#### **Reviewer 1:**

Zeng et al. present NH3 retrieval results from measurements of the Geostationary Interferometric Infrared Sounder onboard FengYun-4B. Compared to previous work, their data product now allows for studies of the diurnal cycle of NH3, due to the geostationary orbit of the satellite. The topic of the manuscript is certainly within the scope of AMT and of scientific importance. However, before publication in AMT, major revisions are necessary, as described below.

We thank the reviewers for his/her constructive comments and suggestions to improve the quality and clarity of our manuscript. We have made major and careful modifications to the original manuscript according to all the comments and suggestions from the reviewers. The major changes include:

- 1. In the main manuscript, we restructured the manuscript and focused on the GIIRS NH<sub>3</sub> retrieval results using just the strong absorption micro-window, 955-975 cm<sup>-1</sup>, and moved the comparison with retrieval results using 920-940 cm<sup>-1</sup> micro-window to the supplementary materials;
- 2. We have made detailed comparison with IASI's most updated version 4 NH<sub>3</sub> data products for the same period (July to December of 2022), in which we carried out spatial comparison and point-by-point collocation comparison to demonstrate the consistency of our retrievals with IASI data;
- 3. Throughout the revised manuscript, we have enlarged all figure fonts to increase readability.

Item-by-item responses to the specific comments are provided below, in which the reviews' comments are in blue, our responses in **black**, and modifications of the original manuscript are indicated by highlight in yellow in the revised manuscript.

#### **General points:**

- Labels and ticks are for most of the figures very small and hard to see without zooming the PDF to 200%. I would suggest to increase font size and line width considerably.

Throughout the revised manuscript, we have enlarged all figure fonts and line width of figures where necessary to increase readability.

- Until the beginning of Section 4 it was not clear to me, if one retrieval using both microwindows is presented here, or if two retrievals using one of the microwindows each are compared against each other. This should be stated much more explicitly in the beginning of the manuscript. It should be also motivated, why two retrieval approaches are compared against each other here. What should the reader learn from this exercise? Or are there also retrieval results shown using both microwindows (as indicated by the caption of Fig. 10)? I am very confused and I am not sure if I know which kind of retrieval was shown in which part of the manuscript.

In the main manuscript, we have re-structured the paper and focused on the GIIRS NH<sub>3</sub> retrieval results using just the strong absorption micro-window, 955-975 cm<sup>-1</sup>, and moved the comparison with retrieval results using 920-940 cm<sup>-1</sup> micro-window to the supplementary materials (see **Supplementary Text S1** and **Figures S4 and S5**). The motivation of this comparison is to investigate the consistency of the retrieval results using two different micro-windows, and to check if different window may give similar results, as a way to quantify the robustness of the spectra features. From the comparison, we do see high consistency in

results from the two micro-windows. We have added related statements in the revised manuscript and the supplementary materials.

- In Section 3.2, I miss general information about the retrieval. What quantities are derived from the retrieval? Is the NH3 concentration derived as a profile or only as a total column? What about temperature and the other trace gases? Until now, this section mainly collects references to theory. This is also important, but does not help to understand what kind of retrieval is performed in this work. Further, specific advantages and drawbacks of the selected retrieval approach should be at least mentioned here. Also the importance of the a priori profile in case of optimal estimation.

In the revised manuscript, we have added more details of the retrieval algorithm, including the state vector, profile retrieval of  $NH_3$ , and other interfering gases, please see Sections 3.1 and 3.2. We have added **Table 1** to show the parameters in the state vector to be retrieved from the retrieval algorithm.

"The parameters in the state vector to be retrieved from the algorithm are listed in Table 1. Vertical profiles of NH<sub>3</sub> and H<sub>2</sub>O are retrieved, while for minor interference gases, total columns are retrieved by scaling an a priori profile. Other parameters to be retrieved include the surface skin temperature, a scaling factor for the atmospheric temperature profile, and the slope and curve for the surface emissivity. We only retrieve the layers below 200 hPa and use the a priori for layers above to compute total columns."

