General comments

The paper describes the application of global ammonia (NH3) emission inversion estimates over 2019-2022. As the current top-down emissions of NH3 are rather inconsistent across spatiotemporal scales, this approach provides a new insight into the NH3 emission budget, at relatively high resolution and daily scales. The inversion uses an IASI averaging kernel (AK) to constrain the profile of NH3 concentrations; results are compared with two global inventories and two top-down estimates. The average estimate shows a higher value, compared to previous budgets, either globally or regionally. The emission results are used to analyze the impact of COVID-19 lockdowns in 2020, as compared with that in 2019. However, the rise of emissions in 2020 seems to be likely due to the decrease in atmospheric NH3 sinks (e.g., NOx and SO2) and induces large uncertainty to these emission estimates.

We are grateful to the reviewer for the positive assessment of the manuscript, valuable comments and helpful suggestions which have helped to make significant improvements to the manuscript. We have addressed all the comments and accordingly revised the manuscript.

The inversion now uses IASI observations to constrain emissions, and other bottom-up inventories and top-down inversions for validation. Would it be possible to use simulated NH3 concentration based on updated emissions to compare with the IASI (or CrIS) concentration or in-situ observations? The current emission validation shows quite large differences between emission products, would the simulated concentration based on this emission estimate also give very different results with NH3 concentration observations? The consistency of this inversion method would be very necessary to check first.

We appreciate the reviewer's suggestion, which helped us strengthen the validation of our atmospheric inversion approach and emission estimates. As suggested by the reviewer, we have now conducted a LMDZ-INCA model simulation using the IASI-constrained NH_3 emission estimates derived from our global inversions for the year 2019 and compared the simulated NH_3 total columns with the IASI NH_3 total column observations. As detailed below, the agreement between the NH_3 total columns simulations and the IASI NH_3 observations across different spatiotemporal scales improved significantly when using the NH_3 emission estimates from inversions in the model simulations instead of the prior CEDS NH_3 emissions.

At the annual scale globally, the spatial Pearson correlation coefficient (r) between the yearly mean model-simulated NH_3 total columns and IASI observations improve from 0.71 (using prior emissions) to 0.90 (using IASI-constrained NH_3 emissions), while the root mean square errors (RMSE) decreases by ~29% from 0.52×10^{16} molec. cm^{-2} to 0.37×10^{16} molec. cm^{-2}. Similarly, at the monthly scale globally, the r value and RMSE between the model simulations with IASI-constrained NH_3 emissions and the IASI observations improve from 0.51 (using prior emissions) to 0.83 (using IASI-constrained NH_3 emissions), while the RMSE decreases by ~34% from 0.88×10^{16} molec. cm^{-2} to 0.58×10^{16} molec. cm^{-2}.

At the monthly scale and across major regions, including India, China, Africa, Europe, South America, and North America, the spatial correlation coefficients (r) and RMSE between the model simulations with estimated NH_3 emissions from inversions and the IASI NH_3 observations are respectively much higher and smaller than when the simulations are based on the prior CEDS NH_3 emissions ((Figure R1.1),. The spatial correlation coefficient (r) between the IASI-constrained NH_3 emissions' simulations of the NH_3 total columns and the IASI NH_3 observations exceeds ~0.8 in most of the regions at the monthly scale for this year 2019 of validation analysis (Figure R1.1). In one of the major NH_3 emitted regions, India, at the monthly scale, the spatial correlation increases from 0.40 to 0.86 and RMSE reduce by ~50% from 3.83×10^{16} molec. cm^{-2} to 1.91×10^{16} molec. cm^{-2} (Figure R1.1). Similarly, over another major NH_3 emission region, China, at the monthly scale, the spatial correlation increases from 0.40 to 0.79 and RMSE reduce by ~27% from 1.19×10^{16} molec. cm^{-2} to 0.87×10^{16} molec. cm-2 (Figure R1.1). It demonstrates the general

improvement brought at different spatiotemporal scales by the update of the NH_3 emission estimates from our inversions, and thus the internal consistency of our global inversion framework despite the rather simple linearization of the chemistry-transport underlying it. This improvement of the fit to the IASI NH_3 observations is a strong indication of the robustness of our inversion-based estimate of the global NH_3 emissions.

