We sincerely thank the reviewers for their insightful comments and valuable suggestions, which have been instrumental in significantly improving the quality and clarity of our manuscript. We have carefully addressed all points raised and conducted thorough revisions throughout the paper. The key changes implemented in response to the reviewers' feedback include:

1. We have substantially improved the comparisons of our data with the most up-to-date inventories, and the data presentation: 1) Our anthropogenic CO emission data are now comprehensively compared with the CEDS-CMIP6 and CEDS-CMIP7 emission inventories, as well as the dataset from Zheng et al. (2019); Our biomass burning CO emission estimates are now systematically compared with the GFED5 dataset; To improve clarity, the visualization of these emissions in Figures 3 (anthropogenic) and 5 (biomass burning) has been updated from 12-month moving averages and monthly means to annual averages.
2. We have added two new figures (Figures 6 and 7) to deepen the analysis of biomass burning. These show monthly averages and monthly trends (2003-2022), alongside the corresponding trends in CO column concentrations attributable to biomass burning changes (2003-2022).
3. We have conducted and present uncertainty analysis for biomass burning emissions and their influence on CO concentrations, revealing that the pronounced interannual trends are predominantly driven by the exceptional wildfire events of 2021.
4. The content of the original Section 3.3 ("Impacts of systematic errors on inferred CO emissions") has been moved to Section 3.4. The new Section 3.4 has been significantly condensed, with its length reduced by approximately 50% compared to the original Section 3.3.

Below, we provide detailed point-by-point responses to each of the reviewers' specific comments.

Reviewer #1

The manuscript «Global CO emissions and drivers of atmospheric CO trends constrained by MOPITT satellite observations» use 20 years of retrieved CO from MOPITT and GEOS-Chem simulations in an 4D-Var assimilation framework to provide inverse estimates of CO emissions. A central finding in this study is a significant long-term decline in global anthropogenic CO emissions, with regional variations. The sensitivity to varying and constant OH as well as using MOPITT CO column or profile data are tested, and the results were not so sensitive to these different choices.

Answer: Thank the reviewer for the valuable comments! The manuscript has been revised carefully based on the comments.

Question: It is very important and useful to evaluate trend in emissions of components important for air-quality and the oxidation capacity of the atmosphere. As the emission inventories to be used in CMIP7 is available (CEDS version 2025, https://github.com/JGCRI/CEDS/) it would be very useful if you can compare your results

with these. It would also be very useful if you could compare your results with the version of CEDS used in CMIP6 as well. As these emission datasets are commonly used in model simulations, and you use a combination of different emission inventories as your prior, the results will be even more relevant for atmospheric modellers.

Answer: We thank the reviewer for this important suggestion! We have added a comprehensive comparison of our inferred anthropogenic CO emissions with the CEDS-CMIP6 dataset, the new CEDS-CMIP7 dataset, and the data from Zheng et al. (2019). As shown in the updated Fig. 3, the comparison reveals that the interannual variability and long-term trend of top-down CO emissions (this work and Zheng et al. 2019) are generally consistent with the CEDS-CMIP7 inventory. Both top-down estimates align more closely with the CEDS-CMIP7 trend than with the CEDS-CMIP6 trend, which highlights the improvements in the updated inventory. However, the emission magnitudes derived from the inverse modelling are higher than those in CEDS-CMIP7 inventory.

Reference:

Zheng, B., Chevallier, F., Yin, Y., Ciais, P., Fortems-Cheiney, A., Deeter, M. N., Parker, R. J., Wang, Y., Worden, H. M., and Zhao, Y.: Global atmospheric carbon monoxide budget 2000–2017 inferred from multi-species atmospheric inversions, Earth Syst. Sci. Data, 11, 1411-1436, 10.5194/essd-11-1411-2019, 2019.

Question: As you wrote at the beginning of the introduction: "Carbon monoxide (CO) is a key atmospheric pollutant produced from incomplete combustion and the oxidation of hydrocarbons.» It is no clear for me how you treat the source from oxidation of hydrocarbons in your setup. You wrote: «The CO source from CH4 oxidation is optimized separately as an aggregated global source, with the a priori uncertainty of 25%.» CH4 has increased by ~133 ppb over the period of investigation. Do you take the increase in methane into account? And what about the oxidation of other VOCs? You wrote that you employ a CO only simulation (on Line 108). How is the atmospheric production treated in these simulations? Biogenic VOC are mentioned, but what about anthropogenic VOC emissions? Is the production of CO from biogenic VOCs sensitive to the different OH fields. Only CO loss is mentioned related to the OH sensitivity.

Answer: In our CO-only simulation, the chemical production of CO from the oxidation of hydrocarbons is included. The CO sources from both bVOCs and CH_4 oxidation are calculated online based on the assumption of instantaneous oxidation by OH radicals. Specifically, bVOC oxidation is treated as a surface source, while CH_4 oxidation occurs throughout the atmosphere. As a result, both production pathways are directly sensitive to the OH concentration field used in the simulation. On the other hand, as demonstrated in the section "Impacts of systematic errors on inferred CO emissions", the difference between the two OH fields (default and TCR-2), which showed a 10-40% discrepancy in OH concentrations, led to only a 3.7% difference in the derived global anthropogenic CO emissions. This limited sensitivity in the emission estimates is attributed to our two-step inversion framework, which is designed to mitigate systematic biases associated with transport and chemistry parameters.

For anthropogenic non-methane VOCs, their chemical production of CO is not simulated explicitly online. Instead, this contribution is accounted for by scaling up the

primary anthropogenic CO emissions by 11%, based on a priori source inventory estimates.

The model uses a prescribed, spatially varying CH_4 field following the default tagged-CO configuration described by Fisher et al. (2017). Upon checking, the specific dataset used is from the year 2013 and is held constant interannually. Over our study period, the global mean CH_4 concentration is about 1850 ppb. Using a fixed 2013 concentration therefore introduces a maximum potential bias of approximately ±4% in the calculated CO source from CH_4 oxidation. Our inversion system treats the global CO source from CH_4 oxidation as an adjustable parameter (with a prior uncertainty of 25%) within the two-step inversion framework. This design allows the system to absorb and correct for such systematic biases in the chemical source. Consequently, the potential bias from not interannually varying CH_4 is expected to have a limited impact on our final inverted anthropogenic and biomass burning CO emissions.

The Methods (Section 2.1) have been revised to provide a more detailed and precise description.

