits the scope of ACP and the scientific idea is new. The article is generally well written and easy to follow. I have the following comments of the paper but I am supportive of publications if these aspects can be addressed.

Major concerns:

1. Please indicate the basis for the satellite data quality screening and the number of valid pixels after eliminating invalid pixels. If the proportion of remaining valid pixels is low, the study results will be misleading and appropriate data supplementation should be performed.

Since the quality of satellite data was greatly affected by cloud cover, we deleted all data with recorded cloud cover >20%, which was more stringent than previous studies (Wang et al., 2020b). Besides, the quality of satellite data was also affected by other factors, such as retrieval method and inversion algorithm. We also deleted the observations with the uncertainty higher than 50% (Fortems‐Cheiney et al., 2016).

We performed a relatively stringent screening of the observations. The further quality constraint is feasible, but more satellite observations will be missing, affecting the spatial continuity of the NH_3 column and the estimated NH_3 emissions. We supplemented Fig. S1 showing the effective pixel count of GIIRS and IASI with the above filtering conditions. The total number of pixels covered by GIIRS (spatial resolution 0.5°) in China is about 3840, and the total number of pixels covered by IASI (spatial resolution of 0.1°) is about 96170.

Fig. S1a shows the number of effective pixels for the monthly average NH_3 column of GIIRS in each overpass period (defined as 2-hour interval) during 2019.11-2020.10. The red dotted line represents the total number of pixels (the sum of the records of 10 overpass periods with 2-hour interval), and the inside of the bar graph is marked as the ratio of the number of valid pixels to the total pixels in a single overpass period. All the monthly average NH_3 columns of GIIRS accounted for more than half of the total effective pixels, and the minimum value of the effective pixels for each overpass period exceeded 25%. GIIRS had the higher number of effective pixels in March, April, and May, while the number of effective pixels in January, July, and December were relatively low, which was consistent with the results (Fig. 3) of the monthly average surface NH_3 concentration by GIIRS in China. It should be noted that due to the lack of observations at 24:00, the observations at 23:00 were not shown in Fig. S1a (the observaed coverage is extremely limited).

Fig. S1b shows the effective pixel count of the monthly average NH_3 column by IASI from 2008 to 2019. The red dotted line represents the total number of pixels, and the outer labels of the pie chart represent the ratio of the effective pixels to the total pixels over China. Here, we only marked a few months with the high proportion in the pie chart. The effective pixels of the IASI NH_3 column showed an overall increasing trend during 2008-2019. The number of IASI NH_3 effective pixels was significantly higher in summer than in winter, and the effective pixel in July 2015 accounted for the highest proportion (>50%). Although the proportion of IASI NH_3 valid pixels was generally lower than GIIRS, most of which was around 35%, the spatial distribution of IASI NH_3 effective pixels in China was relatively uniform without concentration. We had performed interpolation processing to ensure the spatial continuity and integrity of the IASI NH_3 column. In general, the number of available pixels from GIIRS and IASI met the analysis requirements.

We added the following sentences and figures in the supplement.