This analysis and a similar plot of model comparison with the IASI NH_3 observations are now presented and discussed in a new subsection 3.2 of the results section in the revised manuscript.
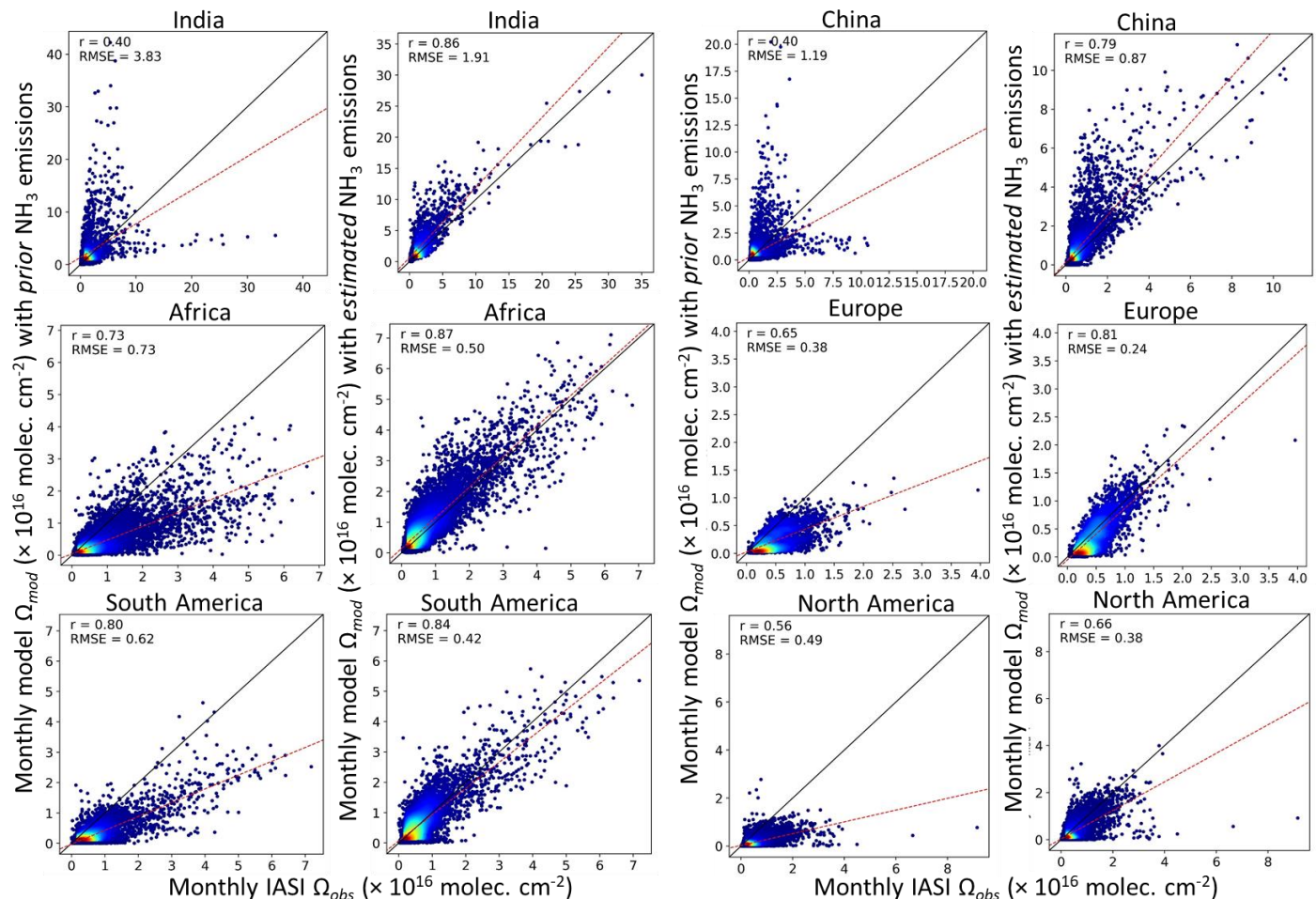