Reference:

Fisher, J. A., Murray, L. T., Jones, D. B. A., and Deutscher, N. M.: Improved method for linear carbon monoxide simulation and source attribution in atmospheric chemistry models illustrated using GEOS-Chem v9, Geosci. Model Dev., 10, 4129-4144, 10.5194/gmd-10-4129-2017, 2017.

Question: You mention that the biomass burning exhibit strong interannual variability, but indicate with only a single number without any uncertainty on the offset of the increase in the anthropogenic emissions. Uncertainties in these numbers should be added. The biomass burning CO emissions have a large interannual variability, and clearly 2021 will impact your trend results at high latitudes. Would it be interesting to look at how the seasonal emissions and impact on concentration have changed over the 20 year period?

Answer: Thank the reviewer for the valuable and constructive suggestions. We have performed additional analyses and revised the manuscript. The main updates are detailed below:

We have now quantified the uncertainties in both the estimated trends of biomass burning CO emissions and their impact on atmospheric CO columns. In the revised manuscript, Table 3 and Table 4 now provide trends based on annual means along with their uncertainties (expressed as ±1 standard deviation derived from the linear fitting analysis). This quantitatively addresses the interannual variability in the trend estimates. To further investigate the uncertainty on a finer temporal scale, we have added two new supplementary figures. Fig. S4 and Fig. S5 (provided in the SI) present the monthly-mean trends and their corresponding uncertainties for biomass burning CO emissions and their column impacts, respectively.

As shown in the new Fig. 7, at both the global and Northern Hemispheric scales, the previously noted positive trend in CO columns attributable to biomass burning was largely driven by the exceptional wildfire activity in 2021. When the year 2021 is excluded from the trend calculation, the long-term trend in CO columns due to biomass burning becomes less pronounced at these broad scales. This highlights the dominant role of interannual variability, as the reviewer suggested, and clarifies that the positive

trend is not a robust feature of the full 20-year period outside of this extreme event. This analysis is now discussed in the revised text.

We agree that examining how the seasonal patterns of emissions and their impacts have evolved over the study period is highly interesting and insightful. First, the new Fig. 6 displays the climatological monthly cycle (2003-2022 average of monthly means) of biomass burning CO emissions for different regions, clearly illustrating their distinct seasonal phasing. Building on this, the new Fig. 7 presents the monthly-mean trends over the 2003-2022 period for both biomass burning CO emissions and their resulting impact on CO columns (Please check Fig. S4 and S5 for the standard deviation of the trends)**.** This analysis reveals a clear and significant seasonal pattern in the trends.

Question: Especially related to biomass burning emissions and corresponding concentration changes but also in general: How are the trends in the manuscript calculated, what are the uncertainties in these trends and are they significant?

Answer: We used linear fitting to calculate the trends (slopes) and their uncertainties (standard deviations). Table 3 and Table 4 now provide trends based on annual means along with their uncertainties; Fig. S4 and Fig. S5 (provided in the SI) present the monthly-mean trends and their corresponding uncertainties.

In general, trends in anthropogenic emissions (Table 2) are mostly statistically significant on an interannual scale. In contrast, interannual trends in biomass burning CO emissions and their impacts on CO columns, calculated from annual averages, are largely statistically insignificant (as shown in Tables 3 and 4). However, when trends are derived from monthly averages, certain months and regions do exhibit statistical significance, particularly when the intense wildfire year of 2021 is taken into account (Fig. S4 and Fig. S5).

Smaller comments:

Question: Abstract: You should mention the model and method used. On L29 you mention posteriori simulations without mentioning the model and method.

Answer: The abstract has been revised to clarify the model and method: "*we analyze changes in global CO emissions and atmospheric concentrations by applying a four-dimensional variational (4D-Var) assimilation framework within the GEOS-Chem adjoint model*".

Question: Throughout the manuscript: I would not use the word "observations" when the MOPITT CO data is mentioned.

Answer: The usage of "observations" associated with MOPITT has been replaced with words such as "retrievals", "measurements" and "data".

Question: L68: "This has profound implications, as CO shares common combustion sources with major greenhouse gases like methane (CH4) and carbon dioxide". I did not fully got what you mean here. CO is emitted due to incomplete combustions. CO2 is also emitted from complete combustion of fossil fuels. We should reduce emissions of greenhouse gases CO2 and CH4 and as a co-benefit CO emissions are reduced. Another co-benefit is less global warming and reduced climate induced biomass burning CO emissions.

Answer: Thank the reviewer for this thoughtful comment! As the reviewer indicated, reducing anthropogenic greenhouse gas emissions yields the co-benefit of lowering CO emissions, and mitigating warming can in turn help reduce climate-driven biomass burning.

Our point here was complementary, focusing on a potential reverse effect: the observed increase in biomass burning CO emissions, which may be offsetting the decline in anthropogenic CO emissions, serves as an indicator that greenhouse gases from the same fires are also likely to increase. If these wildfire-driven greenhouse gas emissions are growing, they could be offsetting a portion of the reductions achieved in anthropogenic greenhouse gas emissions.

The discussion has been revised to clarify this point: "*This has profound implications, as a rise in wildfire CO signals a concurrent rise in wildfire greenhouse gas emissions, which could offset part of the gains achieved from reductions in anthropogenic greenhouse gas emissions*".

Question: L87 and L106: HEMCO is not defined.

Answer: The definition for HEMCO was added.

Question: L113: For the reader it would be good if you can say if there are any trend in OH in that dataset. Looking at Fig. S2, what is the unit on the y-axis?

Answer: Global mean tropospheric OH concentrations from TCR-2 demonstrate a slight increasing trend of $1.0 \pm 0.6 \times 10^3$ molec cm^{-3} yr^{-1} in 2005-2020. The description has been revised.

The y-axis unit, 10^6 molec cm^{-3}, has been added to Fig. S2.

Question: L115: Not all emission inventories used cover all years up to 2023. What is the end year and what do you do with the following years?

Answer: As shown in Fig. 3, a priori emission inventories used in this study have different temporal coverages. For years beyond the end year of a specific inventory, emissions from the last available year within that inventory's coverage were used to fill the subsequent years. This point was clarified in the revised version.

Question: L155: HIPPO not defined yet. ATOM and WDCGG not defined yet either.

Answer: The definition for HIPPO, ATOM and WDCGG were added.

Question: L157: "accumulation of biases over preceding months". Can these biases influence your trend results?

Answer: Given that our 4D-Var inversion uses a time window of one month, a negative bias in CO concentration accumulated from the previous month (i.e., an error in the initial conditions) can affect the current month's emission estimate. This is because the inversion will adjust the emissions within that month's window to compensate for the initial discrepancy, thereby introducing an error into the inferred emissions for that specific month.