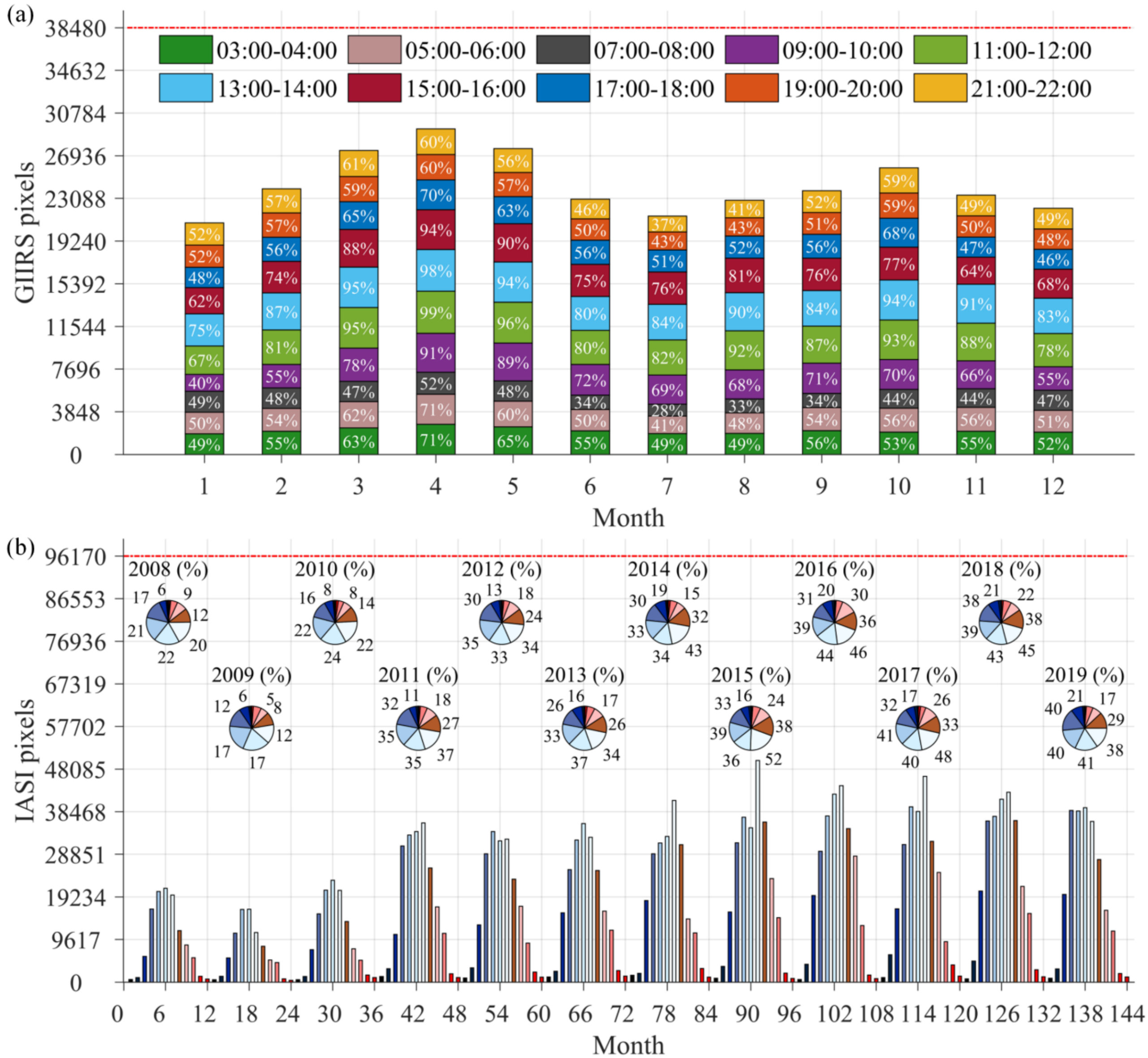

Figure S1. The variation of valid pixels after quality control. (a) Effective pixels of the monthly average NH_3 column by GIIRS in each overpass period (2-hour interval) during 2019.11-2020.10; (b) Effective pixels of monthly average NH_3 column by IASI from 2008 to 2019.

2. In the paper, two kinds of satellite observations are using to estimate surface NH_3 concentration. Figure. 3 and Figure. 7 show the differences in spatial distribution and numerical magnitude between them. Although there are problems of scale conversion, the comparison of the estimation results is of great necessity, especially at similar satellite overpass time.

Clarisse et al. (2021) compared the v3 version of the IASI NH_3 dataset (Van Damme et al., 2021) and GIIRS for NH_3 retrieval in terrestrial areas (Fig. 9). Observations with the difference in measurement time within 15 minutes and the pixel center distance within 2 km are considered as corresponding observations. Overall, the regression slope was found to be 1.14, the correlation coefficient of R_2 was greater than 0.65, and the bias was small. The reasons for the inconsistency included instrument calibration, HRI calculation, processing of surface temperature, systematic biases of neural network, and different observed geometries.

