Point-by-Point Responses to Anonymous Referee #1

We thank Anonymous Referee #1 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-RC1.

This document provides a point-by-point response to Referee #1. 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 case study of using a previously developed Bayesian approach to evaluate emission estimates of C3H8 and C2H6 from global model simulations and aircraft observations from the ATom campaign. The paper compares 3 different model simulations and demonstrates that the results are less dependent on model resolution when using potential temperature (Tpot) rather than pressure as coordinate. The main conclusion of this study is that using Tpot can safe [sic] efforts and costs since coarse resolution simulations are sufficient and provide similar results as more costly high resolution runs."

1.2 Author Response

We substantially revised the manuscript to focus on an analysis approach by creating pseudo data for C_2H_6 and C_3H_8 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 and corresponding SI.

2.1 Referee Comment

"The case study per se could be of interest, specifically to other studies using ATom data, modelers or developers of emission inventories, but in my view there is significantly more work needed before the paper is ready for publication and can provide value to the scientific community. Below some of my major concerns:

Throughout the paper the paper lists a number of limitations on when the methodology can be used and they also applied significant filtering to the data set. More information is needed on how the authors decided on the different filtering and how this could be generalized and be applicable to other cases. How could you decide if this method is applicable and valid when multiple resolution simulations are not done? Have you tested in with other species of lifetimes ~10 days or longer (e.g. CO?)"

2.2 Author Response

The referee highlighted the need for providing more information on data filtering techniques to help make the study generalizable and applicable to other cases. Below we provide our response and in section 2.3 we detail where we made changes to the manuscript.

As potential temperature (θ) is conserved following adiabatic flow, it is best used within certain dynamical conditions. The following conditions may limit its use as an effective zonal coordinate:

- θ is not conserved within moist convection and turbulent conditions, e.g., within the boundary layer.
- Synoptic-scale meteorology has a timescale of about 10 days and a horizontal length scale of greater than 1000 kilometers.[1]

Generally, θ will provide a more precise coordinate framework when: 1) the trace gasses of interest have longer atmospheric lifetimes than the synoptic meteorology timescales (~2 weeks); and 2) the region of study include conditions where θ is conserved, e.g., free extra-tropical troposphere and stratosphere.

Consistent with these constraints, our analysis is restricted to the free troposphere and the extratropics. In this region, variability within large-scale circulation can be well-captured using θ as a zonal coordinate.

Even though C_3H_8 and C_2H_6 have photochemical lifetimes longer than typical vertical and horizontal transport during the winter, they are still somewhat sensitive to the a priori zonal distribution of their emissions. We apply a number of constraints to study well-mixed parcels that are independent from any filtering techniques applied to use θ as a zonal coordinate described above. Thus, we exclude data where highly localized sources influence the mole fraction of these alkanes:

[1] Jacob, D. J. *Introduction to Atmospheric Chemistry*; Princeton University Press: Princeton, N.J, 1999; pp 52-53.

- We reduce the influence from local plumes by analyzing observations in the free troposphere and over the ocean while excluding observations taken within the boundary layer and over land masses where highly local sources exist such as energy infrastructure.
- Nearby biomass burning emissions are identified and excluded using co-measurements of HCN and CO.
- Summer observations are excluded from the analysis, as high temperatures and OH shortens the lifetime of C_3H_8 and C_2H_6 such that nearby sources dominate the variance.
- Similarly, observations in the subtropics are sensitive to transport from the extratropics, where most emissions of C_3H_8 and C_2H_6 originate. We exclude subtropical air parcels using co-measurements with tropopause pressure above 100 hPa (about 5% of data were excluded under this constraint).

Conversely, we exclude alkane observations that are poorly connected to underlying fluxes. Sources of C_3H_8 and C_2H_6 largely originate from northern hemispheric land masses. The lifetime of C_3H_8 and C_2H_6 is less than or equal to a few months during the summer, but air parcel mixing between the northern to southern hemispheres is on the order of a year.[2] As a result, the mole fraction of these gasses is relatively low in the southern hemisphere. The relatively short lifetime of C_3H_8 and C_2H_6 compared to vertical transport owes to the low abundance of these alkanes in the stratosphere. As such, we make the following restrictions:

- We arbitrarily restrict observations above 20 degrees north.
- We exclude stratospheric observations using N_2O as a tracer, which is inert and generally well-mixed in the troposphere but is quickly destroyed in the stratosphere by photolysis and reaction with O^1D.[3]

2.3 Manuscript Changes

Additional information on these filtering techniques was included in Section 2 Methods (lines 53-138), with an additional section added: “Considerations for θ as a zonal coordinate” to highlight techniques specific to this zonal coordinate. We also added two figures in the SI to show filtering. We added additional information related to generalizability in the Introduction (lines 24-51).