Figure R1.1: Comparison of the monthly averages of the IASI NH_3 total column observations (Ω_{obs}) to the corresponding averages of the simulation of these observations with LMDZ-INCA model (Ω_{mod}) over different regions for the year 2019. Each panel shows the correlation coefficient (r) and root mean square error (RMSE) between modeled (from both *prior* and IASI-constrained *estimated* NH_3 emissions from inversions) and observed IASI NH_3 columns. The left column in each panel displays results using prior CEDS NH_3 emissions, while the right column displays results using the *estimated* NH_3 emissions derived from our global inversions. The red dashed line represents the linear regression fit, and the black line denotes the 1:1 line.

Although the finite difference mass-balance (FDMB) inversion approach has been applied to update the anthropogenic NOx emission inventories, it has been rarely used in NH3 emission. Actually, the emission perturbation of 10-20 % is sometimes applied to get the scaling factor (beta). Could the authors further explain why a larger 40 % is applied in NH3 emission and distribution of the beta could be shown to clarify why it should be within the range of 0 to 10?

We agree with the reviewer's remark, and as already discussed in the Introduction section, the FDMB approach is mostly applied to update the anthropogenic NOx emission inventories, and, to our knowledge, the few studies which have investigated this approach for NH_3 emission inversion have applied it at regional scales. Momeni et al. (2023) and Li et al. (2019) applied this approach to derive estimates of NH_3 emissions

in the East Asia with CrIS observations and in the North America with IASI NH$_3$ observations, respectively. However, in this study, we investigate the application of the FDMB approach at the global scale to derive maps of the NH$_3$ emissions at a relatively high temporal resolution worldwide. Figure S1 in supporting information already shows an example of the distribution of monthly mean values of β for July 2019. Although, the values of β are generally less than 1.5 over most of the major NH$_3$ emitted regions worldwide, we applied a constrained on β to limit it within the range of 0 to 10 to avoid unrealistic emission adjustments resulting from any unrealistically large sensitivities.

To address the reviewer's concern about the impact on the inversion results of the selection of the level of perturbations, we have now conducted a sensitivity analysis with a LMDZ-INCA model simulation using a smaller 20% perturbation to the prior CEDS anthropogenic NH$_3$ emissions for the year 2019, in contrast to the original 40% perturbation used in our FDMB inversion setup. The results show that the differences in the resulting budget of the posterior NH$_3$ emissions over 2019 and the globe with the application of the FDMB based on these two levels of perturbations are less than 2%, indicating that the inversion results are not highly sensitive to the choice of perturbation magnitude within this range. The good fit between the posterior simulations using the inverted NH$_3$ emissions and the IASI NH$_3$ observations (see above) further strengthens the confidence in the linearization of the inversion problem based on 40% perturbations to the prior estimate of the emissions. This behavior is similar with that from previous applications of the FDMB method to the inversions of anthropogenic NOx emissions, where different perturbation levels (e.g., 5-50%) to the prior emissions resulted in minimal changes in the posterior anthropogenic NOx emission estimates at global and regional scales (Lamsal et al., 2011; Cooper et al., 2017; Zheng et al., 2020). The use of a 40% perturbation in our NH$_3$ study was motivated by the relatively high uncertainty in current NH$_3$ emission inventories, particularly over regions with strong agricultural sources. Nevertheless, our sensitivity test indicates that this choice (at least within a range of 20-40%) is not a critical parameter of our inversions.

