

Reviewer 2

The paper by Zeng et al., presents a NH₃ retrieval product from the GIIRS infrared sounder on board FY-4B. Geostationary NH₃ measurements are novel and of high interest to the scientific community. The paper is not bad, but there is certainly a lot of room for improvement, both in content, form and depth, which is why I would recommend a major revision, after which the paper needs to be re-reviewed. I would also encourage the authors not to see the revision as a hurdle that needs to be overcome to publish, but rather as an opportunity to improve the paper, and to increase its impact.

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 NH₃ retrieval results using just the strong absorption micro-window, 955-975 cm⁻¹, and moved the comparison with retrieval results using 920-940 cm⁻¹ micro-window to the supplementary materials;
2. We have made detailed comparison with IASI's most updated version 4 NH₃ 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.

A. Major Comments

A.1 General comments on the figures

- As a general rule, the font size of the figures should match that of the text, or slightly smaller. A lot of text is currently simply unreadable on print-out.

Throughout the revised manuscript, we have enlarged all figure fonts to increase readability.

- Again, as a general rule, try to decide whether a figure should be a one-column figure or a two column-figure in the final manuscript, and then utilise exactly the full width or exactly half of the full width of the text. This applies at least to Fig 1C (bottom row could be full page width), Fig 2, Fig 4, Fig 6, Fig 7 and Fig 10.

We have changed the size of these figures. **Figure 1(c)** has been changed to full page width. **Figures 2, 4, 6, 7 and 10** have been enlarged to full page width.

- For large multipanel figures with the same x and y axis, the axis tick labels and axis labels can be removed from the middle panels. This in effect increases drastically the space available for the actual figures. For instance in Figure 8, the degrees East can be removed on top, between the first and second row, and the second and third row (in this way only keep it at the bottom). Likewise, the degrees North can be removed between the first and second column and between the second and third column, and only kept on the left side.

After this, all panels can be enlarged. This applies to Figure 5, Figure 6, Figure 8, Figure 9 and Figure S2. For Figure 5, one colourbar suffices, which can go to the bottom, again allowing for larger and more visible panels.

- Wherever possible, please export figures in a vector format (https://en.wikipedia.org/wiki/Vector_graphics, e.g. pdf), to avoid pixelation of lines and text, especially visible when zooming in on screen. For instance figure 10 or 12 (but ideally for all figures).

In the revised manuscript, we have removed the repeated x- or y-tick labels in **Figure 5, 8, 9, and S2**, and enlarged the figures using the extra space. We have remade the figures to use vectorized format where necessary to increase readability.