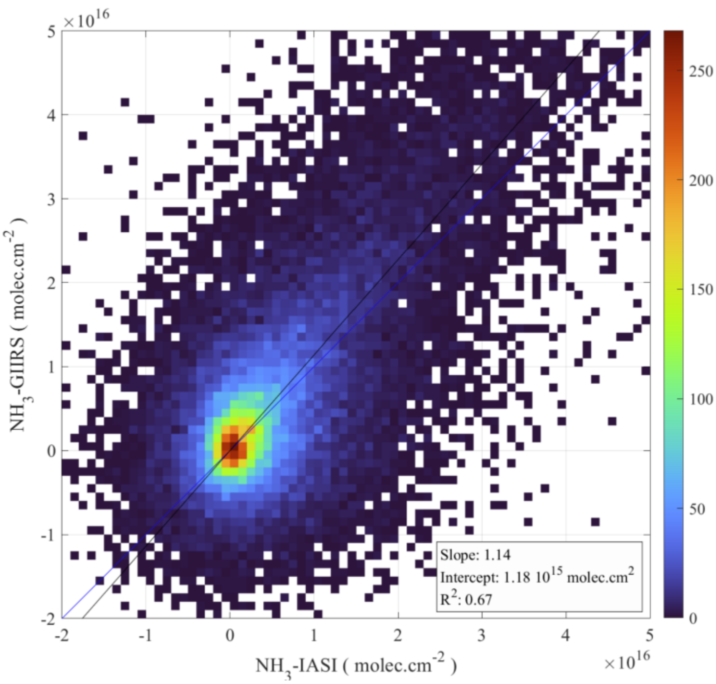

Figure 9. Comparison of IASI and GIIRS NH_3 retrievals in terrestrial regions (Clarisse et al., 2021).

3. Figure. 8 shows an abrupt change of surface NH_3 emissions in China during 2014-2015. The value of surface NH_3 emission is estimated around 17 Tg in 2019, which may be overestimated compared to previous findings. Its accuracy is questionable, and I suggest extending uncertainty analysis.

We agree with the reviewer that NH_3 emissions over China in 2019 may be overestimated. In this study, estimated total NH_3 emissions of China reached 17.77 Tg N yr^{-1} in 2019, which was a high value and had the potential to be overestimated. We added the following sentences into our manuscript.

“Second, we used a relatively fixed average conversion ratio (Fig. S2) to estimate surface NH_3 concentrations and NH_3 emissions in China, ignoring the time-series variation of the ratio, due to the temporal constraint of emission inventory. In this case, non-emission factors led to higher satellite observed NH_3 column, for example, emission reductions of SO_2 and NO_2 led to increased NH_3 column (Lachatre et al., 2019; Fu et al., 2020), which can introduce uncertainty into NH_3 emission calculations using concentration as the main parameter.”

4. The quality of the figures in the paper needs to be improved, and there are errors and inconsistencies in the graphic descriptions, which should be carefully corrected.

We have further checked and updated all figures in the paper, and revised inconsistencies between the descriptions and figures.

Minor comments:

Page 2, line 32: Change "To provide a scientific basis of…" to "To provide a scientific basis for…".

We have changed it as suggested.

Page 3, line 43: Change "Some studies have carried out…" to "Some studies have carried out conducted…".

We have changed "carried out" to "conducted".

Page 3, line 52: "China’s cultivated land area accounts for only 8% of the world, but it consumes about 30% of the world’s nitrogen (N) fertilizer". Please add article references.

We have added references as suggested.

“China’s cultivated land area accounts for less than 10% of the world, but it consumes about 30% of the world’s nitrogen (N) fertilizer (Peng et al., 2002).”

Page 5, line 103: Change "are" to "is".

We have changed it as suggested.

Page 7, line 126: There is a misuse of symbols in units (molec·cm^{-2}).

We have changed it as suggested.

Page 8, line 151: Describe the information of sites in tabular form.

Table S1. The information of the collected hourly measured sites.