This sensitivity test and this discussion are now included in subsection 4.3 in the revised manuscript.

The spatial resolution of the model is 1.27° × 2.5°, which challenges the assumption that there is no transport in a grid, considering the normal wind speed of more than 100 km in a day and the lifetime of NH3 around a day. And the prior inventory (in 0.5°) may not be able to capture the NH3 concentration dynamics at a finer scale, if not overridden by some regional inventories. Moreover, the NH3 is actively reacted with NH4+, so it is worth discussing whether the sensitivity of NH3 and NH4+ together would be better to capture the sensitivity of the NHx (NH3 + NH4+) family to the emission.

The typical lifetime of atmospheric NH$_3$ can reach about one day but generally ranges between few hours and one day, and our model's spatial resolution of 1.27° × 2.5° exceeds the 100 km scale. However, we agree that the model typical length scale can often be reached by the advection of NH$_3$ within its lifetime. This transport to neighboring grids can lead to a spatial "smearing" effect, where emissions are dispersed away from their source grid cell, introducing errors in mass balance inversion approaches (Cooper et al., 2017). This problem of spatial smearing in mass balance inversion approaches is well-documented for short-lived species like NOx (Cooper et al., 2017; Palmer et al., 2003). Such smearing can lead, on average, to the under-estimation of the regional scale emissions, since the approach overlook the fact that the amplitude of the NH$_3$ signal associated to a given area source decreases with the advection downwind (Cooper et al., 2017). For other short-lived species like NOx, some approaches such as smoothing kernels or iterative FDMB inversion approaches have been used to reduce these errors, but the latter is computationally intensive, especially for global inversions. Similar approaches could be explored to NH$_3$ inversions in future work if computational resources allow. We acknowledge this limitation for NH$_3$ inversions in our study and have highlighted it in section 4.3 in the revised manuscript.

In our model setup and inversion framework, the CEDS inventory is re-gridded to match the model resolution. While this inevitably misses some fine-scale features, our study focuses on the broader regional patterns of NH$_3$ emissions rather than point-source inversions. We agree that inversions at higher resolution, based on high-resolution regional inventories (e.g., MEIC, NEI, CAMS-REG, etc.) and high-resolution chemistry transport model simulations can bring more robust information of the more localized sources such

as point sources at sub-national scales. But the above-mentioned limitation of the FDMB approach (somehow ignoring the advection across the chemistry transport model grid cells) would be exacerbated at such a higher resolution. Even using iterative FDMB approach to overcome this spatial smearing effect at finer resolutions, errors in the derived emission estimates can be amplified (Li et al., 2019). Therefore, application of such an inversion approach at the finer resolution may have limitations to accurately estimate the NH3 emissions. It is now discussed in section 4.3 of the revised manuscript.

We agree with the reviewer that an inverse modelling framework including observations of the full reduced nitrogen family (NHx = NH$_3$ + NH4+) and relying on tests of sensitivities of NH$_3$ and NH4+ to changes in NH$_3$ emissions could provide a more comprehensive constraint on NH$_3$ emissions, given the rapid gas-particle partitioning of NH$_3$ to NH4+ under typical atmospheric conditions. However, current satellite retrievals such as those from IASI and CrIS are primarily focused on gaseous NH$_3$. To our knowledge, the current spaceborne instruments have a limited capability to detect particulate-phase NH4+. As a result, the observational constraints in our inversion framework are based only on NH$_3$ columns. Nevertheless, the LMDZ-INCA aerosols-chemistry transport model used in our inversion framework fully represents these chemical conversions of NH$_3$ to NH4+ and the partitioning and deposition processes affecting the entire NHx family. Therefore, the LMDZ-INCA model and, implicitly, our inversion framework account for the fate of NH3 through its interaction with NH4+ when deriving relationships between the NH$_3$ emissions and concentrations. We have added a discussion on this point in section 4.3 of the revised manuscript.

