

In the text below we have included all the referee comments in black, followed by our response in red.

Anonymous Referee #1

A new greenhouse gas simulation in GEOS-Chem is presented, which couples CO₂, CH₄ and CO. The paper presents a detailed evaluation of the new system and compares the model simulations to observations.

It seems likely that this new framework will be useful for inverse modelling studies in the future. The analysis is very thorough, although perhaps some aspects could be cut or moved to the Supplement to help streamline the text (which is quite long). However, I have some concerns about the experiment design and interpretation. I feel that the paper would be suitable for publication in GMD after these concerns have been addressed.

General comments

Experiment design: The novel component of the paper is the description of the coupled CO₂-CO-CH₄ system. However, I found the discussion of the comparisons of the coupled and uncoupled systems very hard to follow in places. I think this is primarily because the various simulations were run with different OH fields and model resolutions (and with/without an OH diurnal cycle, although this doesn't appear to have much impact). To partially address this, an additional OH run is carried out to test the sensitivity to OH, introducing another model into the mix. It seems that the reason for these inconsistencies is historical, although I wasn't entirely sure from the text. It seems to me that this paper could be dramatically improved, streamlined and clarified by carrying out two sets of model runs, using the same OH field, the same resolution, one run coupled and one uncoupled. Given that it's relatively cheap to run these simulations, I'm not sure why this wasn't done. Can the authors explain? If it's to maintain traceability back to earlier GEOS-Chem simulations, I'd argue that this should be a secondary consideration, compared to showing the improvement due to the new simulation setup.

We thank the reviewer for the comments and discussion. This comment (and those from the other reviewers) has highlighted to us that perhaps the manuscript did not clearly describe the aim of our work, and this has led to some confusion about the intent of our work and therefore the experimental design. We have substantially revised the manuscript to better clarify both of these points. Here, we summarise our rationale along with changes we have made in the manuscript.

There was a strong reason for the experimental setup presented in the paper. The aim of the work was to compare the *existing* GEOS-Chem CH₄, CO, and CO₂ simulations that are currently being used by the community to the improved, coupled simulation we have developed. The existing simulations have multiple inconsistencies – in addition to the chemistry being uncoupled, they rely on inputs created at different points in time from different versions of the full chemistry model (including different OH, but also other differences in the chemical schemes). Our goals were to 1) present the problems associated with the existing uncoupled simulations and the way they are currently being used in the GEOS-Chem community and 2) present the improvements introduced by the coupled simulation. We now clarify this explicitly in the revised manuscript.

What we wanted to demonstrate is that if one uses the uncoupled GEOS-Chem CH₄, CO, and CO₂ simulations out-of-the-box, they are not consistent, introducing unnecessary errors when using these simultaneously for a single dataset, inversion, etc. If we were to change to consistent OH fields between the uncoupled (i.e., running consistent full chemistry simulations to produce the

offline chemical fields) and coupled simulation, this would disguise the key limitations and biases in the chemical production/loss in the uncoupled simulations.

Another factor that we did not make entirely clear is that when we referred to the OH being from a particular model version, what this really means is that the chemical production fields were calculated using that model version. The OH distributions are a major contributor to the difference in chemical production between versions, but there will also be additional differences embedded in the chemical production fields (e.g., different emissions). To summarize our experimental setup: the uncoupled simulations use archived chemical production fields created in separate full-chemistry simulations based on what was available at the time each individual offline simulation was most recently updated (CO (v9) and CO₂ (v8)). Recreating all the chemical production fields with the same OH distribution would require re-running the full chemistry simulations for the entire period, which would be significantly more computationally expensive than the offline simulations. Again this would be of limited value, as we would effectively be creating a false narrative: comparing our new coupled simulation to individual CH₄, CO, and CO₂ simulations that do not exist and are not being used by the community.