Name	Class	Lat	Lon	Period	Reference
Xianghe	rural	39.75 °N	116.96 °E	2017.12 -2018.2	(He et al., 2020)
Fudan University	urban	31.30 °N	121.50 °E	2013.7-2014.9	(Wang et al., 2015)
Dianhushan	rural	31.09 °N	120.98 °E	2014.3-2014.6	(Wang et al., 2015)
Gucheng	urban	39.15 °N	115.73 °E	2016.3-2017.5	(Kuang et al., 2020)
Jinshan Chemical Industry Park	industrial	30.73 °N	121.27 °E	2014.1-2014.6	(Wang et al., 2015)

Page 9, line 168: Change "is" to "are".

We have changed it as suggested.

Page 9, line 175: Check the following formula format.

We have fixed it as suggested.

Page 14, line 242: Description in 3.2 is not fitting to Figure. 5.

We have fixed it as suggested.

“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.

Page 15, line 248: There are any errors in Figure. 5, suggest reconstruction.

We have revised and updated Figure. 5.

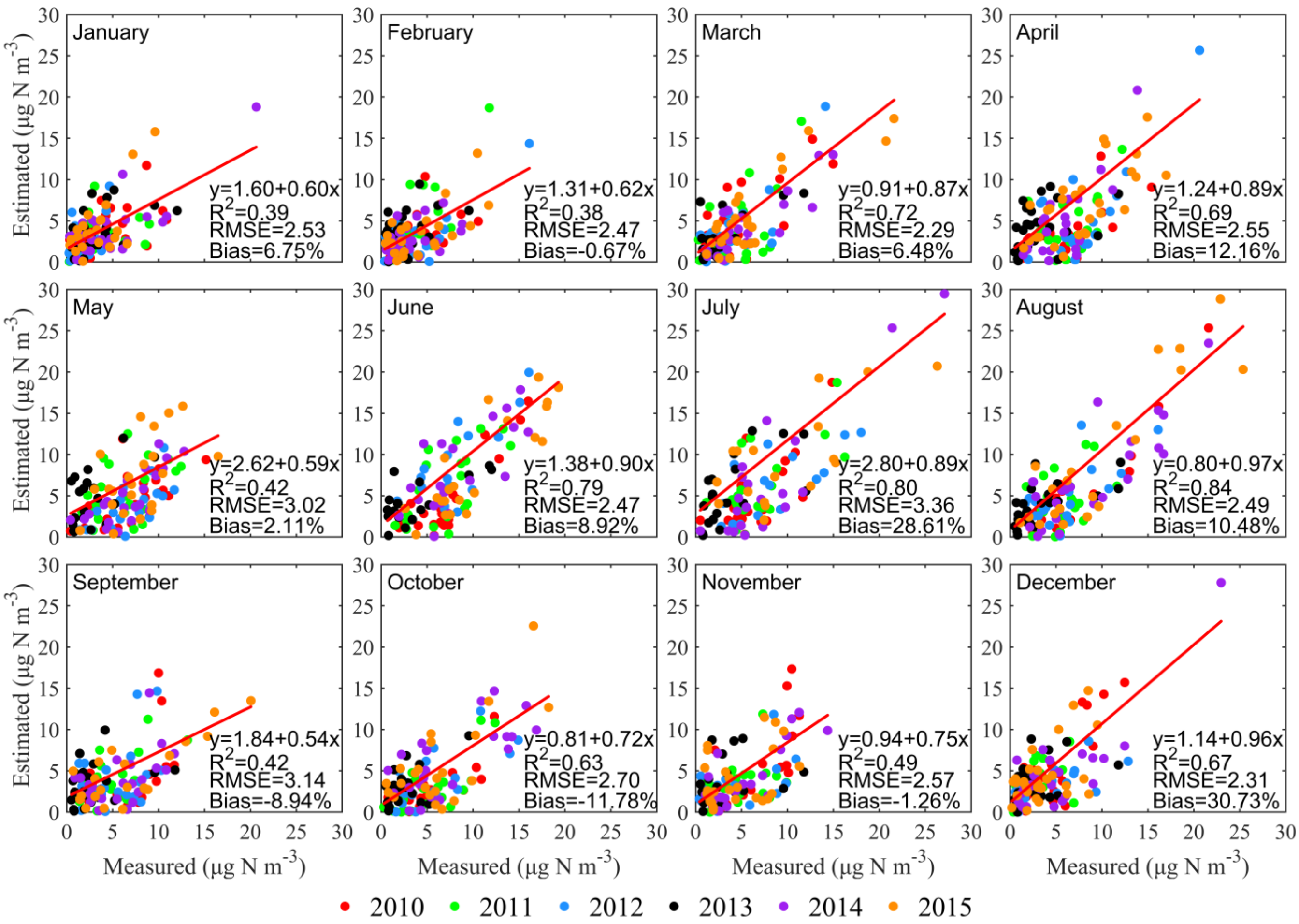

Figure 5. Comparison of monthly average values of IASI-derived and observed NH_3 surface concentrations in 2010-2015.

Page 20, line 304: Why is the time series of the Fengyun geostationary satellite data so short? Can you give an explanation?

Clarisse et al. (2021) reported the first observations of daily atmospheric NH_3 over Asia by satellite FY-4A GIIRS. The fast NH_3 retrieval method originally developed for IASI was used on a full year of GIIRS observed retrieval from November 2019 to October 2020. The dataset of atmospheric NH_3 columns from the FY-4A GIIRS is a short time series product and FY-4A is nearing the end of its lifespan, but it's promising given the future landscape of geostationary sounders.

Page 20, line 305: Change "are" to "is".

We have changed it as suggested.

Response to Reviewer #3:

Comments:

The study estimated the changes of hourly NH_3 concentrations, surface NH_3 concentrations and NH_3 emissions in China using the polar-orbiting satellite (IASI) and Fengyun-4 geostationary satellite. The results show NH_3 concentration in daytime was generally higher than that at night. Satellite-based NH_3 emissions ranged from 12.99-17.77 Tg N yr^{-1} during 2008-2019. The manuscript is overall well organized and written. The analyses are neatly conducted and fit the scope of ACP. Before recommending publish the study, I have the following comments that I think the authors shall address to improve the manuscript.