Two widely used methods for retrieving NH<sub>3</sub> from space have been mentioned in the revised manuscript: "A similar micro-window has been used to retrieve NH<sub>3</sub> using IASI (Clarisse et al., 2010) and GOSAT (Someya et al., 2020), while neural-network-based studies using hyperspectral radiance index (HRI) adopted a much wider window (e.g., Whitburn et al., 2016)."

In addition, we have added the a priori covariance matrix and stressed the importance of the a priori profile in **Section 3.3** in the revised manuscript.

- In Section 4, the retrievals using different microwindows are compared. It is certainly of interest, which spectral region is suited best for the retrieval (or maybe both regions together?), but one important point is missing: There are certainly (temporally and spatially) co-located measurements by IASI, CrIS, ..., which could be used to judge, which of the retrieval approaches better matches the established NH3 columns. I really think this kind of validation approach is missing here. Just saying that microwindow #2 has a stronger signal was already clear by looking at the spectral signature in Fig.2. So what is the point of the comparison of the different retrievals, if not comparing them to independent data?

Thanks for the great suggestion. It is important to carry out comparison with collocated measurements from independent instruments. In the revised manuscript, we have made such detailed comparison with IASI's most updated version 4  $NH_3$  data products for the same period (July to December of 2022). Dr. Lieven Clarisse and Dr. Martin Van Damme who developed the IASI NH3 retrieval algorithm have provided guidance on the comparison and they have joined as co-authors of this manuscript.

Two types of comparisons have been made: (1) Comparison of spatial distribution on representative days. The spatial distribution of NH<sub>3</sub> columns on two representative days: July 07, 2022 (summer, local daytime) when there was mostly positive TC, and December 18, 2022 (winter, local nighttime) when there was

mostly negative TC are compared between GIIRS and IASI; (2) Collocated point-by-point comparison. a spatially and temporally collocated point-by-point comparison between FY-4B/GIIRS and IASI  $NH_3$  retrievals. We consider observations to be collocated when the distance between the centers of the pixels is less than 6km (half of the FY-4B/GIIRS footprint size) and the observation time difference less than 1 hour. In both cases, good agreements are found in general with fitted slopes close to unity.

In addition, we have also conducted an experiment to re-generate a new set of GIIRS  $NH_3$  retrievals using the Gaussian a priori profile provided by IASI to explain the small difference between the two datasets in summer. The result from this experiment demonstrates that the systematic bias between GIIRS and IASI in Summer daytime is caused by the difference in their a priori profile structure.

Please refer to Section 4.3 for the details and results of the comparisons.

- Section 5 seems to be out of place and the title "Discussion" is misleading. In my opinion, this section shows attempts to quantify the errors of the measurements. This is an important task, but it would be interesting to know the uncertainties of the measurements, before diurnal cycles are discussed in Section 4. I suggest to add both subsections 5.1 and 5.2 to section 3, since they also describe the retrieval.

As suggested, in the revised manuscript, we have moved Sections 5.1 and 5.2 to "Section 3.5 Retrieval experiments for quantifying retrieval error."

- Some sources of errors are mentioned in the sections 5.1 and 5.2. Are there any other random or systematic error sources, that are expected to influence the retrieval? I guess the calibration of the spectra should also come with uncertainties, which should be considered for the error of the retrieval results.

We agree that the spectral calibration uncertainty is expected to influence the retrieval. As described in **Section 3.2**, if we just use the spectra noise as described by the NEdT from the GIIRS L1B data product, the averaged reduced  $\chi^2$  value from the optimal estimation NH<sub>3</sub> retrieval is systematically larger than 1.0, suggesting the forward model error is not properly characterized by the spectra noise. So we have enlarged the spectra noise by a factor of 2.0. This extra noise represents the unaccounted uncertainty from the forward model and absorption spectroscopy by the original instrument noise alone, which may come from the spectral calibration uncertainty.

