

We sincerely thank all the referees for reviewing the manuscript and their thoughtful comments and constructive suggestions which have contributed to strengthening the quality of the paper. Responses to referee's comments (black text) are given below, in blue text.

## Author's response to Anonymous Referee #2

### General comments

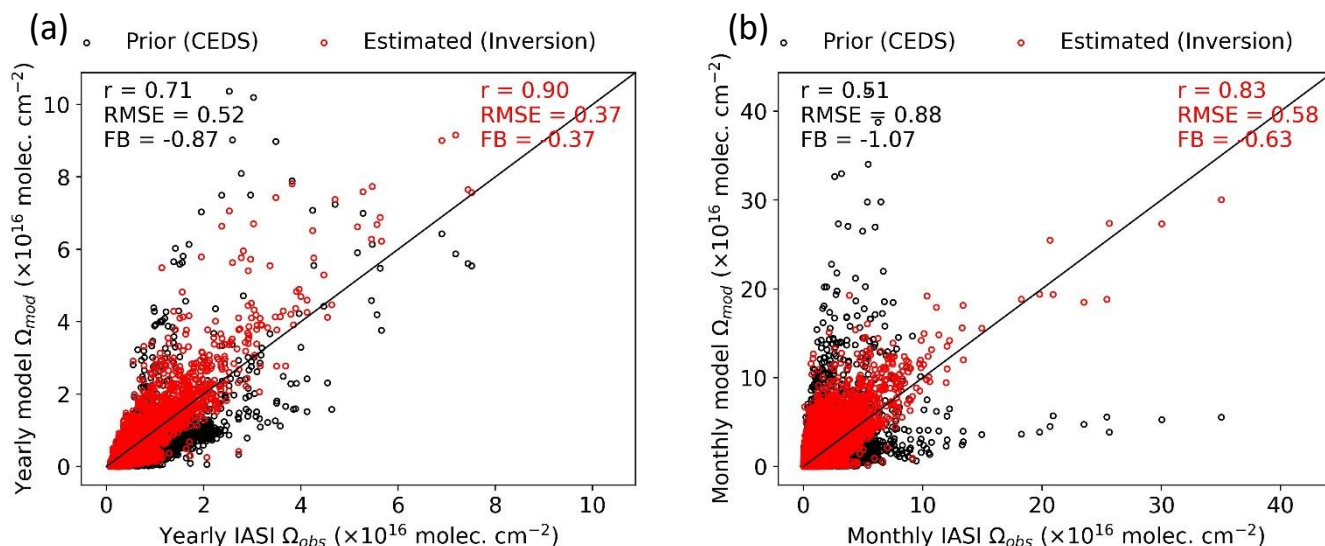
The authors have comprehensively addressed most of my previous comments through a new evaluation section and enhanced discussion of uncertainties. To further strengthen the manuscript, the following minor revisions are required:

We sincerely thank the reviewer for the positive assessment of our revised manuscript and of the improvements made based on his/her comments on the previous version of the manuscript, particularly in the evaluation section and the discussion of uncertainties. We appreciate the reviewer's thoughtful comments and constructive suggestions, which have contributed to further strengthening the quality of the manuscript. We have carefully addressed all the additional comments and suggestions and have revised the manuscript accordingly.

1. The new section 3.2 (Evaluation of estimated emission):

1. Global annual/monthly evaluation results (model vs. IASI) should be presented in a dedicated figure or table (main text or appendix).

Figure 4 in the previous revised version of the manuscript, showed the model vs. IASI comparisons over specific regions for the evaluation of the regional analysis. We have now included the following figure with model vs. IASI comparisons over the globe for the evaluation of the results at global scale in the Supporting Information (Figure S8). This figure was already discussed in the previous revised version of the manuscript.