General comments:

1. Throughout the paper, there are issues with the use of plural vs singular and or verb tense, especially in the use of 3rd person, plural or singular. There is also extensive mixed use of past tense and present tense instead. I strongly recommend using unified tense instead, throughout the paper.

We have re-checked the use of plural/singular, used the present tense, and conducted a careful copyediting throughout the paper.

2. The feedback between surface NH_3 concentration and emissions was calculated by GEOS-Chem. Please describe the simulation process in detail and driven data in SI.

The study used the global 3-D chemistry transport model GEOS-Chem v12.3.0 , developed by Harvard University, to simulate and calculate the conversion ratio of surface NH_3 concentrations and emissions, which was widely used in the field of atmospheric physical chemistry research (Chen et al., 2009; Zhang et al., 2011). GEOS-Chem contained detailed tropospheric gas-aerosol (O_3, NO_x, NH_3, SO_4^{2-}, NO_3^- and NH_4^+, etc.) chemistry, and driven by assimilated meteorological information from the NASA Goddard Earth Observing System (GEOS-5) (https://gmao.gsfc.nasa.gov/). The source programs of GEOS-Chem were freely available from Atmospheric Chemistry Modeling Group at Harvard University (http://acmg.seas.harvard.edu/geos/geos_overview.html). The driver input files contained meteorological data and emission inventory, and the directory file contained the initial concentration file, photolysis mechanism and chemical mechanism files.

In this study, the nested regional model for Asia was driven by assimilated GEOS-5 meteorological data with a horizontal resolution of $1/2^{\circ} \times 2/3^{\circ}$. The GEOS-Chem model here did not consider land-atmosphere bi-directional NH_3 exchange, and the NH_3 flux was parameterized as uncoupled emission and dry deposition processes. Anthropogenic emissions in China are from the Regional Emission in Asia (REAS-v2) inventory. The GEOS-Chem NH_3 concentrations output includes 47 layers from the ground to the top of the atmosphere to capture the NH_3 vertical profiles. Only the inventory data overlapped with the IASI measured period were used, which was sampled and consistent with the model resolution. We have added the following figure in the supplement.