# **Specific points: (**line numbers are given in the beginning of each point)

- 14: "our retrievals are implemented using two different absorption micro-windows": For me this sounds like both microwindows are used in the same retrieval. The following sentences sound more like two retrievals based on the two microwindows are performed and compared. Please clarify. (see also general comment)

In the revised manuscript, we have re-structured the manuscript and focused on the retrieval results from 955-975 cm<sup>-1</sup> micro-window only and moved the comparison using two micro-windows to the supplementary materials.

- 17: Typo: no -> No

Changed.

- 28-36: In this introduction to NH3, it would be also helpful to mention typical atmospheric loss processes of NH3.

In the revised manuscript, to mention the loss process of NH<sub>3</sub>, we added "In addition, ammonia reacts with acids (e.g., H<sub>2</sub>SO<sub>4</sub>, HNO<sub>3</sub>) and produces ammonium containing aerosols that degrade air quality (Seinfeld and Pandis, 2006)."

- 37-44: For completeness, it would be also worthwhile to mention that NH3 has been measured in the upper troposphere of the Asian Monsoon by infrared limb sounders.

In the revised manuscript, we added "In addition, NH<sub>3</sub> has been detected in the Asian summer monsoon upper troposphere using the emission spectra from the infrared limb sounder Michelson Interferometer for Passive Atmospheric Sounding (MIPAS; Höpfner et al., 2016)."

The following reference has been added:

Höpfner, M., Volkamer, R., Grabowski, U., Grutter, M., Orphal, J., Stiller, G., von Clarmann, T., and Wetzel, G.: First detection of ammonia (NH3) in the Asian summer monsoon upper troposphere, Atmos. Chem. Phys., 16, 14357–14369, https://doi.org/10.5194/acp-16-14357-2016, 2016.

- 70: Suggestion: "developed by the group (Zeng et al., 2022) based" -> "developed by Zeng et al., (2022), based"

Changed as suggested.

- 70: Typo: optical -> optimal

Changed.

- 71: absorption feature -> absorption features

Changed.

- 72: The sentence "The primary goals are ..." feels out of place there. It is between two sentences explaining the spectral properties of the retrieval. The sentence could be moved to the beginning of the paragraph (before the sentence "The retrieval algorithm uses ...").

Changed as suggested.

- 74: "our retrievals are ...": It is still not clear to me, if the authors are explaining a single retrieval setup using both microwindows (in this case, this phrase should be used in

singular "our reatrieval is ..."), or if two different retrieval approaches with different microwindows are compared (in this case, this should be written much more specific).

In the main manuscript, we changed to use just the strong absorption micro-window, 955-975 cm<sup>-1</sup>, and moved the comparison with retrieval results using 920-940 cm<sup>-1</sup> micro-window to the supplementary materials

- 75: "their difference in retrievals": Same comment as for line 74.

Please see our response above.

- 90: The introduction of Fig. 1a is misleading. It should be clearly stated in the text that this panel shows NH3 inventory data and no measurements!

We added "..., **as indicated by the bottom-up inventory map** ..." to explicitly state that the data are bottom-up inventories.

- 93: "GIIRS is in principle capable of measuring trace gases": Which other trace gases than NH3 have been retrieved or are planned to be retrieved in the near future?

We added "..., including NH<sub>3</sub> and carbon monoxide (Zeng et al., 2022), ..." in the revised manuscript.

- 99: "comparable to existing infrared sounders": I suggest to give some examples of NedT for infrared sounders, which also measure NH3 columns.

In the revised manuscript, we added "The low instrument noise for FY-4B/GIIRS, comparable to existing infrared sounders (e.g., ~0.2K@280K for IASI and ~0.04K@280K for CrIS; Van Damme et al., 2014; Shephard et al., 2020), makes it possible to accurately retrieve NH<sub>3</sub> over East Asia."