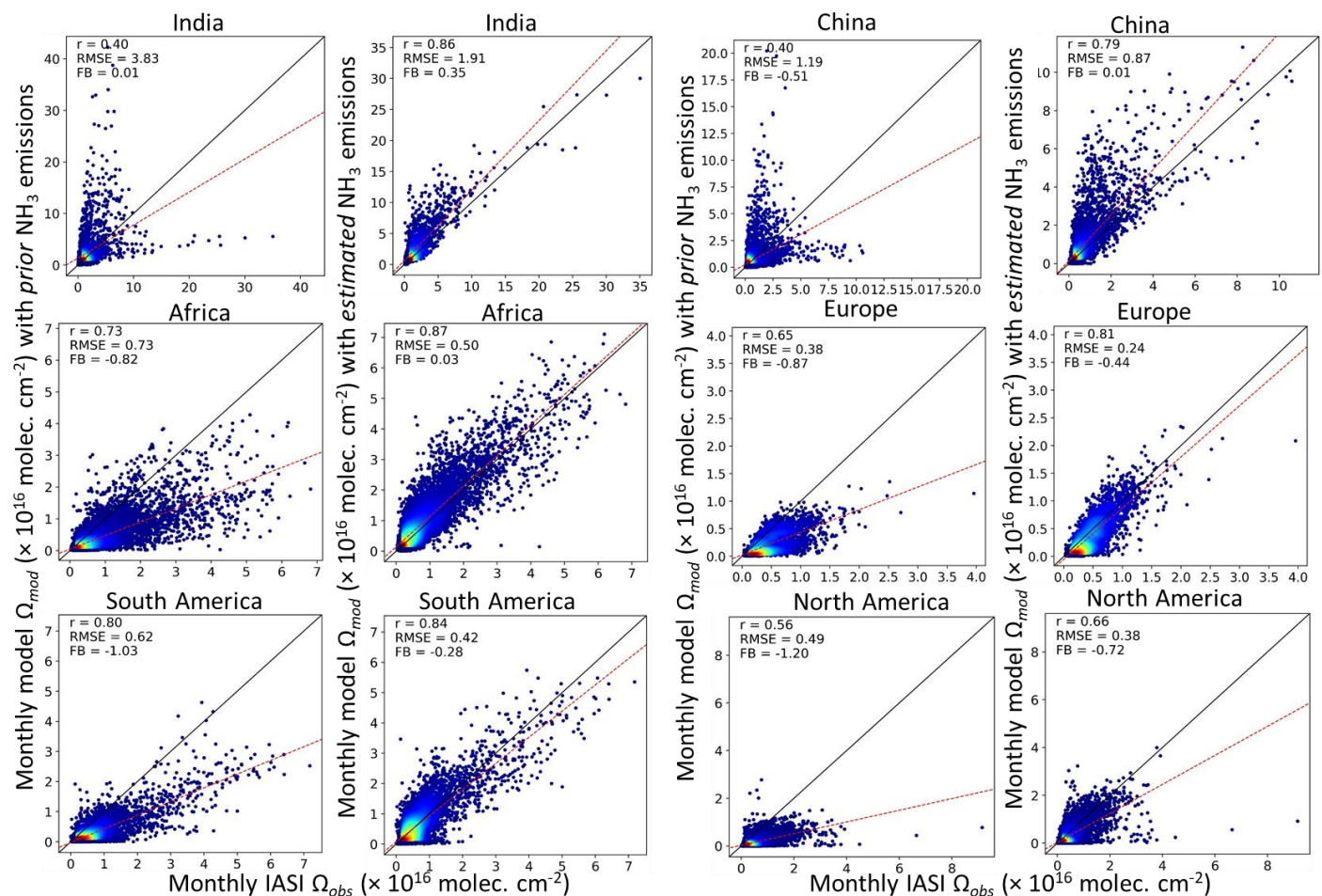
**Figure R2.1:** Comparison of the (a) annually, and (b) monthly averages of the IASI NH3 column observations ( $\Omega_{obs}$ ) over the model horizontal land grid cells at  $1.27^{\circ} \times 2.5^{\circ}$  resolution to the corresponding simulations of average columns with LMDZ-INCA ( $\Omega_{mod}$ ) using the IASI-constrained NH3 emission estimates derived from our global inversions and using the prior CEDS NH3 emissions over the globe and the year 2019. It also shows the correlation coefficient ( $r$ ) and root mean square error (RMSE), and Fractional Bias (FB) from this comparison. The black line denotes the one-to-one line.

2. Clarify how consistency improves at the daily scale (e.g., metrics for 10-day/daily performance), given the inversion's 10-day resolution.