The longer period has been used for spin-up (2010-2018), by using the CEDS global bottom-up gridded inventories as a prior. However, post-2019 was set with the carbon emission growth rate, which I think is inappropriate for the NH3 since 1) NH3 does not have an intense relationship with fossil fuel emissions, as an agricultural-based emission, and 2) they have different trends in anthropogenic sources, but may have a similar reflection on the biomass burning. Instead, the post-2019 prior could be set as invariant and adjust the simulated NH3 columns with the IASI observations, and the derived NH3 emission could be corrected by SO2/NOx change during the COVID lockdown. Or authors could just update it after the release of new CEDS emissions.

As already discussed in the manuscript and as mentioned by the reviewer in this comment, the NH$_3$ emissions are mainly linked to agriculture (e.g., fertilizer use) and do not have a strong relationship with fossil fuel combustions. When extrapolating the timeseries of the CEDS emissions after 2019, the corresponding emission growth rate are derived from the Carbon Monitor dataset by source sector, by month, and by country. These growth rates to the CEDS estimates for 2019 calculations are applied separately for each source sector. However, the Carbon Monitor dataset does not report CO$_2$ emissions from the agricultural sector, and thus, any growth rate does not apply for this sector. We agree that it is the dominant sector. As a result, the NH$_3$ emissions after 2019 remain almost invariant. However, SO$_2$ and NOx are co-emitted with CO$_2$ by the fossil fuel combustion. There are thus large variations in the emissions of SO2 and NOx after 2019, especially, during the COVID-19 period due to reductions in activities associated to fossil fuel combustion. Since the LMDZ-INCA model includes a full chemistry scheme, the changes in NOx and SO$_2$ emissions affect secondary inorganic aerosol formation (e.g., nitrate and sulfate), which in turn can alter NH$_3$ partitioning and atmospheric lifetime through formation of ammonium (NH$_4^+$). Therefore, even though NH$_3$ emissions remained largely unchanged in our extrapolated CEDS-Carbon Monitor prior, the resulting NH$_3$ concentrations were still affected by changes in co-emitted species emissions in the LMDZ-INCA model simulations due to atmospheric chemical interactions. Therefore, the variation in the NOx/SO2 emissions is accounted for in the model simulations from the extrapolated prior CEDS emissions.

We should also remind that in our NH$_3$ inversion framework, which is not Bayesian, the role of the prior estimate of the NH$_3$ (at the monthly scale) is to define the state around which the chemistry transport model is linearized, and as discussed above, the linearization seems to be robust over a wide range of level of perturbations to this prior estimate. Based on this linearization, the inversion aims to perfectly fit the observation, without giving weight to the prior estimate of the NH$_3$ emissions at the monthly scale, which

lessens the impact of the temporal variations of this prior estimates. However, the NOx and SO$_2$ emissions are kept fixed in the inversion process, so that it is more critical to derive suitable temporal variations for these emissions.

Therefore, in the current inversion also, the prior NH$_3$ emissions were also remained invariant throughout the period of this study and they were adjusted based on observed NH$_3$ columns from IASI with varying emissions of SO$_2$ and NOx in LMDZ-INCA model simulations. We have further clarified this in the revised manuscript and as already discussed in the manuscript, we plan to explore multi-species joint inversion of NH3, NOx, and SO2 in future.

Although the paper focuses on the application of the inversion system in a high spatiotemporal resolution, the setup and suitability of the system are not sufficient enough to publish, before more tests and discussions on its sensitivity and consistency. For some parts a more detailed and cleared description could be useful, as described below in the Specific Comments. Overall, the paper is easy to read with a good structure, but could still not be published in the ACP in terms of the above scientific concerns.