- 100: I do not understand the formulation "provide strong constrain" in this context, but I am no native speaker.

We have rephrased it to "makes it possible to accurately retrieve NH<sub>3</sub> over East Asia."

- Figure 1c: It would be helpful to zoom into the used microwindows (in an additional panel?) and highlight the spectral features of NH3, which are used for the retrieval.

A sub-figure has been added to Figure 1(c) that zooms into the used micro-window. In the caption of Figure 1, we added "The micro-window for NH<sub>3</sub> retrieval is also shown in red rectangle and as inset. The NH<sub>3</sub> absorption features are shown in Figure 2."

- 124: It would be good to also mention the possible interfering species and discuss a bit the content of Figure 2.

We added "The micro-window contains strong NH<sub>3</sub> absorption features that are distinguishable from important interference gases (CO<sub>2</sub>, O<sub>3</sub> and H<sub>2</sub>O) in the absorption window, as demonstrated

in Figure 2 from a sensitivity experiment that compares absorptions of NH<sub>3</sub> and perturbed interference gases."

- 128: Please define the acronyms ECMWF and ERA5

Defined.

- 130: Pleas define the acronym CAMS. Are the concentrations or the fluxes used from this data set? Why did the authors decide for this specific CAMS data set? Why are other trace gas data taken from ERA5 instead?

The acronym CAMS has been defined. The concentrations are used from this dataset. The ERA5 is primarily used for gases ( $H_2O$  and  $O_3$ ) because it has a higher temporal resolution (1-hour). For gases that are not available from ERA5, we extract the data ( $CO_2$ ) from CAMS instead.

- Figure 2: N2O is mentioned to be considered for the radiative transfer model, but there are no spectral features shown in this figure. Why?

We have removed  $N_2O$  as an interference gas since it has no significant contribution in this spectral region.

- 134: "Since the NH3 is more ...": It seems like the retrieval grid is introduced and motivated here. I do not understand, why this part starts with a comparison to CO. Maybe, the retrieval is compared to previous work of the authors? I would suggest to start a new paragraph here and then explain, how the retrieval grid is chosen for the NH3 retrieval, motivate the choice and then compare it to similar retrieval setups with the same instrument. I am also missing the information, if the retrieval is on pressure or altitude grid, since some numbers are given in pressure, others in kilometers.

In the revised manuscript, we have started a new paragraph and rephrased the sentences to include all necessary information:

"Since NH<sub>3</sub> is short-lived and highly concentrated in the PBL, we therefore only retrieve the layers below 200 hPa. The forward model uses fixed vertical grids with equally separated layers with similar thickness (about 1 km for layers below 200 hPa and about 5 km for layers above), which is close to the grid settings in Hurtmans et al. (2012) and Clough et al. (2005). The thickness of the bottom layer is variable and determined by the surface pressure of a specific location. The number of layers below 200 hPa ranges from 7 (for high altitude regions such as the Tibet Plateau) to 11 layers (for low altitude regions such as the ocean)."

The retrieval is on pressure grid, as described in the rephrased paragraph.

- 136: "The number of layers for retrieval ranges from 7 (at high altitude) to 11 layers (at low altitude).": I do not understand this sentence. Maybe it becomes clearer, if the altitude grid itself is introduced properly before.

We have rephrased the sentence to: "The number of layers below 200 hPa ranges from 7 (for high altitude regions such as the Tibet Plateau) to 11 layers (for low altitude regions such as the ocean)"

The retrieval grids are also shown in Figure 3(a).

- 147: "The auxiliary parameters include ...": I do not understand the meaning of "auxiliary parameters" in this context. Does that mean that all of these quantities are used by the forward model? Or are some of these parameters also fitted? What exactly are "scale factors"? Are these factors adjusted by the fit?