As noted in our response to reviewer comments on previous version of the manuscript, the current LMDZ-INCA model framework only reads as input  $\text{NH}_3$  emission products with a 1-month temporal resolution. Internally, the model uniformly distributes these monthly emissions across hours, without incorporating diurnal variability. Therefore, in our evaluation protocol, the LMDZ-INCA simulation using the emission estimates from the inversions does not read these estimates at 1-day resolution but monthly averages of these estimates. This is now clarified in the text. We assume that this does not limit our capability to evaluate the inversion estimates based on comparisons to monthly and annually averages of the observations. However, although model performance at the daily scale also improves notably at both regional and global scales, comparisons to daily averages of the observations would probably be misleading, as the current framework of model does not read the day-to-day emission variability as input. It would demonstrate that the improvement of the emission estimate at the monthly scale can support the improvement of the simulations at 10-day scale, but it would not support the assessment of the inversion estimate at the 10-day scale (1-day resolution). Therefore, we restrain ourselves from comparison of the LMDZ-INCA simulation using the emission estimates from the inversions to daily or 10-daily averages of the observations.

3. The regional comparisons show large improvements in RMSE (closer to 1:1 line), and I also noticed the enhancement by mean values. Supplement regional scatterplots with fractional bias metrics to quantitatively demonstrate bias reduction beyond RMSE improvements.

We have included the fractional biases (FB) for each regions in the figure that was already provided in responses to reviewer's comments on previous version of the manuscript (shown below) which is now included in the supporting information (Figure S9). The FB metric quantitatively confirms bias reduction in modelled  $\text{NH}_3$  columns when using IASI-constrained  $\text{NH}_3$  emissions in the model compared to the prior CEDS  $\text{NH}_3$  emissions. Specifically, we observe a clear decrease in FB across all regions, except for India. In the case of India, although the FB remains slightly elevated, the RMSE shows substantial improvement, indicating better representation of the magnitude and spatial and temporal variability, even if the mean offset is not fully corrected. This is now briefly discussed in the revised manuscript.



**Figure R1.2:** Comparison of the monthly averages of the IASI NH<sub>3</sub> total column observations ( $\Omega_{obs}$ ) over the model horizontal land grid cells at 1.27°×2.5° resolution to the corresponding averages of the simulation of these observations with LMDZ-INCA model ( $\Omega_{mod}$ ) over different regions for the year 2019. Each panel shows the correlation coefficient (r), root mean square error (RMSE), and fractional bias (FB) between modeled (from both *prior* and IASI-constrained *estimated* NH<sub>3</sub> emissions from inversions) and observed IASI NH<sub>3</sub> columns. The left column in each panel displays results using prior CEDS NH<sub>3</sub> emissions, while the right column displays results using the *estimated* NH<sub>3</sub> emissions derived from our global inversions. The red dashed line represents the linear regression fit, and the black line denotes the 1:1 line.

## 2. Revised section 4.3 (Sensitivity test and uncertainties):

1. I appreciate the sensitivity analysis authors conducted with a smaller perturbation used (20% vs. 40%) and see the minor impact (2 %) on the emission estimates. Explicitly state whether the 2% emission difference stems from changes in the scaling factor ( $\beta$ ) due to reduced prior perturbations (20% vs. 40%).

$\beta$  is the only variable which changes when changing the level of perturbation. Therefore, the less than 2% change in the emission estimates necessarily stems from the changes in  $\beta$  (due to the reduced prior perturbations). We have explicitly stated this in the revised version.

2. line 872: double check the number of perturbation levels as I only see 5-40 % in three cited references.

In Zheng et al. (2020) they conducted these sensitivity tests by up to 50% perturbation levels, so this range is correct.

3. I agree the spearing effect or spearing length scale statement discussed now, as it is important for the resolution setup in the inversion system. For the full reduced nitrogen family (NH<sub>x</sub>), it is true that satellite such as IASI and CrIS do not detect NH<sub>4</sub><sup>+</sup>. Probably could include NH<sub>4</sub><sup>+</sup> in the calculation of the scaling factor ( $\beta$ ) to better represent NH<sub>x</sub> sensitivity, provided LMDZ-INCA outputs NH<sub>4</sub><sup>+</sup> concentrations. This would enhance the analysis of NH<sub>x</sub> transport.