3.1 Referee Comment

“The highest resolution tested is 0.5x0.625 deg which is still fairly coarse. I suggest to make very clear that this methodology has only been tested on model resolutions global climate models are generally run at.”

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[2] Jacob, D. J. *Introduction to Atmospheric Chemistry*; Princeton University Press: Princeton, N.J, 1999; pp 52-53.

[3] Seinfeld, J. H.; Pandis, S. N. *Atmospheric Chemistry and Physics: From Air Pollution to Climate Change*, Third edition.; John Wiley & Sons: Hoboken, New Jersey, 2016; pp 129.

3.2 Author Response

We added this clarifying language throughout the document.

3.3 Manuscript Changes

Line 10, 12, 34, 37, 41, 42, 44, 245.

4.1 Referee Comment

“Could you please confirm that all simulations use the same base emissions and also state what inventory they are based on. It was not clear from the description. Also in Line 91 it is not clear what is scaled how? You substituted C3H8 with default C2H6? Are there no C3H8 emissions available?”

4.2 Author Response

Referee suggests clarifying base emissions used, what inventory they are based on, and how they were scaled.

Emissions for C_2H_6 and C_3H_8 were computed using a modified version of the Harmonized Emissions Component (HEMCO) Standalone version 3.5.0-rc.1 (Yantosca et al., 2022b) and GEOS-Chem 14.0.0-rc.1 (Yantosca et al., 2022a) on AWS using a public Amazon Machine Image. (Relevant default anthropogenic emissions include Tzompa-Sosa et al. 2017 for C_2H_6 and Xiao et al. 2008 for C_3H_8.) We revised the default emissions using the same methods from Tribby et al., (2022), as these default inventories were shown to underestimate observed C_3H_8/C_2H_6: Briefly, Tribby et al., (2022) they showed that substituting C_2H_6 anthropogenic categories for C_3H_8 greatly improved resulting simulations of in situ observations. The study additionally quantified observed missing high latitude emissions of these alkanes using Bayesian inference. The resulting revised emissions in Tribby et al. 2022 had good agreement with other studies (Figure 5 and Figure S62). We apply the same methods here by scaling all sectors of default C_2H_6 by 1.1 substituting C_3H_8 with default C_2H_6 before scaling by 1.2. Software packages used in this analysis include Matplotlib (Caswell et al., 2022; Hunter, 2007) and Pandas (Reback et al., 2021).

4.3 Manuscript Changes

Lines 111-120 were edited to clarify these methods.

5.1 Referee Comment

“It is highly concerning that the simulations experiences negative concentrations and that this has been fixed by simply setting these to zero. Negative concentrations indicate an issue in the model

or the setup and this simple non-physical fix does not provide high confidence in the model results. If this is a general issue and solution with the model and well documented that this does not lead to issues in the simulated fields, then this needs to be referred to in the paper."

5.2 Author Response

As explained in the initial manuscript, the C_3H_8 and C_2H_6 simulated fields did not display negative concentrations. Several aerosol species with no relationship to C_3H_8 and C_2H_6 did result in negative concentrations, an issue experienced by other users. Fortunately, a new version of GEOS-Chem, v14.3.1, provided a fix for this bug. We repeated the 0.5x0.625 nested simulations with v14.3.1 while still using the same emissions as the 4x5 and 2x2.5 simulations and did not experience these issues with the aerosol species. (Note, only the nested 0.5x0.625 had resulted in negative aerosol.) We include these new simulations for all parts of the analysis. As expected, C_2H_6 and C_3H_8 changed minimally. Below, we list Figures that incorporate these new simulated fields.

5.3 Manuscript Changes

Figure 2-8 and corresponding SI figures.

6.1 Referee Comment

" I do not see convincing information that the emission estimates are less resolution dependent using Tpot compared to pressure. E.g., how would Figure 4 look were you to use pressure. Or how would the numbers in Table 1 change if the emission estimates are using pressure as coordinate?"