A.2: Other major comments

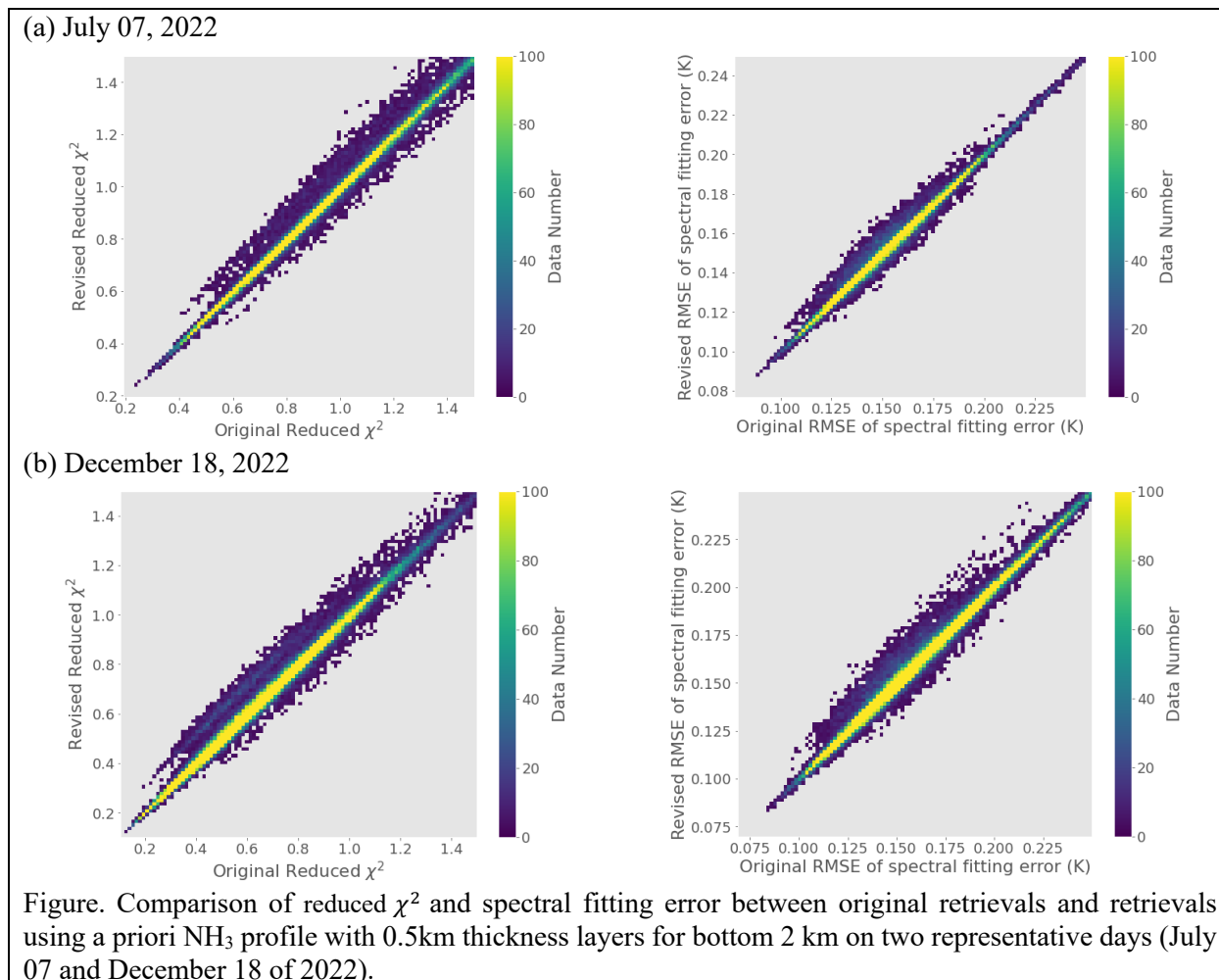
- 1 km layering at the bottom of the atmosphere is not sufficient (at least 500 m is required below 2 km). 1 km is not sufficient for modelling fast varying temperature (e.g. thermal inversions at night), nor for representing the fast NH₃ (especially at night) and H₂O variations (always). I can confidently say that the fit residuals will decrease once a finer layering is adopted (even just for the H₂O fit). The fact also that there was a need to multiply the spectral noise by two, gives a hint that the fits can and should be improved. I realize that redoing all the fits is a major undertaking, but worth it, if the product is going to be further extended in time and used by the community. If a coarse spectral resolution emissivity database was used, I would also encourage to look for better alternatives.

We have tested using the suggested a priori with 0.5km thickness layers for bottom 2km and re-run all retrievals on 2 representative days (July 07 and December 18 of 2022). Theoretically, as the reviewer pointed out, that this would significantly improve the spectral fitting. This is given that the spectra contain enough information to resolve the NH₃ vertical structure in the bottom atmosphere. However, the spectra as in our retrieval can have less than one degree of freedom, suggesting the spectra is not able to correctly resolve lower atmospheric structures, similar to most existing infrared sounders. This can be seen from the comparison of the reduced χ^2 and RMSE of spectral fitting errors from both retrievals (please see the following figure). The reduced χ^2 and the RMSEs of spectral fitting errors from the updated retrievals are basically the same as the original results, suggesting the use of 0.5km thickness layers for bottom 2km may not improve the spectral fit. We therefore continue to use the current vertical grid settings.

For the spectra noise that has been multiplied by 2.0, this is more likely due to the uncertainty in the forward model (such as the used ABSCO lookup table), which is close to random, not systematic.

For the emissivity database, over the narrow micro-window in our retrieval, it is sufficient to assume that the emissivity does change significantly. In our retrieval, we have fit a linear trend and a curve to the emissivity to allow it to change as a function of wavenumber.

We have added relate statements to the revised manuscript.



- Section 3.1 "forward model" already introduces elements of the retrieval. It makes it confusing and not well structured. E.g. the layering used for the atmosphere doesn't necessarily have to match the layering used for the retrieval.

You are right that the layers for the retrieval algorithm can be different from those in the RT model. In the revised manuscript, we have added details about the atmospheric layers used in the forward RT model and for the retrieval algorithm. The rephrased statements are:

“Since the NH_3 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).”