However, their effect on our trend analysis should be minor:

1) Trend of CO emissions: Our 4D-Var system uses initial conditions that have been optimized through sequential Kalman filtering. This effectively mitigates the accumulation of biases from prior months, thereby minimizing their impact on the derived emission trends over time.

2) Trend of atmospheric CO concentration: Our trend analysis is based on a 20-year simulation (2003-2022). The initial conditions (January 2003) are also optimized via sequential Kalman filtering, and the CO emissions used throughout the simulation are constrained by the 4D-Var inversion. Consequently, the long-term concentration trends are not substantially affected by biases accumulated over individual months.

Question: L198: "low cloud observations" unclear what it means. Clouds at low altitudes?

Answer: The text has been updated to a more precise description: "We only consider data with Cloud Description = 2 (cloud free)".

Question: L269: What is the difference between the results in this study and Fortems-Cheiney?

Answer: The major difference between this study and Fortems-Cheiney et al. (2024) lies in the estimated trend of CO emissions over Europe. Fortems-Cheiney et al. reported a continued decline in European CO emissions during 2011-2021, whereas our analysis indicates a slowdown in the decline of European CO emissions after 2017. This discrepancy may be attributed to differences in the treatment of initial and boundary CO conditions: Fortems-Cheiney et al. employed climatological mean CO concentrations, while in this work we used optimized initial and land boundary CO conditions.

Question: L291: Sentence starting with "Comparisons with the CEDS inventory." This is more a description of the anthropogenic emissions in CEDS rather than a comparison with the CEDS inventory. As mentioned above, a comparison with CEDS (both CMIP6 version and CMIP7 version will be useful).

Answer: The original sentence in this paragraph was replaced by a comparison with Zheng et al. 2019 and CEDS-CMIP7: "*Our estimated emission magnitude and trend are similar to Zheng et al. (2019) in India. In comparison, the CEDS-CMIP7 inventory shows a similar trend, but its emission levels are lower than those derived from inverse modeling*".

Question: L299: What is an environmental Kuznets curve?

Answer: This sentence was removed in the revised version.

Question: L303: Would also be useful to compare with other emission inventories, as GFED5 which is recently released. https://www.globalfiredata.org/data.html

Answer: Thank the reviewer for this suggestion! We have added a comparison of our inferred biomass burning CO emissions with the GFED5 dataset. As shown in the updated Fig. 5 and Fig. 6, our emission estimates suggested the historical peak of biomass burning CO emissions by approximately 500 Tg yr^{-1} in 2021, while the overall CO emissions in GFED5 inventory are higher than our estimates in 2003-2022. GFED5

data suggests that boreal Asia's wildfire CO emissions peaked in 2012, different from emission inversion results in this work and Zheng et al. (2023).

Reference:

Zheng, B., Ciais, P., Chevallier, F., Yang, H., Canadell, J. G., Chen, Y., van der Velde, I. R., Aben, I., Chuvieco, E., Davis, S. J., Deeter, M., Hong, C., Kong, Y., Li, H., Li, H., Lin, X., He, K., and Zhang, Q.: Record-high CO_2 emissions from boreal fires in 2021, Science, 379, 912-917, 10.1126/science.ade0805, 2023.

Question: L459: Series of sensitivity experiments. I can not see that these sensitivity experiments to attribute concentration changes to different drivers are defined.

Answer: The following paragraph was added to provide a better definition for the sensitivity experiments:

"*To quantitatively attribute the concentration trends to specific drivers, we conducted a series of sensitivity experiments. The experimental design isolates the influence of individual emission sectors by building a baseline scenario in which all emissions are fixed at 2003 levels to reflect the impact of meteorological condition changes in 2003-2022. Three more sensitivity experiments were then conducted in 2003-2022 in which only one emission category, i.e., anthropogenic, biomass burning, or biogenic VOC emissions, was allowed to vary over time, respectively. The time-varying emissions in these sensitivity experiments were prescribed from the Column-FixOH a posteriori inversion*".

Question: Table 2: What is the unit of the trend. I find it easier to read the table if the unit is added to the table. Eg. after Emissions and Trend. What is the definition of the trend uncertainty estimate.

Answer: In Table 2, the "Emissions" are presented in units of Tg yr^{-1} (teragrams per year). Consequently, the linear trend, which represents the annual change in emissions, has units of Tg yr^{-2} (teragrams per year squared). The "uncertainty" is $\pm 1\sigma$ standard deviation of the estimated linear trend.

To maintain a clean table format, we have chosen to include the unit information in the table caption. However, if the reviewer prefers the units to be listed directly in the table, we would be happy to revise the table accordingly. We appreciate the reviewer's valuable feedback!

Question: Table 3: The trend is missing in this table. Important to also add the uncertainty in the trend calculations here.

Answer: The trends and their uncertainties have been added in Table 3. Thank the reviewer for pointing out this issue!

Question: Fig. 1: Is Fig1c CO emissions? CO production due to photooxidation of biogenic VOCs? As mentioned above, what about anthropogenic VOCs? And methane? The colour scale in a to c should be the same for easier comparison.

Answer: Fig. 1c is actually oxidation of biogenic VOCs. The figure title has been revised to clarify this point. As suggested, we have adjusted Figs. 1a–1c to use identical

color scales, which facilitates a clearer visual comparison across the different CO sources.

For details on the calculation methods for anthropogenic VOCs and CH_4 oxidation, please refer to the response provided earlier. In brief, the CO from anthropogenic VOC oxidation is derived by applying a fixed enhancement ratio to the anthropogenic CO emissions. In contrast, the CO from bVOCs and CH_4 oxidation is calculated online, based on OH concentrations.

Question: Fig. 3: Add y-label. This comment is relevant for several figures. Here it would be very useful if the CEDS emissions to be used in the upcoming CMIP7 as well as those emissions used in CMIP6 (up to 2014) could be added.

Answer: We have checked and updated all figures to ensure the y-labels are added in the revised version. As mentioned in the previous response, we have added comparisons in Figure 3 with the CEDS-CMIP6, CEDS-CMIP7 emission inventories, as well as the dataset from Zheng et al. (2019).

Question: Fig. 4: Maybe due to the limited description of how the attribution to the different sources were made, why did you exclude areas with large biomass burning emissions from the trends in anthropogenic and biogenic VOCs?