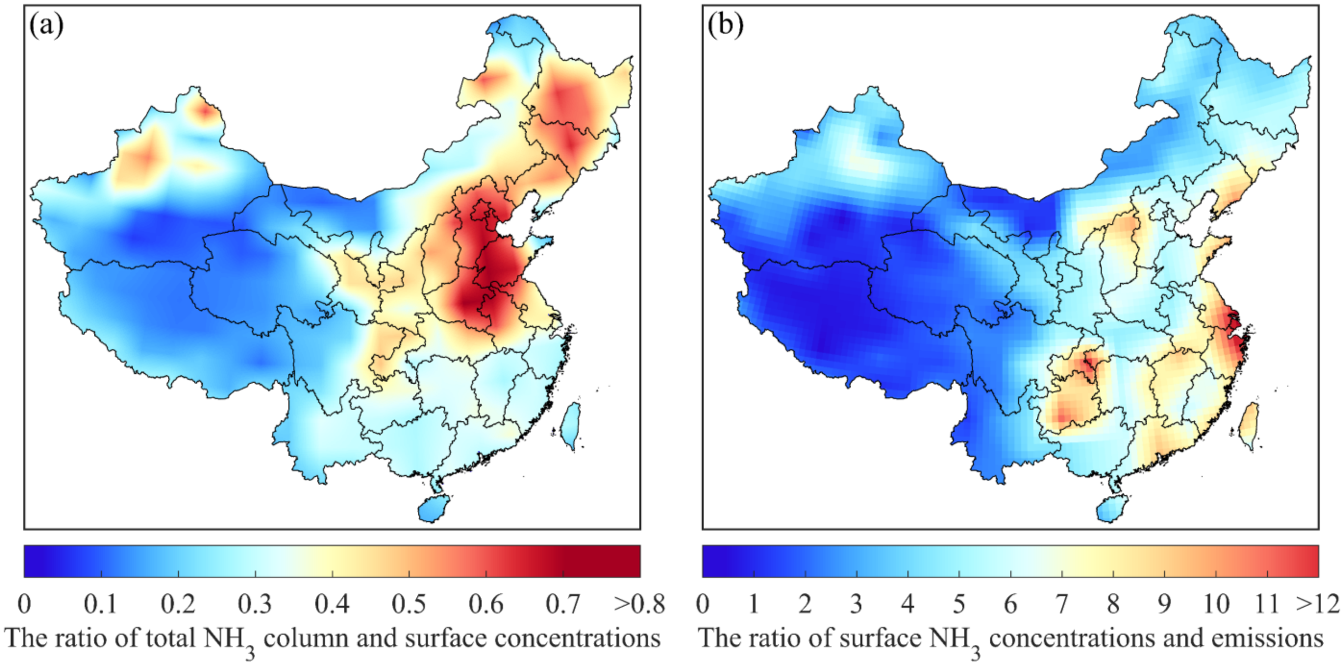

Figure S2. Conversion ratios from GEOS-Chem simulations. (a) The conversion ratio of total NH_3 concentrations and surface NH_3 concentrations. (b) The feedback ratio of surface NH_3 concentrations and NH_3 emissions.

Minor comments:

L56, please further check if the value is 40%, if so, annual farmland NH_3 emission were estimated as 2.4 Tg N yr^{-1} by the IPCC tier 1 guidelines.

We have further checked the citation and the method in the paper calculates NH_3 emissions from China's farmland to be 40% higher than the IPCC in 2008. However, we lacked clarification and the data used in the paper were for the specific year. We have made the following changes to the original text.

“Zhou et al. (2016) calculated the annual farmland NH_3 emission (3.96 ± 0.76 Tg N yr^{-1}) over China in 2008 based on the bottom-up method, which is 40% higher than the emission in the Intergovernmental Panel on Climate Change (IPCC) Tier 1 guidelines (2.89 Tg N yr^{-1}).”

L29, 57, what’s SO_2, NOx, NH_4^+, and IPCC, etc.

NH_3 reacts with acid pollutants (Sulfur dioxide (SO_2) and nitrogen oxides (NO_x)) to form fine particulate matters (such as PM2.5 (particles less than 2.5 micrometers in diameter)), leading to haze pollution. In addition, the deposition of NH_3 and ammonium (NH_4^+) could also cause environmental problems such as water eutrophication, biodiversity loss and soil acidification (Paerl et al., 2014).