In the revised manuscript, we have rephrased the descriptions to:

"The parameters in the state vector to be retrieved from the algorithm are listed in Table 1. Vertical profiles of NH<sub>3</sub> and H<sub>2</sub>O are retrieved, while for minor interference gases, total columns are retrieved by scaling an a priori profile. Other parameters to be retrieved include the surface skin temperature, a scaling factor for the atmospheric temperature profile, and the slope and curve for the surface emissivity. We only retrieve the layers below 200 hPa and use the a priori for layers above to compute total columns."

The scaling factor for the atmospheric temperature profile is adjusted in the spectral fitting process.

- 149: "...which maximize the a posteriori probability given the FY-4B/GIIRS spectra." I do not understand this formulation. Please rephrase.

We have rephrased it to: "which minimizes the spectral fitting error."

- 157: A\_ij has not been introduced until here. Suggestion: "where each element A\_ij of A represents ..."

Changed as suggested.

- 160: Suggestion: "by 2.0 times" -> "by a factor of 2.0"

Changed as suggested.

- 175: "(2) it is not ...": I am not sure, if I understand this correctly. Are the authors trying to say that they would need to rely on model profiles (which may have an incorrect diurnal cycle), if they want to use time dependent a priori profiles?

We have rephrased it to: "(2) it is not applicable to get a reasonable a priori estimate for all hours in a day from just the spectra (e.g., using channel brightness temperature difference as in Shephard et al. (2011) and Warner et al. (2016)) without relying on model simulations, ..."

- 178 and following: I am not sure, if I understood this correctly: Only one profile is used for all of the retrievals. This profile is an average over all land regions in the given lat/lon boxes for 2022. Please try to formulate more precisely here!

We have rephrased the related statement to: "The single a priori NH<sub>3</sub> profile, as shown in Figure 3(a), for all retrievals in the retrieval algorithm is derived from NH<sub>3</sub> simulations ..."

- 180: Since the sentence before was discussing a model, it should be "One year of simulation" instead of "One year of measurements".

Changed. It should be "One year of simulation".

- Figure 3: The error bars of the average NH3 profile reach negative mixing ratios. How is that possible for model simulation results? Is the profile averaged using the mean of all profiles? Or the median? Or something different? Further, which kind of mixing ratio is shown here? Volume mixing ratio? Mass mixing ratio? Please add!

The following statement has been added to the revised manuscript to introduce these in more details: "The negative value toward the lower end of the error bar does not have physical meaning, it is caused by the large standard deviation derived from model simulations that do not strictly follow a normal distribution. The a priori total NH<sub>3</sub> column is about  $1.5 \times 10^{16}$  molecules/cm<sup>2</sup>. To construct the correlation matrix, we used a correlation length of 3 km based on our analysis of the GEOS-CF reanalysis. Most of the layers show correlation lengths between 1 to 3 km and we use upper bound (3 km) to increase the stability of the retrieval system. The covariance matrix calculated based on the a priori error and the correlation matrix is shown in Figure 3(b)."

This is volume mixing ratio. The x-label has been changed.

- Section 3.4: I would expect to perform cloud filtering before doing the retrieval to avoid unnecessary fits of cloud contaminated spectra. I think the subsection headline "postfiltering" is not adequate for the cloud filtering part then. Further, I would expect this part earlier in section 3, since it is performed before the retrieval itself.

Thanks for pointing this out. In the revised manuscript, we have moved the cloud screening part to the end of Section 2.

- Figure S1: It would be more interesting to see the histograms before applying the filters and with an enlarged x-axis.

We have added the histogram before applying the filtering in the Supplementary Figure S2.

- 199: Suggestion: "suggesting satisfying goodness of fit." -> "suggesting good fit quality."

We have changed it to: "suggesting a satisfactory goodness of fit".

- 200: I do not understand the formulation "do not have enough constrain from the observed spectra" in this context. Please rephrase.

We have rephrased it to: "In the following analysis, an extra filter based on DOFS may apply to exclude data with low DOFS and, therefore, low information content extracted from the observed spectra."