Answer: The reason for excluding grids with high biomass burning emissions when calculating trends for anthropogenic and biogenic VOCs in Fig. 4 is as follows:

Treatment for Fig. 4 (grid-cell emission trends): Biomass burning emissions exhibit strong seasonality, whereas anthropogenic emissions show relatively weaker seasonal variation. To prevent periods of intense biomass burning from obscuring the long-term trend of anthropogenic sources, we excluded months in which the contribution of biomass burning exceeded 50% at each grid cell when calculating trends for non-biomass burning emissions. This approach ensures that the derived trends more accurately reflect the actual changes in anthropogenic emissions, without being biased by short-term, seasonal biomass burning signals.

Treatment for Fig. 3 (regional-averaged time series) and Table 2 (regional trends): For the regional average analysis (e.g., over Southeast Asia) on an annual mean basis, we did not apply the above monthly exclusion. This is because, at a large regional scale, the influence of biomass burning on the long-term trend of annual total anthropogenic emissions is relatively limited and does not substantially alter the overall direction or magnitude of the anthropogenic emission trend. Thus, the regional-averaged trends remain robust in representing the long-term changes in anthropogenic emissions.

The Caption of Fig.4 has been revised to provide better clarification.

Question: Fig. 5: Here you both show the large interannual variability of the biomass burning emissions and the seasonal variability. One of the main point you made is the offset of the air quality improvement due to reduction in anthropogenic CO emissions by the increase in biomass burning CO emissions. Clearly the year 2021 will impact the results. How robust is the trend you calculate. Would it be more interesting to look at trend in different seasons and the impact on air quality for seasons. At high latitudes, vegetations do not burn during winter.

Answer: As mentioned in the previous response, we have conducted and present uncertainty analysis for biomass burning emissions and their influence on CO concentrations. In general, interannual trends in biomass burning CO emissions and their impacts on CO columns, calculated from annual averages, are largely statistically insignificant (as shown in Tables 3 and 4). However, when trends are derived from monthly averages, certain months and regions do exhibit statistical significance, particularly when the intense wildfire year of 2021 is taken into account (Fig. S4 and Fig. S5).

Question: Fig. 7: "Only stations with 20 year observations (the time range between the first and last observations) during 2003-2022 are included" Can you simply say, "Only stations with observations from 2003 to 2022 are included"?

Answer: We rechecked our analysis and found that the original analysis used stations with at least 10 years of observational records from 2003-2022. We have therefore revised all analysis involving WDCGG surface stations (including figures and tables), now using stations with a minimum of 14 years of data. This standard (i.e., 14 years of data) was made because applying a longer data record threshold would have resulted in an insufficient number of stations. The updated analysis does not alter the main conclusions of this study.

Reviewer #2

This paper is relevant, well written and helps to explore recent trends in CO concentrations, which have not been clearly understood previously. It has potentially important impacts for the link between carbon monoxide and climate-change (and wildfires) and I endorse the publication of this manuscript, provided the suggestions below are addressed.

Answer: Thank the reviewer for the valuable comments! The manuscript has been revised carefully based on the comments.

Question: My primary concern is the structure of the manuscript. I would suggest moving Section 3.3 to allow a more cohesive paper and have explained this in more detail in the specific comments below. A clearer model description (and explanation of the tagged-CO mode) is also required to explore how the VOCs and OH are being treated by the model and a budget comparison to other studies would benefit the paper. Some minor changes to the figures are suggested in the final section of comments, mostly relating to consistency between the plots. More generally, there are assumptions made throughout the manuscript which are not clearly explained and instances where comparisons are made between model runs or observations and model runs where it is not clearly defined which way the comparison is being done, and so the difference becomes difficult to interpret. These need to be addressed before publication.

Answer: We sincerely thank the reviewer for this comprehensive and constructive feedback. We have thoroughly addressed these concerns throughout the manuscript, and the key improvements are summarized as follows:

Regarding manuscript structure: Following the reviewer's primary suggestion, we have moved the original Section 3.3 to become the new Section 3.4. Furthermore, this section

has been significantly condensed (by approximately 50%) to improve the paper's cohesiveness and flow, as also suggested in the specific comments.

Regarding model description clarity: We have substantially revised Section 2.1 to provide a clearer and more detailed explanation of the tagged-CO model. This includes explicitly elaborating on the treatment of the chemical sink (linearized with archived OH fields) and the sources from VOC oxidation (including biogenic and anthropogenic precursors) and CH_4 oxidation.

Regarding budget comparison and assumption clarification: We have added a comprehensive comparison of our inferred anthropogenic CO emissions with the latest inventories (CEDS-CMIP6, CEDS-CMIP7) and the study by Zheng et al. (2019) (updated Fig. 3). Throughout the manuscript, we have carefully reviewed and clarified ambiguous statements, assumptions to enhance interpretability.

Regarding figure consistency: We have standardized the color schemes across all figures and improved the visualization of data ranges (e.g., adjusting color bars to start at zero where appropriate) to ensure consistency and clarity.

We believe these revisions have significantly improved the manuscript's clarity, structure, and robustness. Detailed point-by-point responses to each specific comment are provided below.

Specific Comments:

Question: Line 24: suggest changing atmospheric chemistry to atmospheric oxidation capacity

Answer: Changed.

Question: Line 26: clarify 'global environmental conditions'.

Answer: This sentence has been revised: "*Accurate quantification of its global emissions and the underlying driver behind its atmospheric trends is essential for understanding changes in global atmospheric environment*".

Question: Line 28: remove 'here'.

Answer: Changed.

Question: Line 29 - 30: Change 'the a posteriori' and 'the a priori' to 'a posteriori' and 'a priori'.

Answer: All relevant locations in the manuscript have been changed.

Question: Line 35-36: 'In contrast' is confusing as the opposite to the decreasing trend in anthropogenic emissions is an increasing trend, not interannual variability, which is the next statement made. In Line 305 it is stated that there is no significant long-term trend in the biomass burning emissions. Make this clear in the abstract as it currently reads as though there may be an increasing trend.

Answer: the word "In contrast" was removed in the revised version. Thank the reviewer for pointing out this issue!

Question: Line 37: In the conclusion, 47% is discussed, not 37%. I think this is perhaps the CO column concentration, but 37% is not mentioned again. Please clarify.

Answer: The 37% represents the offset effect globally, while 47% is specific to the Northern Hemisphere. To maintain consistency with the revised conclusions, we have made revision in the Abstract: "*in particular, the intense 2021 wildfires substantially offset the anthropogenic emission-driven decline in atmospheric CO over the Northern Hemisphere*".

Question: Line 55: Include further information on the impacts of CO on air quality and climate.