We thank the reviewer for this suggestion to improve the inversion system by including NH<sub>4</sub><sup>+</sup> in the calculation of the scaling factor ( $\beta$ ) to better represent NH<sub>x</sub> sensitivity. Our model, LMDZ-INCA, does provide NH<sub>4</sub><sup>+</sup> concentrations, which can enhance the analysis of NH<sub>x</sub> transport, particularly given the important role of NH<sub>4</sub><sup>+</sup> in atmospheric chemistry and deposition. However, as noted in our response to reviewer's comment on previous version of the manuscript, implementing such a refinement in the inversion would ideally require corresponding observational constraints for NH<sub>4</sub><sup>+</sup>. In the absence of direct satellite observations of NH<sub>4</sub><sup>+</sup> (unlike NH<sub>3</sub>, which is detected by IASI or CrIS), it remains challenging to evaluate or adjust the NH<sub>4</sub><sup>+</sup> component within the inversion framework. Nevertheless, this is an important consideration for future developments, especially if reliable observational proxies or retrievals of NH<sub>4</sub><sup>+</sup> become available.

3. Post-2019 Emission: Add a supplementary figure comparing prior annual anthropogenic emissions of NH<sub>3</sub>, NO<sub>x</sub>, and SO<sub>2</sub> (2019-2022) to contextualize post-2019 NH<sub>3</sub> variability. I look forward to future work on joint inversions of these species.

We have added a figure (Figure S1) of these species' emissions in supplementary material.

4. line 492: Clarify "misrepresentation of seasonal variation": Do both IASI and LMDZ-INCA fail to capture observed seasonality? This is particularly relevant given IASI's higher NH<sub>3</sub> columns during non-growing seasons in agricultural regions (e.g., EU/NA).

We have clarified the sentence further to specify that the "misrepresentation of seasonal variation" refers specifically to the prior CEDS NH<sub>3</sub> emissions, which lack accurate seasonality, particularly in the major agricultural regions. This limitation in the prior NH<sub>3</sub> emissions propagates into the modeled NH<sub>3</sub> columns simulated from the LMDZ-INCA.

As already mentioned in this paragraph, we also acknowledge that the elevated NH<sub>3</sub> retrievals observed by IASI during non-growing seasons in some regions such as Europe and North America could also be partly influenced by retrieval uncertainties, such as surface temperature effects or low thermal contrast, especially in winter months, which can affect satellite sensitivity.

5. line 603-604: how much is the emission contribution from biomass burning compared to anthropogenic emissions in these region?

We have computed the contribution of biomass burning (BBG) emissions relative to anthropogenic (ANT) emissions ( $BBG / ANT \times 100$ ) in the specified regions. The BBG-to-ANT ratios are as follows:

**South America:** BBG emissions contribute ~11% in 2021 to maximum ~15% in 2020 relative to anthropogenic emissions.

**Africa:** The contribution ranges from ~15% in 2022 to ~17% in 2021.

**North America:** BBG emissions account for around ~5% of anthropogenic emissions in most years, with a notable increase to ~26% in 2021, likely due to elevated fire activity that year.

These information are included in the revised manuscript.

## **Author's response to Anonymous Referee #3**

### **General comments**

I realize that this is a revised manuscript, but I have not reviewed any earlier version of this manuscript. In this regard, I have gone through the revised-track-changed version of the manuscript along with author's response to the Reviewer #1 and #2 comments. In this study, the authors investigate global ammonia emissions from 2019-2022 by using satellite observations from IASI and a chemistry-transport model called LMDZ-INCA. The study updates ammonia emissions using the finite difference mass-balance through an atmospheric inversion technique. Furthermore, the paper examines regional variations in ammonia emissions and their seasonality, addressing discrepancies with current inventories and influences from COVID-19. I found the objective of this paper very relevant for publication in ACP. The paper is mostly well written but needs some very minor language editing work (could be taken care of by copy-editor) for a better readability to the general reader (though I have not pointed out those changes). My observations are that both the Reviewers found the paper interesting and well written; and I also have the same opinion regarding the aim, objectives, methodology and conclusion of the research work presented in the manuscript. I spend some time to go through the Reviewer #1 critiques and corresponding responses by the authors. To me, authors have done a commendable job in addressing the major issues raised by the Reviewer #1. The responses look well augmented and easy to follow by a general reader who is not specialized domain. Overall, I am satisfied with the scientific idea and analysis presented in the paper and authors' responses and recommend acceptance of the paper in the current form.

We are very grateful to the reviewer for the thorough reading of our revised manuscript and for the positive evaluation of the scientific objectives, methodology, and conclusions of our work. We are especially grateful for the recognition of the efforts made in addressing the previous reviewers' comments and for the acknowledgment of the clarity and relevance of our revised version of the manuscript.