- 215: "Fortunately, no large systematic bias is observed ...": I think, there is a banana-shape feature of the correlation (but I may be wrong, see my comment on Figure 4). If there is this kind of banana-shape, what does this mean for the retrieval performance? How do you know, which microwindow selection is better? As already mentioned in the general points, I think such correlations are more helpful against independent measurements, e.g. from IASI, CrIS, ...

In the revised manuscript, the comparison of NH<sub>3</sub> between two micro-windows has been moved to the **Supplementary Figure S5**. One-to-one lines have been added in the updated figures.

The scatter plot appears to be a banana-shape mainly because of the saturated color scheme used for the data points. In the revised manuscript, the figure has been re-plotted using a color bar with log10 scale. We have also added an extra figure with DOFS>0.7 to demonstrate how the agreement may improve with retrievals with high DOFS. The revised figure shows that the two datasets agree very well, and there is no such "banana" shape in the data. The data agreement can also be seen in the histogram figures of NH<sub>3</sub> difference, that shows an even distribution without systematic bias. For the comparison with DOFS>0.7, we see that the agreement improves. The comparison suggests that the two retrievals agree well with each other, and no significant bias exist, especially for retrieval with high DOFS.

In the revised manuscript, we restructured the paper and focused on the 955-975 cm<sup>-1</sup> micro-window only, which shows a higher DOFS and has been widely adopted by several other retrieval data products. In addition, we have made detailed comparison with IASI's most updated version 4  $NH_3$  data products for the same period (July to December of 2022). Please refer to **Section 4.5** for the details of comparison.



Part of Figure S5. (b) the retrieved NH<sub>3</sub> columns filtered by DOFS>0.5. For the comparison of columns, in total, 1.1 million data points are available. The correlation coefficient between the two column datasets is 0.82 with a root-mean-square-error of  $9.2 \times 10^{15}$  molec/cm<sup>2</sup>. The histogram is also shown; (c) the retrieved NH<sub>3</sub> columns filtered by DOFS>0.7. In total, 0.5 million data points are available for comparison. The correlation coefficient is 0.86 with a root-mean-square-error of  $8.6 \times 10^{15}$  molec/cm<sup>2</sup>. The mean errors are  $2.0 \times 10^{15}$  and  $2.7 \times 10^{15}$  molec/cm<sup>2</sup>, respectively, for (b) and (c).

Figure 4: I think, a 1:1-line would be very helpful here to guide the eye. For me it looks like there is a banana-shape in both correlations, but this is not discussed at all in the text. Maybe my eye is wrong here, but a 1:1-line would help!

In the revised manuscript, the comparison of NH<sub>3</sub> between two micro-windows has been moved to the **Supplementary Figure S5**. One-to-one lines have been added in the updated figures. Please see our responses to your last comment.

- 229: I suggest to replace "non-source" by "background"

Changed as suggested.

- 239: "A typical "butterfly" shape can be seen in almost all cases." What about the other cases, e.g. North-India in July? A short comment would be helpful.

In North India in July, there was no enough data are available. Short comment "**except for North India in July when there are much less observations due to clouds**" has been added to the revised manuscript.

- 244-251: Fig. S2 shows very large TC for the Tibetan Plateau for BJT 14-15 h. However, the DOFS are very low here (almost zero). How does this agree with the mentioned "strong correlations between DOFS and TC"?

As we explain in the text, the DOFS is a function of both TC and NH<sub>3</sub> abundance. In Tibetan Plateau although it has large TC, the DOFS is low because of its very small NH<sub>3</sub> columns.

Related statement has been rephrased in the revised manuscript:

"This strong correlations between DOFS and TC or NH<sub>3</sub> abundance is also reflected in the spatial maps of DOFSs ..." and "The source regions in North China Plain and North India have higher DOFSs than other non-source regions, such as Tibetan Plateau although it has large TC."