- Section 3.2 I think a comprehensive table with all the forward/retrieval parameters would go a long way in making this section more understandable, and the paper more reproducible. For an AMT paper it is essential that these important technical aspects are

fully transparent. E.g. a table with 1st column: Parameter name, 2nd column, levelling used in forward model, 3rd column: levelling used for the retrieval, 4th column: a priori, 5th column covariance matrix (i.e. % variability if diagonal, and description of off-diagonal elements, e.g. correlation length). This table should at least include all parameters that are retrieved, but also the important atmospheric and surface terms (temperature/pressure)

Thanks for your great suggestion. Part of Section 3.1 has been rephrased to comprehensively introduce the input variables necessary to drive the RT model. In addition, we have added Table 1 that includes descriptions of all parameters in the state vector to be retrieved from the retrieval algorithm. The variable names, no. of variables, a priori values, a priori uncertainty, and necessary descriptions are included. The following statements have been added to Section 3.2:

“The parameters in the state vector to be retrieved from the algorithm are listed in Table 1. Vertical profiles of NH₃ and H₂O 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.”

- Figure 3: please provide an additional panel with the a priori covariance matrix. Showing the a priori NH₃ profile is only showing half of the story. How was it calculated? Also from model output? Please discuss this matrix at some length, as the choice of covariance matrix is absolutely key (both diagonal and off-diagonal elements).

The a priori profile and the a priori covariance matrix are important inputs in the optimal estimation-based retrieval algorithm. In the revised manuscript, we added descriptions of the details of the used a priori and the covariance matrix. The following statements have been added or rephrased:

“The single a priori NH₃ profile, as shown in Figure 3(a), for all retrievals in the retrieval algorithm is derived from NH₃ simulations from the Goddard Earth Observing System composition forecast (GEOS-CF; Keller et al., 2021) model developed by NASA's Global Modeling and Assimilation Office (GMAO). One year of simulation in 2022 is used to get the mean and standard deviation of NH₃ vertical distribution. To avoid over sampling of the background regions, only simulations in the representative land regions in east Asia (20°-60°N and 110°-120°E) and south Asia (20°-40°N and 70°-100°E) are used. 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 NH₃ column is about 1.5×10^{16} molecules/cm². 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).”

- Section 4: comparing two retrievals is very interesting, but obviously also opens the door to a lot of questions. Currently the reader remains rather unsatisfied.

In the main manuscript, since the main focus of this paper is the development of an NH₃ retrieval algorithm for FY-4B/GIIRS, we have re-structured the paper and focused on the GIIRS NH₃ retrieval results using just the strong absorption micro-window, 955-975 cm⁻¹, and moved the comparison with retrieval results using 920-940 cm⁻¹ micro-window to the supplementary materials (see **Supplementary Text S1 and Figures S4 and S5**).

* For the column retrievals, would it not be better to apply averaging kernels in the comparison, to remove the impact of the a priori all together (and to what extent it is used by both retrievals). Perhaps both could be shown (with and without AVKs applied). I am not sure what is the best approach (which averaging kernel to apply to what), but there is certainly literature out there to guide you.

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 the results from the two micro-windows. The small difference between the two micro-windows may be traced back to the slightly different sensitivity. We have added related statements in the revised manuscript and the supplementary materials.