Answer: We have restructured the first two paragraphs of the Introduction. The first paragraph now primarily discusses the importance of CO, including its climate relevance, while the second paragraph focuses on the scientific question. In the first paragraph, the description regarding climate relevance is presented as follows:

"*By modulating the abundance of OH, changes in CO concentrations indirectly affect the atmospheric lifetime of methane (CH_4). Furthermore, CO shares common combustion sources with major greenhouse gases like CH_4 and carbon dioxide (Worden et al., 2017; Zheng et al., 2023)*".

Question: Line 65: Explain further 'growing sensitivity to climate change'.

Answer: The discussion on climate relevance has been integrated into the first paragraph of the Introduction. Consequently, this sentence has been revised: "*biomass burning emissions exhibit strong interannual variability*".

Question: Line 92: In particular.

Answer: Changed.

Question: Line 107: What is the model top altitude?

Answer: The pressure at the top of the model is 0.01 hPa. Changed.

Question: Line 108: More detail needed to explain the tagged-CO mode.

Answer: Thank the reviewer for this suggestion! Section 2.1 has been revised to provide more details about the tagged-CO mode. The first paragraph (Section 2.1) primarily discusses CO sinks: "*the chemical sink of CO is linearized with archived monthly mean OH fields*". The second paragraph focuses on CO sources: "*The contribution of co-emitted anthropogenic VOC sources is considered by scaling up anthropogenic CO emissions by 11%. Biogenic emissions are simulated using the Model of Emissions of Gases and Aerosols from Nature, version 2.0 (MEGANv2.0, Guenther et al. (2006)). CH_4 oxidation source is considered by using a prescribed, spatially varying CH_4 field following the default tagged-CO configuration (Fisher et al., 2017). The CO sources from both biogenic VOCs and CH_4 oxidation are calculated online based on the assumption of instantaneous oxidation by OH radicals*".

Question: Line 123: Comment on the trends in CO budget and compare to other budgets e.g. Zheng et al. 2019 (https://essd.copernicus.org/articles/11/1411/2019/). This could be included as text or a table, and if possible include sink sizes also.

Answer: We thank the reviewer for this important suggestion! We have added a comprehensive comparison of our inferred anthropogenic CO emissions with the CEDS-CMIP6 dataset, the new CEDS-CMIP7 dataset, and the data from Zheng et al. (2019). As shown in the updated Fig. 3, the comparison reveals that the interannual variability and long-term trend of top-down CO emissions (this work and Zheng et al. 2019) are generally consistent with the CEDS-CMIP7 inventory. Both top-down estimates align more closely with the CEDS-CMIP7 trend than with the CEDS-CMIP6 trend, which highlights the improvements in the updated inventory. However, the emission magnitudes derived from the inverse modelling are higher than those in CEDS-CMIP7 inventory.

Question: Line 134: What is the 50% assumed a priori error based on?

Answer: We assume 50% uncertainty because of the combination of combustion-related CO sources and the oxidation source from biogenic VOCs following previous studies (Jiang et al., 2013; Jiang et al., 2017). The citation has been added.

References:

Jiang, Z., Jones, D. B. A., Worden, H. M., Deeter, M. N., Henze, D. K., Worden, J., Bowman, K. W., Brenninkmeijer, C. A. M., and Schuck, T. J.: Impact of model errors in convective transport on CO source estimates inferred from MOPITT CO retrievals, J. Geophys. Res.-Atmos., 118, 2073-2083, 10.1002/jgrd.50216, 2013.

Jiang, Z., Worden, J. R., Worden, H., Deeter, M., Jones, D. B. A., Arellano, A. F., and Henze, D. K.: A 15-year record of CO emissions constrained by MOPITT CO observations, Atmos. Chem. Phys., 17, 4565-4583, 10.5194/acp-17-4565-2017, 2017.

Question: Line 134: Consider renaming as the GC-original simulation also uses a priori emissions inventories, so naming the simulation with the Kalman filter as a priori is confusing. Suggest name change to GC-Kalman.

Answer: We appreciate the reviewer's suggestion! After careful consideration, we have decided to retain the name "GC-a priori" for the following reasons:

1. Although a sequential Kalman filter is applied to optimize initial and land boundary conditions in GC-a priori, its primary purpose is to provide a low-bias baseline using unadjusted emissions. Renaming it to "GC-Kalman" might lead readers to believe it involves the use of Kalman Filter for emission optimization, which is not the case and might cause methodological confusion with the 4D-Var.

2. The term "a priori" accurately reflects the fact that this simulation uses a priori emission inventories without optimization of emissions, which is a key point of contrast with the subsequent 4D-Var-based experiments (e.g., Column-FixOH, Profile-FixOH, Column-VarOH) where emissions are optimized. Maintaining the name "GC-a priori" helps emphasize the distinction between a priori emissions and optimized emissions, rather than shifting focus to the data assimilation methods.

Question: Line 157: Clarify how model spin-up period was decided.

Answer: We employed a sequential Kalman Filter to assimilate MOPITT CO data and optimize atmospheric CO concentrations, with the assimilation period spanning from

October 1, 2002, to December 31, 2022. Consequently, for the first month of the 4D-Var emission inversion (January 2003), the spin-up period consisted of three months of sequential Kalman Filter assimilation. For subsequent months of the emission inversion, the initial conditions were taken directly from the Kalman Filter-optimized CO concentration fields at the corresponding time points.

The description in the text has been supplemented to highlight the time period of the sequential Kalman Filter assimilation: October 1, 2002, to December 31, 2022.

Question: Line 159: (and S3 caption). What are the red-grids referring to? The figure shows a pink mask over the oceans to my eye, not red grids.

Answer: As the reviewer pointed out, oceanic grid points are indeed marked in pink rather than red. The description has been changed.

Question: Line 175: Include a description of how OH (and VOCs) is being modelled before the experiments are explained. I think it would also add clarity to the paper to instead rename these experiments with Profile and Column not Col and Prof as shortening by only a few letters seems unnecessary and will aid the flow of the text.

Answer: For details on the modelling methods for OH and VOCs, please refer to the response provided earlier. In brief, the chemical sink of CO is linearized with archived monthly mean OH fields; Biogenic emissions are simulated using MEGAN, and CO sources from VOCs oxidation are calculated online based on the assumption of instantaneous oxidation by OH radicals.

As the reviewer suggested, all abbreviated names in the text and figures (Col-FixOH, Prof-FixOH, Col-VarOH) have been changed to their full names (Column-FixOH, Profile-FixOH, Column-VarOH).

Question: Line 198: Justify the exclusion of data below this threshold.

