Point-by-Point Responses to Anonymous Referee #2

We thank Anonymous Referee #2 for their time and effort in reviewing our work and appreciate their recommendations for ways to improve the manuscript. Following many of their suggestions, we believe the revised manuscript is an improvement upon the original submission. The referee comments below are taken from https://doi.org/10.5194/egusphere-2023-2227-RC2.

This document provides a point-by-point response to Referee #2. The responses follow the sequence: (1) comments from the referee, (2) authors' response, (3) authors' changes in manuscript.

1.1 Referee Comment

"This paper presents a Bayesian inversion estimating ethane and propane emissions using aircraft data from the ATom campaigns and GEOS-Chem predictions at different horizontal resolutions (4x5, 2x2.5, and 0.5x0.625 degrees nested). The authors argue that the use of potential temperature as a zonal coordinate results in a comparison to aircraft data that is less sensitive to model resolution than comparisons on lat-lon-pressure coordinates, and inversions for longer-lived trace gas species can thus be performed at coarser resolution to save time and cost."

1.2 Author Response

We no longer use the Bayesian approach in this study. We substantially revised the manuscript to focus on an analysis approach by creating pseudo data for ethane and propane as sampled by the DC8 aircraft during the Atmospheric Tomography (ATom) aircraft mission. We sample simulations from the highest resolution (0.5 x 0.625 degree) GEOS-Chem High Performance v14.3.1 and then evaluate the correlation of these pseudo data with lower resolution simulations and simulations sampled +/- 5 days from the pseudo data. We apply a simple statistical analysis that illustrates the value of potential temperature as a vertical coordinate in comparing sparse observations with a GCM.

1.3 Manuscript Changes

Figures 4-8.

2.1 Referee Comment

"While the results of this study certainly have potential value for other work quantifying sources of longer-lived trace gases, this paper is not suitable for publication without major revisions. As is, it assumes the reader is intimately familiar with the previous study on which it is based

(Tribby et al., 2022), and reads as a sort of proof-of-concept of that specific work (a Bayesian inversion based on 4x5 degree model predictions and comparisons to ATom data on potential temperature coordinates). More detail is needed throughout the paper in order for this to serve as a standalone study. A discussion and conclusions section are also needed after the Results section to demonstrate the broader context of this work and its applicability to other research problems (and to complete the paper!). Also, many of the claims made throughout the paper are currently too qualitative, and the authors must do more to quantify their findings and demonstrate how they improve upon inversions using lat-lon-pressure coordinates. I also question whether the analysis used is the best approach for testing their hypothesis. I have made specific line-by-line suggestions below."

2.2 Author Response

We have made major revisions to address these concerns. First, we have provided a new analysis approach to test this hypothesis and no longer use the Bayesian approach, we have added a discussion and conclusion section, and more quantitative detail with RMS, RMSE, and simple linear regression analysis.

2.3 Manuscript Changes

Figure 4-8.

3.1 Referee Comment

"Lines 46-51: More information about the data exclusion process here is needed. Presumably these are being done to limit the dataset to those observations whose variance is dominated by synoptic-scale transport from the midlatitudes? How do the authors confirm their screening can "sufficiently reduce subtropical influence"? How do the results vary if these screening criteria are changed, and what recommendations can the authors make for other studies using this approach?"

3.2 Author Response

More information about the data exclusion process was provided, for considerations when using potential temperature as a coordinate (to help generalize this work) as well as filtering specific to the alkanes of interest. We also provide additional figures in the SI related to the filtering methods.

3.2 Manuscript Changes

Lines 53-138.

4.1 Referee Comment

"Lines 90-93: Please note which emission inventories were used rather than just referring to them as "default". Also, please explain how the scaling factors for the ethane and propane emissions were derived. I think I found the details in Tribby et al. (2022)—they are related to observed ethane:propane ratios?-- but more information is needed here."

4.2 Author Response

We added clarifying language for the emissions and addressed a similar question in R1 comments, 4.2 author response.

4.3 Manuscript Changes

Lines 111-120.

5.1 Referee Comment

"Line 99: While I see how some of variability across latitude is maybe collapsed when moving from pressure to potential temperature coordinates, the differences between Figure 2 and 3 are pretty subtle and I disagree with the authors' claim that the curtain plots in Figure 3 look "very similar" at the different horizontal resolutions. A more quantitative comparison of the differences between simulations is warranted here."

5.2 Author Response

We provide quantitative comparisons with our new analysis approach including simple linear regression, RMS, and RMSE.

5.3 Manuscript Changes

Figures 4-8.

6.1 Referee Comment

"Line 122-127 and Figure 4: A visual inspection is not sufficient here. Please provide some quantitative statistics to demonstrate good agreement between the simulations and aircraft data at all three horizontal resolutions and with the inclusions of the +/- 5 day window when using potential temperature as a coordinate. Also, please show how these statistics are improved relative to a comparison using lat-lon-pressure coordinates. Lastly, why was the 5-day time window chosen for comparison?"

6.2 Author Response

We have provided new quantitative comparisons between simulations and observations for pressure and potential temperature as a vertical coordinate and how the statistics were improved

(RMS, RMSE, simple linear regression comparing slope and Pearson Correlation Coefficient). The 5-day time window was chosen arbitrarily to approximate global chemical transport error.

6.3 Manuscript Changes

Figures 4-8.

7.1 Referee Comment

" Line 137-150: This should all be moved to the Methods section rather than the Results."

7.2. Author Response

These methods were removed, as we no longer utilize the Bayesian approach (see RC1, 6.2 author response).

7.3 Manuscript Changes

Lines 137-150 (Submission 1) removed.

8.1 Referee Comment

"Lines 151-158 and Table 1: This section needs the most additional work to be useful to the broader community. At the very least, it must include a comparison to inversions using lat-lon-pressure coordinates to prove that the resulting alpha is less dependent on model resolution when using potential temperature. Also, while the authors mention they did one test using the 4x5 degree simulated data as the observations in a 0.5x0.625 degree inversion, I question why the authors rely mostly on inversions using real observations rather than simulated ones in this analysis. It seems much easier to quantify the reliability of the results in the latter case, when the true flux is known and can be perturbed. Also, on that note, the authors need to address why the 95% credible interval is [0.71, 0.86] in the simulated inversion—shouldn't it be approaching 1.0 in this case? I'm unclear if that's the case for the inversion using real observations, but more discussion is needed to explain the behaviors here. Also, how dependent are the results on the spatial distribution of the emissions (i.e. if the authors used a prior with the same global flux but different spatial distribution)? Such a test would help demonstrate the limitations and applicability of this work in other contexts."

8.2 Author Response

We are no longer utilizing the Bayesian approach (see RC1, 6.2 author response).

8.3 Manuscript Changes

Figures 4-8 (new analysis), removal of Table 1.

9.1 Referee Comment

"Line 102: I think this should be referring to Figures S4-S7?"

9.2 Author Response

These now refer to Figures S9-S12.

9.3 Manuscript Changes

Lines removed.