In the new coupled simulation, we are calculating (online) the chemical production of CO (from loss of CH₄) and CO₂ (from loss of CO) using consistent (v9) OH fields and comparing these to the out-of-the-box uncoupled CO and CO₂ simulations to demonstrate the inconsistencies and biases if the GEOS-Chem user runs the default out-of-the-box uncoupled simulations. One thing that comparison does not highlight is inconsistencies between the out-of-the-box CH₄ and CO simulations. The out-of-the-box uncoupled CH₄ simulation uses v5 OH, while the out-of-the-box uncoupled CO simulations uses P(CO)_{CH₄} based on a full chemistry simulation with v9 OH. This means that in the uncoupled simulations, L(CH₄) and P(CO)_{CH₄} are *not* equivalent (although, in reality, they should be). The reason we added the additional (orig-OH) coupled simulation was to demonstrate the impact of this discrepancy, by determining what the coupled simulation would look like if we calculated P(CO)_{CH₄} (and follow-on parameters) from the same L(CH₄) as in the out-of-the-box CH₄ simulation. We understand that this is a fairly subtle point, and that its inclusion alongside the coupled and uncoupled simulations may have hampered the readability of the manuscript.

To improve the paper, we have made a number of changes in the revised manuscript. In addition to clarifying the above points throughout, we have also made the following substantial changes:

1. We focus the analysis on comparison of the uncoupled and coupled simulations only, removing the orig-OH simulation from the main comparison discussion and figures. Instead, we have added a short subsection with discussion about the orig-OH simulation using a 1-year simulation to more clearly make the point outlined above. By focusing on the coupled and uncoupled simulation results, we now clearly show the value of our improved simulation relative to the out-of-the-box simulations.
2. To simplify the analysis, we now focus the results on CO and CO₂ (and their chemical production terms) and remove parts of the CH₄ analysis.

Comparison to observations: The conclusions state that the new model improves the fit to the observations (L488 – 490). However, I don't think that this can be concluded. Since we do not know the "true" flux magnitude for these gases, we can't be sure that the coupled simulations are really improving the fit to the data, or just compensating for some bias(es) in the flux fields. For example, for methane, it is stated (L446) "The coupled-origOH results show a positive bias with overestimated CH₄ values for all sites; however, using globally more abundant OH fields (v9-01-03) in the coupled simulation resolve this large bias". However, it could well be that the new fields are simply compensating for some bias in the (highly uncertain) emission field.

We agree with the reviewer. In the revised manuscript, we rephrase and correct these statements and highlight that reduced bias in the coupled simulation could be compensating for biases in the emissions.

Specific comments:

L107 (Eq. 2): I don't think this equation works. For closure, I think there also needs to be a term representing the net flux from/to the troposphere.

We thank the reviewer for alerting us to the fact that we did not adequately describe this equation (or indeed the other equivalent equations) in the manuscript. We now explicitly state in the revised manuscript that these equations only describe the changes in the emission, deposition, production and loss terms that occur within each grid box and that advective transport fluxes between grid boxes (including between the troposphere and the stratosphere) are in addition to the terms described in each equation.

L113: [OH] has already been defined.

[OH] definition removed.

L129 (Eq. 7): Again, need flux to/from the troposphere.

See above; we now clarify this for all relevant equations.

Figure 3: Is this figure relevant?

Figure 3 highlights well the spatial differences in OH between the coupled and uncoupled simulation. Differences in the OH spatial distribution have a direct impact on the spatial distribution and analysis/understanding of the CO and CO₂ chemical fields. However, due to the reduction of the orig-OH simulation analysis we move both Figure 2 and 3 to the Supplement.

L226 – 227: Some of this inter-annual variability is present in the uncoupled simulation. Is the change really so marked in the coupled version?

We certainly observe stronger inter-annual variability in the coupled simulation, as well as changes in the chemical fields that are not present in our uncoupled simulation. In response to comments from Reviewer #3, we have clarified this discussion to explain the source of the inter-annual variability in the uncoupled simulation.

L231 and Figure 5 caption: The figure shows the CO production, not the "Changes of the CO production". The change can be inferred from the figure, but is not directly shown.

We have removed 'changes' from both L231 and Figure 5 caption.

Figure 4: Add an x-axis. Also, it's not clear why a bar chart is the best way to present this. How about a line graph?

We have added the years as the x-axis (and axis label 'Years'). Figure 4 shows the global summary of the different region budgets presented in the Supplement (Fig S3-S6) that are difficult to show as a line graph. Hence for consistency we used the same plotting procedure (i.e., bar graph with the same colouring). We now state in the figure caption "*Regional distributions are shown in Figs. S3-S6 in the Supplement*" to better make the link between this figure and the ones in the Supplement.