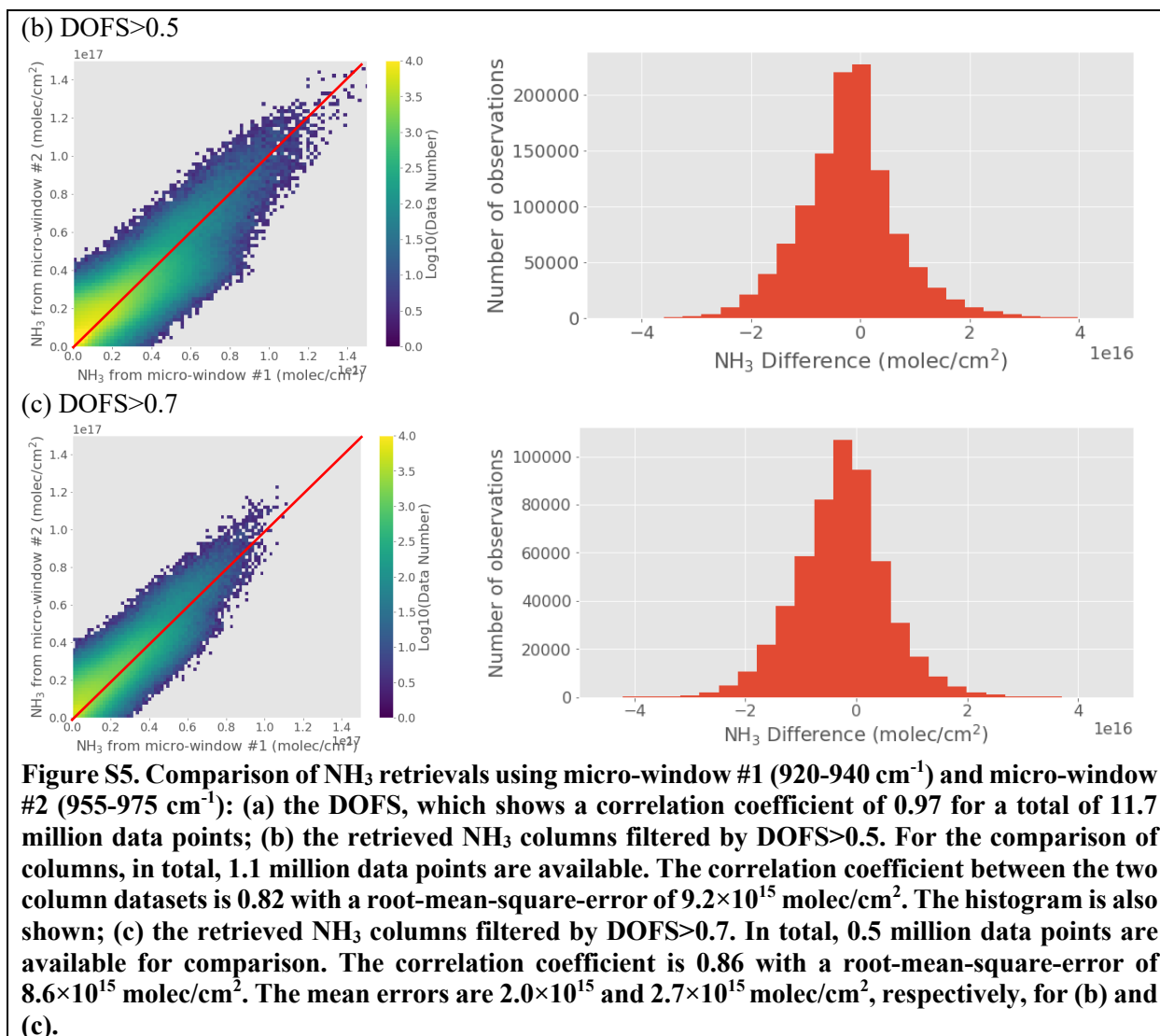
Since the main focus of this paper is the development of an NH₃ retrieval algorithm for FY-4B/GIIRS, we did not apply the AK correction, but instead mentioned it in the supplementary text as a potential cause of the difference between the two retrievals.

* Figure 4b is misleading, as it is highly saturated with the colourbar as-is. I would propose to use a colourbar with log scale (e.g. 1 to 10000 or higher). Also, the vast majority of observations have a column below 5×10^{16} , and for these points, there seems to be a clear bias between the two retrievals, where the micro-window 1 is low-biased. I disagree strongly that "no large systematic bias is observed" (perhaps non-surprisingly because of the lower information content - but in that case applying averaging kernels would show this, and the bias should disappear). I would not show observations above 1×10^{17} , to allow higher level of detail for the low columns.

In the revised manuscript, the comparison of NH₃ between two micro-windows has been moved to the **Supplementary Figure S5**. One-to-one lines have been added in the updated figures.

Following your suggestion, we plot the scatter plots using a colourbar with log scale. 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 significant "banana" shape in the data. The data agreement can also be seen in the histogram figures of NH₃ 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⁻¹ micro-window only, which shows a higher DOFS and has been widely adopted by several other retrieval data products. We have made detailed comparison with IASI's most updated version 4 NH₃ data products for the same period (July to December of 2022). Please refer to **Section 4.5** for the details of comparison.