We thank the reviewers for his/her thoughtful comments and acknowledge that there was a need to further assess the sensitivity and consistency of the inversion setup and the robustness of the estimated NH$_3$ emissions based on new model simulations. As discussed in answer to the previous comments, we have revised the manuscript, including additional LMDZ-INCA simulations, using the posterior estimates of the NH$_3$ emissions, and with a different level of perturbation of the NH$_3$ prior emissions, new comparisons to the IASI NH$_3$ column observations, and clearer explanations and discussions regarding the points raised by the reviewer. We hope these revisions address the reviewer's concerns and demonstrate the robustness and suitability of our approach for the global atmospheric NH$_3$ inversions using satellite observations and chemistry transport model.

Specific comments

line 20 'all the spatiotemporal scales': provide concrete ranges of spatial scales (e.g., regional to global, 0.1° to 2° resolution) and temporal scales (e.g., daily, seasonal, interannual variations from 2010 to 2018)

We aimed at making a very general statement here. We prefer to decrease its scope rather than detail it, especially since, due to strict words limit in the abstract, this detailed information is difficult to provide here.

line 25 'prior CEDS inventory's anthropogenic NH3 emissions': if only update the global anthropogenic NH3 emissions, consider modifying the title correspondingly to accurately reflect the focus

Modified.

line 31-32 Post-2019 emission trends: this conclusion highlights a limitation in the post-2019 prior emission, particularly for NOx and SO2, which may propagate unrealistic trends in NH3. I recommend explicitly addressing their impact to correct this unrealistic NH3 emission trend.

See our answer to the general comment on this above. Furthermore, again, due to strict words limit in the abstract, it is not possible to discuss such a topic here. However, it is addressed in discussions and conclusions sections.

line 74: NH3 emission estimates or NH3 concentrations? The majority of the paragraph is talking about NH3 observations, but latter you also mention the NOx emissions, a bit unclear.

We agree that the sentence could be clearer. The intention was to highlight that most NH$_3$ emission estimates derived from the satellite data are based on NH$_3$ observations from instruments mainly IASI and CrIS. This is clarified further in the revised manuscript.

line 212: which kind of pre-/post-retrieval filters you applied, except for the cloud coverage?

IASI-ANNI-NH3-v4 data product accompanied these pre- and post-retrieval flags and we follow the recommendations of the data user guide. The pre-filter removes measurements with erroneous L1 or excess

cloud coverage. The post-filter flags retrievals with limited or no sensitivity to the measured quantity (Clarisse et al., 2024).

line 267: any cases for NH3?

Yes, we have now included Cao et al. (2022) and Ding et al. (2024) here which used CriS NH_3 observations with averaging kernels for ammonia emission estimates.

line 383-386: As shown in Figure 2, IASI NH3 columns are much higher than the model simulation. Assuming you still keep those negative values in IASI retrievals, does this bias arise from underestimated agricultural emissions in the prior inventory or systematic biases in IASI retrievals?

Previous validation studies of earlier IASI ANNI NH_3 retrieval products (e.g., with version 3) showed relatively good agreement with in situ and FTIR measurements (Guo et al., 2021; Wang et al., 2020). Although, the IASI ANNI NH_3 v4 product introduces important improvements compared to the earlier versions and expects minimal biases, a comprehensive validation of this version has not yet been conducted and such a validation is anticipated in upcoming studies (Clarisse et al., 2024). Therefore, the bias between IASI NH_3 columns and LMDZ-INCA model simulations mainly reflect an underestimation of agricultural NH_3 emissions in the prior inventory, as well as a misrepresentation of their seasonal variation, but we cannot fully rule out remaining retrieval uncertainties in the absence of comprehensive validation of this version of IASI NH_3 retrievals. We have now clarified this point in section 3.1 of the revised manuscript.

Technical corrections

Acronym consistency: consistently use the acronym 'AK' for 'averaging kernels' from top to bottom, the same applies to others

Updated accordingly.

line 97-99: add some references to support

Added.

line 111 'EDGAR': full name

Added.

line 239 and 276 'interpolated onto the model horizontal grid': which interpolation method?

We use conservative regridding by ensuring that the total mass (e.g., emissions) is preserved during the interpolation. This information is now provided in section 2.2 of the revised manuscript.