Answer: In the 4D-Var method, the adjoint forcing is calculated based on (simulation minus observation) / observation error, where the observation error is a combination of 10% uniform error and the MOPITT CO retrieval error covariance in this study. Consequently, satellite-observed CO columns within extremely low ranges (e.g., less than 5×10^{17} molec/cm^2) may have low observation errors due to the 10% uniform error (calculated as satellite-observed CO column concentration × 10%), which increase the weight of those observations in the cost function. If some extremely low column observations are inaccurate, they could adversely affect the overall performance of the 4D-Var assimilation. For this reason, a threshold of 5×10^{17} molec cm^{-2} was applied in the 4D-Var assimilation following previous studies (Jiang et al., 2013; Jiang et al., 2017).

The description has been revised to provide a better clarification: "*The threshold (5×10^{17} molec cm^{-2}) was selected to prevent the influence of certain potentially inaccurate, extremely low-concentration observations, which may also have low observation errors in the cost function, on the 4D-Var assimilation (Jiang et al., 2013; Jiang et al., 2017)*".

References:

Jiang, Z., Jones, D. B. A., Worden, H. M., Deeter, M. N., Henze, D. K., Worden, J., Bowman, K. W., Brenninkmeijer, C. A. M., and Schuck, T. J.: Impact of model errors in convective transport on CO source estimates inferred from MOPITT CO retrievals, J. Geophys. Res.-Atmos., 118, 2073-2083, 10.1002/jgrd.50216, 2013.

Jiang, Z., Worden, J. R., Worden, H., Deeter, M., Jones, D. B. A., Arellano, A. F., and Henze, D. K.: A 15-year record of CO emissions constrained by MOPITT CO observations, Atmos. Chem. Phys., 17, 4565-4583, 10.5194/acp-17-4565-2017, 2017.

Question: Line 199: Is there any danger of a systematic bias being introduced by only using day-time data? For example, anthropogenic emissions likely having a diurnal cycle. Clarify if comparisons to observational data have been corrected, or if effort has been taken to compare to day-time observations.

Answer: The MOPITT data used in this study is from the joint retrieval combining TIR and NIR radiances. Since the NIR channel relies on reflected solar radiation, only daytime data are considered in this work. This criterion is primarily based on previous research, particularly the work of the MOPITT science team. The impact of the diurnal cycle on the emission inversion was not considered in the present study. We thank the reviewer for highlighting this issue and will carefully evaluate the influence of the diurnal cycle in our future work.

References:

Worden, H. M., Deeter, M. N., Edwards, D. P., Gille, J. C., Drummond, J. R., and Nédélec, P.: Observations of near-surface carbon monoxide from space using MOPITT multispectral retrievals, J. Geophys. Res.-Atmos., 115, D18314, 10.1029/2010jd014242, 2010.

Hedelius, J. K., Toon, G. C., Buchholz, R. R., Iraci, L. T., Podolske, J. R., Roehl, C. M., Wennberg, P. O., Worden, H. M., and Wunch, D.: Regional and urban column CO trends and anomalies as observed by MOPITT over 16 years, J. Geophys. Res.-Atmos., 126, e2020JD033967, 10.1029/2020jd033967, 2021.

Gaubert, B., Anderson, J. L., Trudeau, M., Smith, N., McKain, K., Pétron, G., Raeder, K., Arellano, A. F., Granier, C., Emmons, L. K., Ortega, I., Hannigan, J. W., Tang, W., Worden, H. M., Ziskin, D., and Edwards, D. P.: Nonlinear and Non-Gaussian Ensemble Assimilation of MOPITT CO, J. Geophys. Res.-Atmos., 129, 10.1029/2023jd040647, 2024.

Tang, W., Gaubert, B., Emmons, L., Ziskin, D., Mao, D., Edwards, D., Arellano, A., Raeder, K., Anderson, J., and Worden, H.: Advantages of assimilating multispectral satellite retrievals of atmospheric composition: a demonstration using MOPITT carbon monoxide products, Atmos. Meas. Tech., 17, 1941-1963, 10.5194/amt-17-1941-2024, 2024.

Question: Line 223: Smaller not lower.

Answer: Changed.

Question: Line 228: Not clear why the units have swapped in these lines from the above units.

Answer: In Lines 223-225, we compared the model simulation results with satellite-observed column concentrations; therefore, the unit molec cm^{-2} was used. In Lines 226-228, we compared the model simulation results with aircraft observations; hence, the unit ppb was used.

Question: Line 246: This section would benefit with a description as to how source types are discerned in the model (e.g. how emissions are determined to be anthropogenic or biomass in origin). This may be placed earlier with the expanded definition of the tagged-CO mode.

Answer: Thank the reviewer for this suggestion! The following paragraph was added at the end of Section 3.1:

"*Given this confidence in the system's performance, we now present the central findings of this study: the long-term evolution of CO emissions. As mentioned in Section 2.1, the combustion-related CO sources and the oxidation source from biogenic VOCs are combined, and thus, the inverse system optimizes total CO emissions within each model grid cell. The subsequent attribution of emissions to specific source types (e.g., anthropogenic, biomass burning) in an individual grid cell is based on the relative contribution of each source category from a priori emission inventories. Specifically, a posteriori emission for a given source type in a grid cell is calculated by applying the grid-scale scaling factor (the ratio of a posteriori to a priori total emissions) to the corresponding a priori emission of that source type. Different sources can finally be calculated because each source category possesses distinct spatial patterns and seasonal variations*".

Question: Line 248: 7-14% higher when? Is this per year, or over the whole period? Clarify which experiment is higher (only the a priori is mentioned).

Answer: The 7-14% range represents the increase of a posteriori CO emissions relative to a priori emissions over the entire study period (2003–2022). Specifically, the three inversion experiments show the following differences compared to a priori: Column-FixOH: 14% higher than a priori; Profile-FixOH: 7% higher than a priori; and Column-VarOH: 10% higher than a priori.

The description in the manuscript has been revised: "*At the global scale, anthropogenic CO emissions based on three inversion configurations are estimated to be 7-14% higher than a priori values (Table 2) in 2003-2022*".

Question: Line 251: Global anthropogenic emissions?

Answer: Changed.

Question: Line 273: China or East China?

Answer: It is eastern China. Changed.

Question: Line 274: The growth period until 2007 is only clear in the GC-original line. Consider including a growth rate value to clarify.

Answer: The description for the first stage has been revised to "a slight growth until 2007".

Question: Line 300: India's growth rate does not appear to be significant in recent years. Include figures to demonstrate this point.