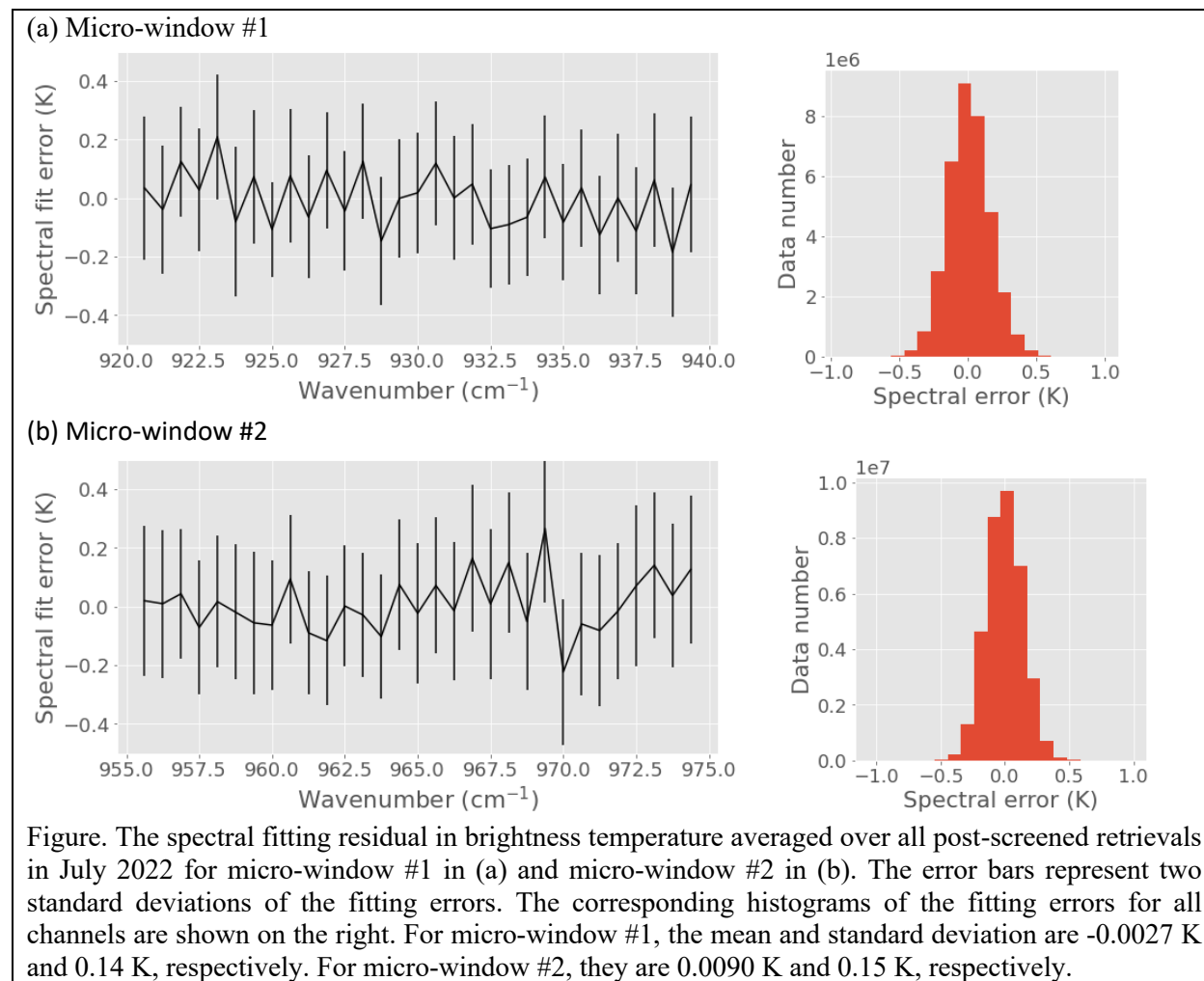


* For the comparison, it would be good to add a figure showing the mean residual (calculated-observed as a spectrum) for one or two selected regions, for both retrievals. For a well-behaved retrieval, there should be no systematic features, and noise should average out.

The following figure shows the spectral fitting residual in brightness temperature averaged over all post-screened retrievals in July 2022 for micro-window #1 in (a) and micro-window #2 in (b). The error bars represent two standard deviations of the fitting errors. The corresponding histograms of the fitting errors for all channels are shown on the right. For micro-window #1, the mean and standard deviation are -0.0027 K and 0.14 K, respectively. For micro-window #2, they are 0.0090 K and 0.15 K, respectively.

The spectral fitting errors in brightness temperature (BT) are small but show systematic patterns, that are persistent among observations at different hours, can be seen from the averaged fitting residual from all spectra. These patterns are likely caused by the uncertainty in molecular absorption properties in the

ABSCO lookup tables. Fortunately, these patterns are not correlated with the absorption feature of the target gas NH_3 and are not expected to significantly affect the retrievals of NH_3 .



