

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

Anonymous Referee #3

General Comments

The authors present coupled greenhouse gas simulations with GEOS-Chem focusing on CO₂/CH₄/CO and its global chemical interactions. Such simulations will be definitively needed towards a consistent description of long-term atmospheric chemistry and for realistic assessment of climatic change by earth system models. I fully acknowledge the work done here and I am convinced that the new developments implemented in GEOS-Chem are a big step towards these goals. When reading the abstract, I got the impression that the paper follows a clear outline by first comparing coupled and uncoupled simulations and then evaluate the new model version with observational data. The authors present a sound and informative introduction to the scientific and computational problem of consistent chemistry simulations, which comprise processes representing a broad range of timescales as well as trends and interannual variability. I also liked the detailed budget term quantification as presented in Table 2. Unfortunately, I got lost after different versions of the OH input fields were introduced. For me it remained unclear why the authors are not able to stick to a single OH field which then can be used for both, the uncoupled and the coupled simulations. In the discussion part of the manuscript, the introduction of a third simulation (coupled-origOH) lead to unnecessary confusion and an overload with information details which made it hard for me to extract the major conclusions. Overall, I would not recommend to publish the paper in its current form but I encourage the authors to submit a revised manuscript based on more consistent coupled and uncoupled simulations.

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 why the OH fields were different between simulations, as well as for introducing the coupled-origOH simulation. 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. What we wanted to demonstrate in the paper is that if one uses the uncoupled GEOS-Chem CH₄, CO, and CO₂ simulations out-of-the-box (which is how the GEOS-Chem community is using them), the chemical production fields (calculated using GEOS-Chem full chemistry simulations) are not consistent as they were created at different times using different versions of the full chemistry (and therefore different OH). Because the archived chemical fields used by the out-of-the-box uncoupled simulations were calculated with different OH fields, the resulting CH₄, CO, and CO₂ distributions were inconsistent with one another. The coupled simulation resolves these inconsistencies, which are partly driven by the lack of coupling and partly by the different existing chemical production fields. If we were to change to consistent OH fields between the uncoupled and coupled simulations, this would disguise the key limitations and biases in the chemical production/loss in the uncoupled simulations. It would also 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.

One thing that the comparison between the uncoupled (out-of-the-box) and coupled simulations 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 simulation 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 for introducing the coupled-origOH 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 has 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.

We would kindly ask Reviewer #3 to also visit our detailed reply to Reviewer #1's general comment about the experimental setup for further explanation of these points. We anticipate that the improvements we introduce in the revised manuscript will much better highlight and clarify the aim of our work and the reasoning behind our experimental design.

Specific Comments

Line 49: What do you mean by "outside source regions"?

"outside source regions" was referring to areas/regions that are not dominated by strong anthropogenic point emissions. We clarify this in the revised manuscript.

Lines 49-56: For CO budget terms you can also refer to Stein et al. (2014).

We thank the reviewer for suggesting the additional reference, which we have added to the revised manuscript (both the text and Table 2).

Line 51: Publication year is missing.

Fixed.

Lines 52-54: It would be interesting to see also the numbers for the chemical production by NMVOCs (used as input for your simulations).

We now state the P(NMVOC) numbers in the revised manuscript. On lines 52-54 we list known literature values for this chemical production with references (320–820 Tg CO yr⁻¹, Holloway et al., 2000; Bergamaschi et al., 2000; Arellano Jr. and Hess, 2006; Duncan et al., 2007; Zeng et al., 2015; Fisher et al., 2017) while in Section 2.2 (coupled simulation description) we present the numbers used as input in our simulations (480 Tg CO yr⁻¹).

Lines 60-63: Can you give a reference here?

We have added references in the revised manuscript that highlight differences in the chemical

fields: Nassar et al., (2010), Fisher et al., (2017), Duncan et al., (2007), Suntharalingam et al., (2005), Zeng et al., (2015).

Lines 82-84: Is the spin-up time sufficient? You doubt this later on (Lines 506-508).