Answer: This sentence has been revised: "*India exhibited a continuous growth in anthropogenic CO emissions from 2003 to 2009, followed by a period of slower increase, with an average annual increase of 0.5-0.8 Tg yr^{-1} in 2003-2022*".

Question: Line 316: Add time period to the statement regarding a dramatic increase.

Answer: This paragraph has been extensively revised to highlight the differences in the seasonality and trends of wildfire emissions between the boreal North America and boreal Asia regions:

"*Emissions from high-latitude coniferous forests have shown different long-term trends between boreal North America and boreal Asia over the past two decades (Figs. 5a-b). Peak fire activity in boreal North America occurs during June-August (Fig. 6a); boreal Asia experiences its primary fire season in June-August, with a secondary peak often observed in March-May (Fig. 6b). When excluding the exceptional wildfire year of 2021, summertime biomass burning CO emissions in boreal North America exhibited an overall declining trend from 2003 to 2022 (Fig. 7a). In contrast, boreal Asia experienced a general increase in summertime biomass burning CO emissions during the same period, even when 2021 is omitted, though the trend is less pronounced than when including that extreme year (Fig. 7b)*".

Question: Line 319: What do the two regions discussed contribute to global biomass burning emissions in other years (i.e. is 2021 unique?)

Answer: As shown in new Fig. 7, when the year 2021 is excluded, no pronounced increasing trend in wildfire CO emissions is observed at both the Northern Hemispheric and global scales. This indicates that the extreme wildfire emissions in 2021 represent a distinct perturbation on wildfire trend.

Question: Line 322: Has global burned area decreased globally or just in these regions discussed?

Answer: In the revised version, we have adjusted the relevant statement. We cite Zheng et al. (2023) to explain the peak in wildfire emissions from high-latitude coniferous forests in 2021, emphasizing the climatic drivers (e.g., concurrent extreme droughts across the Northern Hemisphere) behind this anomaly. The discussion specifically mentioning burned area has been removed. This adjustment allows the discussion to remain more closely aligned with the core theme of emissions.

Reference:

Zheng, B., Ciais, P., Chevallier, F., Yang, H., Canadell, J. G., Chen, Y., van der Velde, I. R., Aben, I., Chuvieco, E., Davis, S. J., Deeter, M., Hong, C., Kong, Y., Li, H., Li, H., Lin, X., He, K., and Zhang, Q.: Record-high CO_2 emissions from boreal fires in 2021, Science, 379, 912-917, 10.1126/science.ade0805, 2023.

Question: Line 330: Explain how climate variability impacts the CO emissions in this case.

Answer: In the revised version, we have reorganized the discussion in this part, and the reference related to climate variability has been removed:

"*The trend shift in CO emissions are consistent with the sharp reductions in annual deforestation rates in the Brazilian Amazon from 25396 km^2 yr^{-1} in 2003 to 7000 km^2 yr^{-1} in 2010 (Deeter et al., 2018)*".

Reference:

Deeter, M. N., Martínez-Alonso, S., Andreae, M. O., and Schlager, H.: Satellite-Based Analysis of CO Seasonal and Interannual Variability Over the Amazon Basin, J. Geophys. Res.-Atmos., 123, 5641-5656, 10.1029/2018jd028425, 2018.

Question: Line 341: Figure 4 has total emissions not combustion emissions. I think this also includes the NMVOC related sources of CO and so Figs 4j-l should not be discussed simply as combustion.

Answer: Thank the reviewer for pointing out this issue! The citation of the figure has been changed to "Fig. 4" instead of "Figs 4j-l".

Question: Line 349: And industrialization in some regions.

Answer: Changed.

Question: Line 361: This section would benefit from some organizing/ structure. For example, a full description of the anthropogenic emissions and then biomass burning and then global rather than swapping between the two continuously.

Answer: Thank the reviewer for this important suggestion! The content of the original Section 3.3 ("Impacts of systematic errors on inferred CO emissions") has been moved to Section 3.4. The new Section 3.4 has been significantly condensed, with its length reduced by approximately 50% compared to the original Section 3.3.

Question: Line 407: Explain the compensation by CO emissions. Can it be determined what the VOC source of CO is doing in this comparison? Is there any compensation through this route?

Answer: This part has been expanded to include a discussion on OH and biogenic CO sources and sinks:

"*Variations in OH concentrations influence the oxidation of biogenic VOCs to CO and their subsequent chemical loss. These two counteracting processes establish a complex balance, ultimately reflected in the inverted estimates of biogenic CO sources. Specifically, the Column-VarOH inversion yields an average global biogenic CO sources of 391.4 Tg yr^{-1} in 2003-2022, approximately 3.9% lower than the 407.6 Tg yr^{-1} in Column-FixOH inversion*".

Question: Line 422: Add more detail about the optimized boundary conditions (sometimes they are discussed as land boundary and sometimes as ocean boundary). Is 50% a good approximation for both the Var and Fix OH experiments? Consider the use of this section if the results are so limited. It may be better placed as supplementary information or in the methods section rather than results.

Answer: We sincerely thank the reviewer for this thoughtful comment! We have reviewed the manuscript and ensured that in the revised version, all descriptions related to boundary conditions now accurately and consistently use the term "land boundary

conditions". This adjustment eliminates previous ambiguity between "land" and "ocean" boundary conditions.

The statement that "optimized boundary conditions can reduce the influence of OH-related uncertainty on CO emission inversions by about 50%" was drawn from our earlier regional inversion study (Jiang et al., 2015b), which focused on North America for 2004-2005. This number was a rough estimate rather than a precise quantitative result. We acknowledge that its applicability to the Var and Fix OH experiments in the present study is uncertain. Therefore, in the revised manuscript, we have removed discussion about this approximate estimate.

We have substantially condensed the section in question, reducing its length by approximately 50%. The content has been streamlined to focus only on the most directly relevant outcomes. Consequently, we believe it is now more appropriately retained in the Results section. This placement allows readers to follow the discussion of sensitivity experiments without checking discussions in the supplement.

Reference:

Jiang, Z., Jones, D. B. A., Worden, J., Worden, H. M., Henze, D. K., and Wang, Y. X.: Regional data assimilation of multi-spectral MOPITT observations of CO over North America, Atmos. Chem. Phys., 15, 6801-6814, 10.5194/acp-15-6801-2015, 2015b.

Question: Line 441: This section may be better placed before section 3.3. Alternatively, as suggested above, section 3.3 may be better placed with the supplement or methods section as the assumptions described mean results are difficult to interpret (e.g. due to the optimized land condition). I think section 3.3 is interesting and important to include, but the manuscript would benefit from a more coherent structure.