* Did you also try to retrieve using the entire NH_3 band? What were the results? Can both retrievals be combined to provide a combined column? If columns are simply averaged, what to do with the corresponding retrieval uncertainty and averaging kernel? A discussion is missing on this. In the NH_3 dataset that is shared online, what are the variables that are included?

The retrievals were not done using the entire band. We expect the retrievals using a wider window may show difference that can be traced back to the sensitivity difference. In the main manuscript, we focused on the GIIRS NH_3 retrieval results using just the strong micro-window, $955\text{-}975\text{ cm}^{-1}$, and moved the comparison with retrieval results using $920\text{-}940\text{ cm}^{-1}$ micro-window to the supplementary materials.

In the file shared online, the variables include:

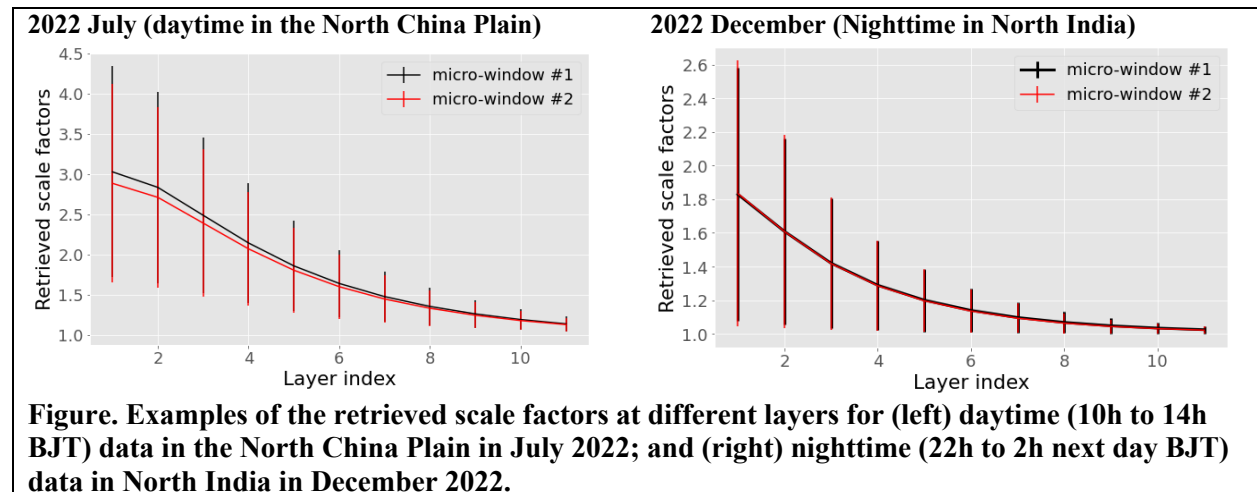
- 1) [Obs_Lat]: latitude of the observation
- 2) [Obs_Lon] : longitude of the observation
- 3) [Obs_Hour] : decimal hour (UTC) of the observation

- 4) [Obs_SZA] : solar zenith angle of the observation (degree)
- 5) [Obs_VZA] : viewing zenith angle of the observation (degree)
- 6) [AP_SurfSkinT]: a priori for the surface skin temperature (Kelvin)
- 7) [AP_SurfP]: a priori for the surface pressure (hPa)
- 8) [AP_SurfEmissivity]: a priori for the surface emissivity
- 9) [AP_AirMidLayerPres]: atmospheric pressure at mid-layer (hPa)
- 10) [AP_AirMidlayerTemp]: atmospheric temperature at mid-layer (Kelvin)
- 11) [AP_NH3_ColumnProf]: a priori for NH3 partial column profile (molecules/cm²)
- 12) [AP_NumRetLayer]: number of layers used in the retrieval algorithm (<=11 layers)
- 13) [AP_NumPresLayer]: number of layers in the RT model (19 layers)
- 14) [Ret_NH3_ColumnProf]: Retrieved NH3 partial column profile (molecules/cm²)
- 15) [Ret_SurfT]: Retrieved surface skin temperature (Kelvin)
- 16) [Ret_AirMidlayerTemp_SF]: Retrieved scale factor for [AP_AirMidlayerTemp]
- 17) [Ret_RedChi2]: Reduced Chi square from the retrieval algorithm
- 18) [Ret_IfConverge]: Indicator of converge (1) or not (0)
- 19) [Ret_NH3_ErrorDiag]: The diagonal of retrieval error covariance matrix for NH3
- 20) [Ret_NH3_AverageKernel]: averaging kernel matrix for NH3
- 21) [Ret_NH3_FitRes_Rad]: spectral fitting error relative to the spectral continuum
- 22) [Ret_NH3_FitRes_BT]: spectral fitting error in brightness temperature (Kelvin)