Our spin up period totals to 11 years, and we used this time period based on the recommendation from the GEOS-Chem team. The recommended spin up period for CO₂ and CH₄ is 10 years (http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_restart_files). We clarify this in the revised manuscript, and have removed the comment on lines 507-508 referring to insufficient spin-up time.

Lines 96-97: I would expect that a single reference full chemistry simulation is used for all simulations presented here.

Please refer to our response to the general comment above. In addition, we now clarify in Section 2.1 that the uncoupled simulations run here are the out-of-the-box versions available in GEOS-Chem v12.1.1, which were developed independently at different points in time and therefore use chemical production fields that come from different full chemistry simulations. We better highlight in the revised text that these discrepancies between full-chemistry versions are a major source of bias (and limitation) in the uncoupled CH₄, CO and CO₂ simulations; and that this significant limitation is now removed by introducing the coupled simulation.

Line 134: How does GEOS-Chem handle Biomass burning emissions? It is known that such emissions need to be emitted throughout the troposphere following a vertical profile.

As our intention is to understand the impact of the coupling (including consistent production and loss fields) relative to the out-of-the-box uncoupled simulation, we treat emissions from biomass burning identically between the coupled and uncoupled simulation, using the treatment found in the out-of-the-box uncoupled simulations. However, for CO₂ we made a small modification by updating the simulation to use QFEDv2 emissions that were not available in the out-of-the-box uncoupled CO₂ simulation. We implemented this following the method used in the CH₄ simulation and we did this to have consistent biomass burning emission types across all three species. We now describe the biomass burning emissions treatment in section 2.2. Briefly: for all three gases we use daily QFEDv2 emissions (Table S1, Supplement) with additional diurnal scale factors. For CO we use vertical partitioning of the emissions where 35% of the biomass burning emissions are emitted above the Planetary Boundary Layer (again, following the default setup in the uncoupled CO simulation). For CO₂ and CH₄, the emissions are emitted at the surface only and transported to higher altitudes via mixing. We acknowledge that the CH₄ and CO₂ emissions treatment should be improved to match that for CO; however, for this work we aimed to keep all aspects of the simulations not associated with the chemical coupling as consistent as possible with the default version of the uncoupled simulations.

Line 295: Exchange “tropospheric column” by “mid troposphere”.

Fixed; however, this part of the discussion will be moved into the Supplement based on the comments from Reviewer #1.

Lines 348-349: I would expect to have the same P(NMVOC) for all model runs. Give numbers!

P(NMVOC) is consistent in the simulations where it is used as an input field: uncoupled CO and coupled simulation. In these simulations the P(NMVOC) is based on v9 full chemistry simulations and additional offline processing as described in Fisher et al., (2017). However, in the uncoupled CO₂ simulation, the P(CO₂) chemical fields were archived by Nassar et al., (2010) using the v8 full chemistry simulation (with differences in chemical scheme that would implicitly include differences in P(NMVOC)). Intermediate terms, including P(NMVOC), were not archived, limiting us from specifying the numbers in the revised manuscript.

Table 1: Publication year is missing.

Fixed.

Figure 1: I like this figure. It could even improve if you orient your coupled and uncoupled flows from left to right.

Figure modified as suggested.

Figure 4: From your description I would expect that $P(\text{CO})_{\text{CH}_4}$ (for all years) and $P(\text{CO}_2)$ (for 2010-2017) remains exactly constant for the uncoupled runs (except for leap years).

That is correct. The chemical production input fields have no inter-annual variability in the case of CO and after year 2010 for CO₂; however, Figure 4 does show a change. The reason for this is: 1) as the reviewer noted leap years during 2008, 2012 and 2016 that will lead to higher production (since the figure shows total production summed over the year) and 2) interannual variability in the meteorological fields (e.g., pressure levels, tropopause height) affecting the calculation of the total tropospheric budget. We clarify this in the revised manuscript.

Figure 7(b): This reads like “Surface Loss” as parameter.

Fixed, however, this Figure will be moved into the Supplement based on comments from Reviewer #1.

References:

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