Figure 1 clarification: what is the difference between 'LMDZ-INCA original' (orange) and 'LMDZ-INCA without AK' (red). Besides, the AK is higher with the lower pressure (higher elevation), but why the largest discrepancy happens at around 600-800 hPa

Here "LMDZ-INCA original" refers to the modelled NH_3 mole fraction vertical profile on the LMDZ-INCA model's native 79 vertical levels and "LMDZ-INCA without AK" represents the NH_3 sub-columns calculated after interpolating this profile onto the 14 pressure levels used by the IASI ANNI-NH3-v4 retrieval, but without applying the averaging kernel (AK). In contrast, "LMDZ-INCA with AK" includes the effect of the AK, making it more comparable to the satellite retrieval.

Regarding the behavior observed around 600-800 hPa despite almost smoothly varying AK values with increasing altitudes above the surface: this is mainly due to the interaction between the vertical structure of the modelled NH_3 profile and the thickness (or pressure width) of the sub-columns. The NH_3 sub-column represents the mass of NH_3 in each pressure layer, so layers with both significant NH_3 concentrations and wider pressure intervals can result in larger NH_3 sub-column values even if the AK is not at its peak there. Consequently, even modest AK values at higher altitudes, combined with substantial NH_3 mass in thick pressure layers, can lead to amplified contributions to the total column.

It is clarified better in section 2.3 of the revised manuscript.

line 312 'given hourly': 9-10 AM?

Yes, IASI has a morning overpass around 09:30 local solar time (LST). Since the LMDZ-INCA model outputs are in UTC and each IASI pixel also includes a UTC timestamp corresponding to the local overpass time, we match each IASI observation with the modelled NH3 profile at the corresponding nearest UTC hour.

319-320: what is the definition of the 'high-quality IASI pixels' it looks rational if you exclude negative columns as long as the negative IASI NH3 total column has been kept after the filtering process.

Here, "High-quality IASI pixels" means the dataset obtained after applying recommended quality flags accompanied with the data product. As recommended in the data product documentation and its publications, we don't exclude negative NH_3 columns at pixel levels during the initial averaging of IASI NH_3 columns onto the model grids. However, for subsequent steps such as for inversions, we use only the non-negative gridded values at the model grid levels, in order to ensure physical consistency in the flux estimates and analysis.

line 453-465: the description of the gap-filling method is important but consider moving it into the 'Material and methods' part

Moved to "Material and Methods" section.

line 658-660: it is inequivalent to compare your anthropogenic emissions with Luo 2022 and Dammers 2022, since they also included the natural sources (e.g., biomass burning). But your emission estimates (98 Tg yr-1) are still higher than Luo's (78 Tg yr-1), which is quite interesting and worth discussing by comparing with your spin-up stage (prior to 2019). I would like to see such a comparison in a Table or Figure.

The biomass burning contribution to the total NH_3 emissions is much smaller than the anthropogenic one (at least, according to our prior emission products). Therefore, this comparison doesn't impact much our analysis. It is a spin-up stage without inversion so, we think, this comparison could be misleading and we prefer to avoid it.

line 704: for India comparison, there is a new study you could check:
https://egusphere.copernicus.org/preprints/2025/egusphere-2024-3938/#discussion

Thanks for this reference. We cited this in the revised version.

line 785: is it possible to quantify the uncertainty for your emission estimates, via satellite retrieval errors/number and model transport biases?

As mentioned briefly in the "Uncertainties and Limitations" section, the current framework of our atmospheric inversion does not provide uncertainties in our emission estimates. However, there are a few studies (Cooper et al., 2017; Koukouli et al., 2018) which tried to get some information about the uncertainties in their estimates using basic or FDMB inversion approach, propagating the observation errors. While implementing a similar approach could be considered in future work, it is beyond the scope of the current study. We have discussed it further in section 4.3 of the revised manuscript.