We have added a data user guide in the shared file online

* Are there differences in retrieved profiles? Could you again for selected region(s) show the average retrieved NH3 profile (+ a priori?) with both windows? Alternatively, can you show the averaged scaling factor profile (retrieved NH3 profiles divided by their a priori).

The following figure shows examples of the retrieved scale factors at different layers for (top) daytime (10h to 14h BJT) data in the North China Plain in July 2022; and (bottom) nighttime (22h to 2h next day BJT) data in North India in December 2022. We can see that the retrieved scale factor profiles agree well between the two micro-windows.



* Figure 10: Can you add thermal contrast to this figure? (compare e.g. Fig. 4 of Clarisse et al., 2021). This would allow discussing better the variable sensitivity as a function of time.

You could also add the mean or median DOFS (unfiltered, total average).

In the revised manuscript, we have added Figure A2. It is an associated figure for Figure 10 on the diurnal change of TC and DOFS for the North China Plain and North India. Different from Figure 10, no extra filters have been applied.

- Section 5

* I would call this rather "Retrieval experiments".

Changed as suggested. In the revised manuscript, we have moved this to Section 3.

* Did you add synthetic noise? (ideally generated randomly from the noise covariance matrix)

Yes. As explained in the text, the assumed noise according to the spectra noise of FY-4B/GIIRS is added to the simulated spectra.

* I found section 5.2 not that interesting. Much rather it would be nice (see also comment above) to show and discuss the retrieved NH₃ profiles along side temperature profiles and averaging kernels. This could be done with averages, but also on individual observations, showing cases e.g. of extreme temperature inversion (which forces the profile to a narrow band), very large thermal contrast, etc.. The paper does not nearly go deep enough on this aspect.

The original Section 5.2 has been moved into the supplemental materials. Following your suggestion, we added Figure A1 to show examples of the a priori and retrieval NH₃ profiles and the corresponding AK row vectors for three cases: (a) positive TC with TC=13.37K and DOFS=0.89 from daytime measurement on July 07, 2022; (b) negative TC with TC=-6.7K and DOFS=0.91 from nighttime measurement on December 18, 2022; and (c) weak TC with TC=3.39K and DOFS=0.23 from early evening measurement on July 06, 2022.

The following statements have been added to Section 4.1:

“Measurement sensitivity of NH₃ is driven by TC and the NH₃ abundance (Clarisse et al., 2010). This is illustrated in the Appendix Figure A1(a) and (b) for a large positive and negative TC. As described above, while a positive TC leads to stronger absorption features, a negative TC causes spectral emission features allowing the detection of NH₃ also during the night (see also the example GIIRS spectra shown in Clarisse et al. (2021)). In both cases we see that the averaging kernels peak at the surface and the posteriori uncertainty in the retrievals of the surface layer are largely reduced compared to the a priori uncertainties. However, when the TC is small, as in the Appendix Figure A1(c), the DOFS values become smaller, and the AVK peaks higher up in the atmosphere (in this case, in the second layer). The retrieved value remains close to the a priori and the posteriori error is almost the same as the a priori, indicating low information content of the measurement. These examples illustrate the importance of TC for infrared sounding of boundary layer NH₃. An important advantage of GEO compared to LEO IR sounders is that they make observations throughout the day, such that optimal measurement conditions (large TC) can be found more readily. The diel variations of TC and DOFS are illustrated in the Appendix Figure A2 for North China Plain and North India. LEO IR sounders like IASI with an equator crossing times

at 9:30 am and 9:30 pm LT in general do not measure at the time where measurement sensitivity (or DOFS) is largest. The optimal time is found around noon.”

B. Minor comments

- Please detail exactly how thermal contrast was calculated (because "lower atmosphere") can mean a lot of different things. Also, is surface temperature used or the brightness temperature of the surface (the difference being the surface emissivity)

We have changed “lower atmosphere” to “the lowest atmospheric layer”. The surface skin temperature is the physical temperature (in Kelvin) of the surface skin.

- Figure 1c: x-label should be wavenumber, not frequency

Changed as suggested.