- 256: "We can see the diurnal AK values ...": First, the quantity "averaged averaging kernel row" should be introduced and what we can learn from it. Further, in Fig. 7 the x-axis-label should be corrected to "averaged averaging kernel row". I also miss a altitude-resolved averaging kernel plotted for different altitudes without averaging it, at least for an example in the supplement.

We have changed it to "Averaged averaging kernel diagonal vectors", which are measures of the DOFS for each vertical layer. The x-axis label has also been changed accordingly. We have rephrased the statement to "... using the corresponding averaging kernel diagonal vectors, which are measures of the DOFS for each vertical layer."

In addition, an example of an altitude-resolved averaging kernel plotted for different altitudes without averaging it is shown in the **Appendix Figure A1**.

- 293: "The data gaps ...": I would have expected that cloud filtering would also considerably impact data availability, in particular in North India during the Monsoon season. In fact, in Fig.

10, no time series can be shown for North India in July-August. I miss a comment for this at all in Section 4.3.

Thanks for your suggestion. In the revised manuscript, we added the following statement: "The cloud filtering has considerably impacted data availability, in particular in North India during the Monsoon season in July-August."

- 297: "Interestingly, ...": Comparing the steps of the diurnal cycle of different months in Figures 8 and 9 with the half-year average inventory in Fig. 1a seems not like a fair comparison. On the one side, diurnally- and seasonally-resolved satellite measurements with data gaps in North India (a region specifically mentioned in the comparison) are compared to a half-year average. It's apples and oranges. Further, in July-August, there are times, in which North China Plain shows higher NH3 columns than North India.

In the revised manuscript, we have removed the comparison with bottom-up inventories, and rephrase the statement to be: "As explained in Wang et al. (2020), the causes of high NH<sub>3</sub> loading in North India, which are slightly different from that in the North China Plain, are due to the weak chemical loss and weak horizontal diffusion in North India."

### - 307: Typo: capture -> captured

Changed.

# - 308: "In addition, ...": To which months does this sentence refer to? Further, would more precipitation also wash-out the highly water soluble NH3 from the atmosphere?

It is referring to the summer months. We have rephrased the sentence to "In addition, the relatively low temperature and higher humidity **in the nighttime, relative to the daytime**, contribute to the conversion from NH<sub>3</sub> to particulates that leads to a lower NH<sub>3</sub> concentration."

The removal of  $NH_3$  from the atmosphere is call wet deposition. It is an important mechanism of  $NH_3$  loss. However, it is not the main reason for the diurnal cycle as discussed in this paragraph.

- Section 4.3: This section should be restructured. In the first paragraph, the diurnal cycles shown in Fig. 10 are already briefly introduced, then the next paragraph starts with some explanations of the observed diurnal cycles, while in the end of the section, the diurnal cycles are again introduced again, but in more detail. I suggest to start a new paragraph for the description and discussion of Fig. 10. In this paragraph, please first describe, what can be seen in the figure, and then explain, why it makes sense, what we see.

We have re-structured the section (now Section 4.2) following your suggestions. The explanation of the observed diurnal cycles has been rephrased as:

"The general diurnal cycle of NH<sub>3</sub> columns can be primarily explained by three possible driving factors, as concluded in the summary in Clarisse et al. (2021), including the day-night difference in

agriculture activities as a major source of NH<sub>3</sub>, the temperature dependence of NH<sub>3</sub> emissions driven by diurnal and seasonal temperature changes, and the conversion between NH<sub>3</sub> gas and particulate driven by the day-night change of meteorological conditions. These can be used to interpret the quantitative analysis of the diurnal cycle as shown in Figure 10 for North China Plain and North India."

Please see Section 4.2 in the revised manuscript for the details.

- Figure 8/9: I do not understand, why the order of the panels is different compared to Fig. 5. It would be easier, if this would be consistent throughout the manuscript. E.g. keep it in a way that the diurnal variation is visible within a row and the seasonal changes are visible within a column. Maybe it would be best to change Fig. 5 (and S2).