L244: "Hemispheric" seems preferable to "regional" to describe the table.

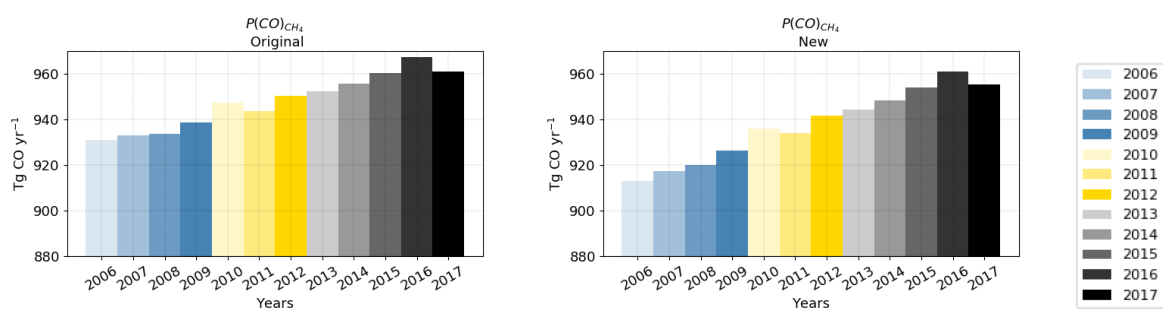
'regional' changed to 'hemispheric' in both the text and Table 2 caption.

Figure 7: Perhaps move this, and the discussion, to the Supplement. I'm not sure it adds too much. **In the revised manuscript, both Figure 7 and the related discussion have been removed from the main text.**

L340: I think a comma would be preferable to a semi-colon between "seasons" and "but". **Changed to a comma.**

L431: The offset in the modelled values will lead to a small difference in the CO production, etc., compared to the real atmosphere (i.e. CO production from CH₄ will be over-estimated, since the model spin up leads to higher CH₄). Is this effect important?

We thank the reviewer for highlighting this. The higher initial CH₄ levels (due to the spin up) in the coupled simulation will indeed lead to stronger CO production after reaction with OH. This was an error in our original work that we have now corrected. To correct this, we re-ran the coupled and uncoupled CO₂ simulations with all the offsets (in both the CH₄ and CO₂ initial fields) corrected prior to the simulations (instead of the post-simulation correction we used previously). The figure below shows the original results (with the offset present in the chemical term) and the new corrected result. Using the corrected restart files leads to lower P(CO)_{CH₄} values; however, the overall analysis in the paper remains the same, since the coupled results still show higher values relative to the uncoupled CO simulation and follow the inter-annual variability as in the original analysis. In the revised manuscript we have updated all the results to use the corrected simulation and we modified the description of the offset correction.



L449: "resolves", rather than "resolve" (although, see general comment... I don't agree with this statement!)

We have rephrased this statement, as described in our response in the general comment (comparison to observation).

L464 / 465: Notwithstanding the issues around flux magnitudes, these differences seem very small compared to all the other uncertainties in the system. I think it'd also be fair to say that the model changes had a negligible impact on the comparison with the observations here.

We have rephrased/removed this based on our response above to the general comment (comparison to observations).

L488 – 491: I think these lines in particular (and many others throughout) need to be revised in light of my general comment regarding the potential impact of uncertain fluxes.

As above, we have rephrased this statement.

L593: I don't think you can say that the v5-07-08 fields are incorrect based on this analysis, given the emissions uncertainty.

In this specific case, we have robust and strong evidence that the v5-07-08 fields are incorrect. The chemical scheme used in GEOS-Chem v5-07-08 (circa 2004) was much simpler than modern chemical schemes and is now extremely outdated. For example, in v5-07-08 the model

underestimated lightning NO_x emissions, and thus ozone, in the northern extratropics. Further, there is clear evidence that the v5-07-08 OH fields are incorrect since they produce an unrealistic seasonal cycle in CO_{CH4} that is corrected when using newer OH fields. We now explain this in the new dedicated sub-section on the orig-OH simulation.

L607 – 609: As above, I think this conclusion needs to be removed.

As above, we have rephrased this statement.

Appendix A: Titled as “Appendix A: Appendix A”

Fixed.