- Figure 2: y-axes labels inconsistent (diff vs difference)

Changed as suggested. In the revised manuscript, this figure has been moved to Figure S4.

- Figure 2: I would not show a priori x_1 or x_2 , it doesn't contribute or provide any new information. In this way, the second row can be removed altogether, since this difference is shown in the bottom panel. Also please refer to the appropriate section for the definition of a priori (which is not introduced yet at this stage of the paper).

Thank you for your suggestion. We have removed the results with a prior x_1 or x_2 . We added “**based on the a priori NH₃ profiles shown in Figure 3**” in the figure caption to link to the a priori profile.

- Line 159: How was the measurement error covariance matrix determined?

The following statement has been added to define the measurement error covariance matrix:
“ **S_ϵ is the measurement error covariance matrix, which is assumed to be a diagonal matrix constructed using the spectra noise estimates**”

- Line 170: this is technically incorrect. It all depends on the magnitude of S_a . If the retrieval is poorly constrained (very large S_a), then the DOFS will naturally be high, as all info comes from the measurement rather than the a priori. Conversely, if the retrieval is very (too) tightly constrained, the DOFS will always be small.

We have rephrased this statement to be:

“**For example, a DOFS of 1.0 means that, given the assumed S_a , at least one independent piece of information can be retrieved from the spectral measurement to constrain the vertical distribution of NH₃. Note that the DOFS is highly dependent on the magnitude of the assume S_a , an indicator of the a priori knowledge. If S_a characterizes a weaker constraint, indicating less a priori knowledge, the DOFS will be higher as relative more information will be taken from the measurement.**”

- Line 191: I guess this should read satellite zenith angles instead of solar zenith angles?

Changed as suggested.

- Line 234: this is factually incorrect, one can have a lower thermal contrast with increasing surface temperature. TC is a temperature difference, and for instance ($T_{surf}=290$, $T_{air}= 280$ K) has a higher thermal contrast than ($T_{surf} = 300$ K, $T_{air} = 299$ K). In fact the entire passage lines 233 - 237 need to be rewritten. I suggest the authors consult again the papers they reference just above, for proper terminology and physical explanations.

We have rephrased the statement to be:

“When thermal contrast is close to zero, measurement sensitivity is low, and DOFS are close to zero. Large positive TC increases sensitivity and results in NH₃ spectral signatures that are seen in absorption. Large negative TC also allows for sensitive measurements, this time allowing NH₃ spectral signatures to be seen in emission. Negative TC corresponds to the situation where the atmosphere is warmer than the surface, allowing to decorrelate the surface layer with the rest of the lower troposphere.”

- What is the spectral resolution of the emissivity atlas that was used. Please mention this in the manuscript.

We have added the following statement in Section 3.1:

“The emissivity values at 925 cm⁻¹ and 1075 cm⁻¹ are used to estimate the a priori emissivity for the retrieval micro-window. Two factors (slope and curvature) are used in the state vector to scale the wavelength dependent emissivity values”

C. English. Although in general not bad, there are several typos/grammar mistakes. I definitely did not try to be exhaustive, so here are just some that I noted:

We have changed the following. At the same time, our co-authors (including L. Clarisse and M. Van Damme) have carefully proofread the paper.

- Line 15 and 75: to imply > to quantify / study / analyse (?)

Changed to “to quantify”

- Line 310 availabel

Changed to “available”

- Line 77: remaining > remainder of (?)

Changed to “the remainder of”

- Line 83: inconsistency east Asia vs East Asia

Changed to “East Asia”

- Line 100: constraints (?) + remove "measuring" + remove "column"

Changed to “makes it possible to accurately retrieve NH₃ over East Asia”

- Line 113: "can be referred to" > can be found in

Changed to “can be found in”

- Line 134: the NH3 > NH3

Changed.

- Line 135: remove therefore

Changed.

- Line 233: "detectivity" please rephrase

Changed to "Previous studies by **Clarisse et al. (2010, 2021)** and **Bauduin et al. (2017)** using IASI observations have shown that the DOFS is primarily driven by the TC."

- Line 241: Please rephrase

Changed to "In NH₃ source regions (e.g., North China Plain and North India), the DOFSs are higher for the same TC compared with non-source region (e.g., Mongolia), suggesting the contribution from higher NH₃ concentration to the total information content."

- Line 244: This strong correlations > This strong correlation

Changed.

- Line 294: "are resulted" > result from

Changed.