We have changed the structure of **Figures 8** and **9** such that, to be consistent throughout the paper, the rows represent different months.

-339: "The error bar represents one standard deviation.": So the error bars rather show variability within the region than an error/uncertainty of the measurement? Since this variability is quite large compared to the observed diurnal cycle, this variability should be discussed in Section 4.3 somewhere.

We added the following statements in the revised manuscript:

"Note that the variability of NH<sub>3</sub> columns within the region is large as shown by the error bars (one standard deviation). This large variability is a result of NH<sub>3</sub>'s short life time and the spatial heterogeneity of its emissions."

- Section 5.1: In the end of the section, a noise error is estimated for the retrieval. It would be interesting, how this error is for the diurnal cycles shown in Fig. 10 compared to the signal of the diurnal cycle itself. Is the error larger or smaller than the variability (which is shown in Fig.10 with error bars)? Is the amplitude of the diurnal cycle larger than the scaled random error from this exercise?

To compare the uncertainty of the retrievals with the diurnal variabilities, we added the following statements: "Moreover, when compared with the averaged uncertainty of a single retrieval (1.37×10<sup>16</sup> to 1.67×10<sup>16</sup> molec/cm<sup>2</sup> as derived from the retrieval experiment in Section 3.5), the day-night contrast of the averaged diurnal variations of NH<sub>3</sub> columns as shown in Figure 10 may not be significant for the North China Plain in September-October and the North India in November-December."

- Figure 11: For me, it looks like there is a low bias for the NH3 retrieval: There are considerably more dots far away from the 1:1 line in the lower-right half of the plot than in the upper-left part. And a non-negligible number of the extreme outlier have a DOFS > 0.5. This should be discussed in the text. Do the authors have any idea about the source of this systematic error?

The data points on the lower-right half are mostly retrievals without high information content (low DOFS). As a result, these retrievals are close to the a priori value, which is about  $1.5 \times 10^{16}$  molecules/cm<sup>2</sup>.

For those outliers that have high DOFS but with poor agreement, they are likely caused by large difference of the real NH<sub>3</sub> profile and the a priori profile. Because the satellite retrieval cannot resolve all layers, the extrapolation of the retrieval profile scaling may lead to large bias if the truth NH<sub>3</sub> profile is far away from the a priori profile. Related statements have been added to the revised manuscript.

## - 385: I think, the reference should be to Section 5.1

Changed.

- 386: In Section 5.1, also absolute uncertainties are given. That would be also helpful here.

Added. The absolute root-mean-square errors for both cases are about the same  $3.4 \times 10^{15}$  molec/cm<sup>2</sup>.

Figure 12 b/c: A 1:1-line would be helpful here. Further, it seems like the correlation splits into "branches", in particular for Fig. 12b. So, the correlation does not evenly scatter around the 1:1 line. This should be discussed in the text. Are there any explanation for this behavior?

The Figure has been moved to the **Supplementary Figure S6**. 1:1 line has been added to (b) and (c). As you mentioned, the correlation does not scatter along the 1:1 line. For (b) with retrieved NH<sub>3</sub> column using the reduced PBL excess profile, the retrievals are underestimated compared with the original retrievals. For (c) with retrieved NH<sub>3</sub> column using the enhanced PBL excess profile, the retrievals are overestimated compared with the original retrievals. These differences are mainly caused by the a priori total columns.

- 429: The given link only leading to the FY-4B/GIIRS CO data, but no NH3 data is available there. Please add the missing data to the website, or give the correct link. For this and all other mentioned data resources, it would be further better to have the data versioned and tagged with a DOI.

The associated  $NH_3$  datasets and a data user guide have been uploaded to the PKU Opendata repository (<u>https://opendata.pku.edu.cn/dataverse/FYGEOAIR</u>). The designated DOI for this dataset is: <u>https://doi.org/10.18170/DVN/VJ4MLO</u>.