We have added the following content to explain them. “…, which is 40% higher than the emission in the Intergovernmental Panel on Climate Change (IPCC) Tier 1 guidelines (2.89 Tg N yr^{-1}).”

L113, correct word ‘is’ to ‘are’, please check similar mistake throughout the manuscript.

It is fixed now, and we have double-checked and corrected similar errors in the manuscript

L201, in Figure 1, check NH_3 concentrations (kg N ha^{-1}) or NH_3 concentrations (ppb).

We have revised the units of NH_3 concentrations in Figure 1 to 10^{15} molecules cm^{-2}.

L201, in Figure 1, the figure shows the 2019-2020 average or sum, please check similar mistake throughout the manuscript.

The time series of NH_3 columns by GIIRS was between 2019.11 and 2020.10. We showed the spatial variation of the monthly average NH_3 columns at 10 overpass times for GIIRS from 2019-2020. We have revised the similar mistake throughout the paper and made the following changes to the figure captions.

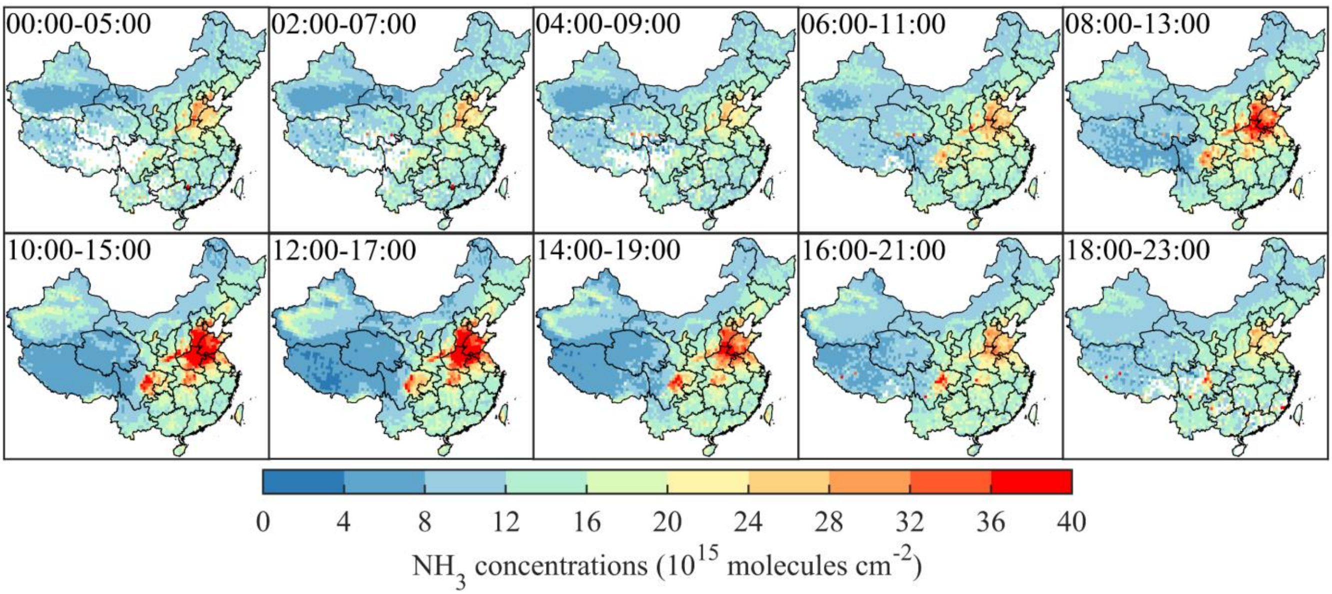

Figure 1. Monthly average NH_3 concentrations for each of the 10 GIIRS overpass time periods during 2019-2020.

L282-283, need the data link or reference.

We have added references as suggested.

"The North China Plain is China's granary, with developed agriculture and animal husbandry, high population densities and strong human activities (including vehicle emissions) (Zhang et al., 2006; Wang et al., 2020a)."

Reference

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