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Reply on RC2

Michael Sitwell et al.

Author comment on "An ensemble-variational inversion system for the estimation of ammonia emissions using CrIS satellite ammonia retrievals" by Michael Sitwell et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-549-AC2, 2022

Thank you for your comments. Reviewer comments are in italics.

1) *I am not sure why the author spent much effort about the difference between log-space and linear-space H(observational operator) to justify the 'hybrid' technique. Is that because of the scientific importance? If that is the efficient approach, then 3.3 should be shortened and briefly explain the benefit of the compromised approach. (Those descriptions and testing results are too technical to this journal)*

The hybrid technique is a novel comparison method that has a significant impact on the inversions, as shown in the 'Results' section. The discussion around this technique is to emphasize that the comparison method chosen can dramatically change the results of the inversions. As this is a new technique, we would like the details of this method documented, but have moved more of the details to a new appendix (Appendix B) to streamline the main text. Additionally, the plot showing the operator selection using the hybrid method (what was labeled as Fig. 3) has been moved to the Supplement.

In the interactive discussion, Anonymous Referee #2 requested additional details of the hybrid technique (under the comment 'Sensitivity of constants' in the comments RC1) and in response we actually added details of the hybrid method to a subsequent version of the manuscript. Moving more of the details to a new appendix and Fig.3 to the Supplement is our attempt at trying to balance these comments with the comments from Anonymous Referee #2.

2) *I understand why the column comparisons with the averaging kernels for this work. But if the operator has higher sensitivities with the vertical profile, is that any possibility to compare the satellite data and model at a specific level only with the highest sensitivity (such as 700hPa or near-surface levels only)?*

I would image that this approach possibly could yield fairly similar results to using the full profile (assuming the a priori was still accounted for). While many profiles are sharply peaked at a particular level, not all profiles have such a narrow peak. Also, different profiles will peak at different levels. So since we had access to the full profile, we used the information from the full profile.

3) *The author has to comment more about the reason for GEM-MACH performance before and after the inversion since readers do not know much about the potential weakness or*

biases of the generic model performance. We don't determine the meaning of changes by this work well.

To help address this comment, in addition to adding more discussion on this point, we thought it would be beneficial to rearrange the 'Results' section to better highlight these differences. Previously, the 'Results' section was subdivided into subsections by result type (i.e. a subsection looking at the inversion result, another subsection looking at the surface NH3, etc…). In the revised manuscript, the 'Results' section instead starts with a subsection describing the 'before' case (Subsection 4.1), followed by a subsection describing the inversion (Subsection 4.2), then by a subsection describing the 'after' case (Subsection 4.3). Additional comments on the results from specific ground stations, biases, and emissions sources were also added (see 'Tracked Changes' version of the manuscript).

4) *The ammonia has a relatively short lifetime and the author claimed that the ammonia concentrations have increased. How is the degree of underestimation of NH3 emissions and the trends over the other continents? The comparison of this work to other regions(or studies) will be informative as well.*

Currently there are limited ammonia inversion studies outside of North America.

One study that examines ammonia inversions over Europe is currently under review for 'Journal of Geophysical Research: Atmospheres' (https://doi.org/10.1002/essoar.10507960.1). While the authors of this paper are coauthors on the JGR manuscript as well, the JGR manuscript uses a different model (GEOS-Chem) and inversion method (4D-Var). In the case of the unidirectional flux scheme, the inversion increases emissions in most of Europe in the Spring and Summer (with areas like Northern Italy as an exception).On the other hand, emissions are decreased in many places in Europe during the fall and winter. However, the annual emissions are increased by the inversions in most of Europe.

Another relevant manuscript that is currently under review in ACP and available in preprint is 'Data assimilation of CrIS-NH3 satellite observations for improving spatiotemporal NH3 distributions in LOTOS-EUROS' (https://doi.org/10.5194/acp-2021-473). This study uses CrIS NH3 retrievals to estimate NH3 emissions over Germany and parts of Belgium, and the Netherlands. Assimilation done with a LETKF increased emissions throughout this region, with increases as much as 30% for the total emissions over 2014-2018.