Answer: Thank the reviewer for this important suggestion! The content of the original Section 3.3 ("Impacts of systematic errors on inferred CO emissions") has been moved to Section 3.4. The new Section 3.4 has been significantly condensed, with its length reduced by approximately 50% compared to the original Section 3.3.

Question: Line 459: Describe the sensitivity experiments, perhaps in the methods section.

Answer: The following paragraph was added to provide a better definition for the sensitivity experiments:

"*To quantitatively attribute the concentration trends to specific drivers, we conducted a series of sensitivity experiments. The experimental design isolates the influence of individual emission sectors by building a baseline scenario in which all emissions are fixed at 2003 levels to reflect the impact of meteorological condition changes in 2003-2022. Three more sensitivity experiments were then conducted in 2003-2022 in which only one emission category, i.e., anthropogenic, biomass burning, or biogenic VOC emissions, was allowed to vary over time, respectively. The time-varying emissions in these sensitivity experiments were prescribed from the Column-FixOH a posteriori inversion*".

Question: Line 460: Reference Figure 9a).

Answer: Changed.

Question: Line 486: % sign missing.

Answer: Changed.

Question: Line 490: This is not concomitant as higher biomass burning emissions are not naturally associated with the lower anthropogenic emissions.

Answer: This paragraph has been extensively revised to highlight the impact of the extreme wildfires in 2021 on the trend of CO column concentrations:

"*This attribution analysis highlights the substantial impact of extreme wildfire years on the CO budget. Although anthropogenic emission reductions lowered Northern Hemisphere CO columns by approximately 0.51% yr^{-1}, the intense biomass burning emissions in 2021 introduced a positive perturbation of about 0.24% yr^{-1} in the full-record trend, thereby offsetting a considerable fraction of the anthropogenic-driven decline*".

Question: Line 510: Be clear about the direction of comparison for the bias (observations to model or model to observations). In these comparisons is this the column total difference? Below there is reference to the surface concentrations.

Answer: Thank the reviewer for raising this point! We have clarified in the text that the bias is calculated as model minus observation. The comparison here refers specifically to the model simulations against the aircraft-based observations, and thus the unit is ppb. The mention of surface concentrations earlier in the paragraph is provided as broader context, but the quantitative comparison with surface observations is not included in this section for brevity.

Question: Line 516: Is this total reduction in emissions (or burden?) or should this be annual?

Answer: The unit is Tg yr^{-1}. Changed.

Question: Line 520: Make clear any links between biomass burning emissions and the difference in climate over the period of the study.

Answer: Thank the reviewer for the valuable comment. We have reviewed and refined the usage of the term "climate" throughout the text. In the revised version, we have avoided using "climate" as a direct explanation for temporal changes or spatial patterns in biomass burning emissions. It is now used to describe the general influence of biomass burning on climate, or the influence of "climate modes" (e.g., ENSO events) on biomass burning activity.

Figures and Tables:

Question: 1: Colourbar is misleading, how can it go below zero? Suggestion to start the white at zero for a) b) c).

Answer: We have standardized the color bars across all figures and improved the visualization of data ranges (adjusting color bars to start at zero where appropriate) to ensure consistency and clarity.

Question: 4: Include the exclusion of certain grid-cells in the text and give a reason for this.

Answer: Biomass burning emissions exhibit strong seasonality, whereas anthropogenic emissions show relatively weaker seasonal variation. To prevent periods of intense biomass burning from obscuring the long-term trend of anthropogenic sources, we excluded months in which the contribution of biomass burning exceeded 50% at each grid cell when calculating trends for non-biomass burning emissions. This approach ensures that the derived trends more accurately reflect the actual changes in anthropogenic emissions, without being biased by short-term, seasonal biomass burning signals.

The Caption of Fig.4 has been revised to provide better clarification.

Question: 4: 6: Suggestion to not include data for the oceans if it is being ignored in the analysis. This data could be masked out.

Answer: We thank the reviewer for the thoughtful suggestion! In our revision, we have chosen to continue showing the oceanic OH distribution in order to maintain a globally consistent visualization, which aids in the overall interpretation of inter-dataset differences. However, to prevent confusion, we have explicitly noted in the caption that oceanic OH variations have negligible influence on the scaling factor differences, due to the land boundary conditions: "*Please note that due to the use of land boundary conditions, differences in OH concentrations over the ocean in the left column figures have a negligible effect on the differences in scaling factors shown in the right column figures*". We believe this addition addresses the underlying concern while preserving the informational value of the full global map.

Question: 7: Move titles to above the plots.

Answer: Changed.

Question: 8 and 9: These colour schemes look different to those used in previous figures. Ensure this is consistent and also perhaps one which has more differentiation in the negative end of the bar would be beneficial as the blues are quite hard to distinguish.

Answer: The color schemes for all figures have been adjusted to ensure a consistent presentation. The visualization of the dark blue regions (negative values) has been optimized.

Question: S1: Include a key for the colours used in e) and d)

Answer: Thank the reviewer for this suggestion! The color schemes of Fig. S1 have been standardized, and the visualization of data ranges has been improved by adjusting the color bars to start at zero. Furthermore, the following note has been added to the caption of Figure S1: "Since Eastern China, India, and Southeast Asia are geographically adjacent, different colors are used in Fig. S1e to distinguish these three regions".

Question: S4: Fix the titles and the detail on this figure is difficult to pick out. Suggest trying larger marker points or a more differential colour scheme to make the differences clearer.

Answer: Thank the reviewer for the valuable suggestion! We acknowledge that the details in the original figure were difficult to discern, and we appreciate your

recommendation to use larger marker points or a more differential color scheme for clarity.

We have carefully reconsidered the figure's purpose. This figure aimed to compare the differences in atmospheric CO concentrations between a priori simulations, a posteriori simulations, and aircraft observations. However, the visual distinction between a priori and a posteriori simulations is subtle. This is because our a priori simulation already incorporated optimized monthly CO initial conditions using sequential Kalman filter based on MOPITT satellite measurements, which reduced the model bias from -16.2 ppb (in the Original Simulation, GC-Original) to -3.4 ppb (in GC-a priori). While a posteriori emissions further reduced the bias to a range of -2.9 to -1.6 ppb, the change relative to a priori simulation is limited, making visual differentiation challenging as the reviewer pointed out.

Given that these quantitative bias values are already provided in Table 1, and to avoid any potential confusion, we have removed the original Figure S4 in the revised manuscript. We believe this approach maintains clarity while ensuring that the key findings are adequately presented through the tabulated data.