6.2. Author Response

We no longer use the Bayesian approach in this study. Replicating our Bayesian model analysis using pressure proved challenging, as our Bayesian statistical model assumed a good linear correlation between the gas and the vertical coordinate. The correlation of the alkanes with pressure tended to be more scattered. This introduced high uncertainty in our interpretation of the Bayesian results and when comparing the two coordinates against each other. Additionally, we realized that our application of this Bayesian approach was likely confusing to readers since our simulations already incorporated revised emissions and thus we were not expecting the inversion to indicate a large missing source. We anticipated this confusion would be likely to detract from the key point of the manuscript.

Instead, we have 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.

6.3 Manuscript Changes

Figure 4-8.

7.1 Referee Comment

“Figure 2 and 3: the 0.5x 0.625 degree runs look significantly different from the coarser resolution results for 23 Feb for both pressure and Tpot. How do the authors explain this?”

7.2 Author Response

While some days are predicted to have slightly higher mole fractions, these data did not appear to significantly affect or skew RMS, RMSE, and slope comparisons.

7.3 Manuscript Changes

Figures 4-8.

8.1 Referee Comment

“Need to specify r2, rmse etc. for Figure 4 and related Figures in Supplement.”

8.2 Author Response

We have now included RMS, RMSE, and Pearson Correlation Coefficient for this analysis.

8.3 Manuscript Changes

Figures 4-8 and related SI figures.

9.1 Referee Comment

“The results section needs to be separated into a methodology and an actual results/discussion section.”

9.2 Author Response

We have moved methodology-related text into the methods section.

9.3 Manuscript Changes

Lines 53-138.

10.1 Referee Comment

“Methodology: Please be clear how you averaged and filtered the data. To what degree does the +/- 5 days sampling before and after the observation time contribute to reducing the resolution dependence? Effectively you degrade the higher model resolutions more than the coarsest.”

10.2 Author Response

The referee brings up an interesting point of how the +/- 5 day sampling method may degrade the higher resolution more than the coarser simulations. If this +/- 5 day sampling approach were to have the effect of significantly degrading the higher resolution simulations compared to the coarser ones, we would expect to see a dependence of the strength and uncertainty of the correlation with the sample day, but that is not the case. In Figure 5 the RMSE does not show a clear trend with sample day, and in Figure 8 the daily slope and pearson correlation coefficient do not show a significant reduction in the confidence interval for the actual flight path day, nor does the magnitude of the pearson correlation coefficient significantly vary according to sample day.

10.3 Manuscript Changes

We added some discussion on this concept, lines 240-246.

11.1 Referee Comment

“Significantly more in-depth analysis and discussion on the results are needed (see comments above). The confidence range in Table 1 does not provide a strong indication for missing sources. Without knowing what the base emissions are, whether they are representative for the year and how they compare to other inventories, you cannot say that there are missing sources but differences could also be due to uncertainties in emission factors, underestimation of emissions from specific sectors etc. It is also not clear to me, if the emissions were scaled upfront by 1.1 and 1.2 for C2H6 and C3H8, respectively and the scaling factors (Table 1) are in the order 0.8-1(1.1) and 0.9-1.1(1.2), then the original emissions might not be too low at all if the lower ranges apply.”

11.2 Author Response

Please see response in section 6.2.

11.3 Manuscript Changes

Please see response in section 6.3.

12.1 Referee Comment

“Table 1 results for the highest resolution run are actually more different from the two other simulations. This might be related to my question related to Figures 2&3 or simply indicates that there is still some resolution dependence? Have you rerun your simulations with the lower and upper ranges of your estimates to see whether you improve the comparison to aircraft data?”

12.2 Author Response

While we have not rerun the simulations with the lower and upper ranges of the revised emissions, RMS and RMSE do not indicate a strong resolution dependence.

12.3 Manuscript Changes

Figure 4 and 5.

13.1 Referee Comment

“It also needs to be clearly stated how much new information this study provides beyond what has been done in Tribby et al. (2022).”

13.2 Author Response

We added clarifying language.

13.3 Manuscript Changes

Line 34-36.

14.1 Referee Comment
“The paper is missing a Summary/Conclusion.”

14.2 Author Response
We added this section.

14.3 Manuscript Changes
Lines 240-251.