

We thank the referee for the useful comments. In what follows, we have included the original referee comments in black, followed by our response in blue, with new text shown in **bold**.

Anonymous Referee #2

The manuscript 'Improved method for linear carbon monoxide simulation and source attribution in atmospheric chemistry models illustrated using GEOS-Chem v9' by Fisher et al. describes an update of the linear CO method which is available in the GEOS-Chem model. Details about the technical implementation (usage of CO production rates from a prior model run instead of yield rates for NMVOC emissions), as well as an comparison of the 'old' and 'new' version with observation data are given. In addition an example of the new source attribution capabilities of the method is presented. As the paper describes an improvement of a current model it is in general well suited for GMD, before publication, however, major revisions are necessary.

General comments:

The manuscript is in general well written, but especially the description of the model details lacks in clarity. I would suggest to condense the evaluation (more details below) and to describe the model updates as well as the general performance of the model with respect to changes of the CO production rates in more detail.

The authors seem to describe three main updates. The first updates are the improvements described at the end of the Sect. 2.2. These changes are applied in both presented model simulations. Accordingly, the term 'original CO-only simulation' is somewhat misleading as these model results are different from the 'CO-only' method which is the 'status quo' prior to the changes described by the manuscript.

We have clarified the text in Section 2.2 to explain that these updates are effectively bug fixes and that these are part of the current standard ('status quo') GEOS-Chem CO-only simulation (v11-01). The only non-standard change described in this section is the OH fields, discussed below. We are sensitive to the concern that the term "original" is misleading, and have replaced in with "base" throughout the manuscript (except in the abstract, where we feel it would be confusing without additional context).

The paragraph in question now reads:

P5, lines 25-32: "**We also fixed a few minor problems in the v9-01-03 CO-only simulation to bring it up-to-date with the current public version (v11-01) and to make it more compatible with the full chemistry simulation. Specifically, we implemented optional non-local mixing in the planetary boundary layer (Lin and McElroy, 2010) and the centralized chemistry timestep (http://wiki.seas.harvard.edu/geos-chem/index.php/Centralized_chemistry_time_step) – both of which are defaults in the v9-01-03 standard full chemistry simulation but were missing from the CO-only simulation. We also added diurnal scaling of the monthly mean [OH] fields based on the cosine of the solar zenith angle. This is the same method used in all other offline simulations in GEOS-Chem, and is an objective improvement compared to using a single monthly mean value in each grid box. All of these updates are included in both the base and improved versions compared below.**"

The main update are the improvements described in Sect. 2.2.2. The description of these updates seems insufficient to me. For readers which want to implement this method in their own model implementation details are missing. For people of the GEOS-Chem community which might be interested in using the method a user manual or similar (in the supplement) is missing. In addition not all simplifications/assumptions are discussed in detail (see below for details).

We have addressed all the specific comments and suggestions below, and hope that this now covers all previously missing implementation details. We have also expanded the Section 7 on Code Availability to more clearly explain how users in the GEOS-Chem community can apply the changes:

P21, lines 14-19: “The standard GEOS-Chem code is freely accessible to the public by following the guidelines at <http://wiki.geos-chem.org/>. Updates described here will be included in the standard code once this paper has been accepted, likely after version 11-02. In the interim, the version 9-01-03 code used here is available at https://github.com/jennyfisher/GEOS-Chem_TaggedCO_v9.01.03_updated. **The standard GEOS-Chem code is distributed as a git repository, and the code updates described here can be added to a standard v9-01-03 code repository by pulling the branch linked above.** The provisional version of v11-01 where we have implemented these updates is available by contacting the authors.”

We have also added a link to the user manual, which provides all details for compiling and running GEOS-Chem (none of which have changed in this version):

P21, lines 19-20: “**Instructions for running the GEOS-Chem model, including the CO-only simulation, are provided in the GEOS-Chem User’s Guide:** <http://acmg.seas.harvard.edu/geos/doc/man/>.”

In general I would recommend to give proper version numbers to the different updates to make the model changes clearly traceable (e.g. 1.0 original Version, 1.1 updates described in Sect 2.2 etc.).

Because the GEOS-Chem model itself (of which the CO-only simulation is one part) already has version numbers, we feel it would be confusing to replace the names with a version. In the absence of short names that could clearly and succinctly encapsulate the differences between the two versions of the simulation, we prefer to retain simulation names “base” and “improved”, which we use consistently throughout the text and figures.

To track the model development it would also be very interesting to show the difference of the model results between ‘version 1.0’ and ‘version 1.1’.

As described above, most of the minor changes described in Section 2.2 were essentially bug fixes (already made in the public version 11-01), and we have updated the text to clarify this. The only non-standard change described in this section is the OH fields. As described below, we have added an extra figure to the Supplement (new Fig. S1) to show the magnitude of the changes due to OH (in essence, comparing ‘v1.0’ and ‘v1.1’).

Further I am missing a discussion about how the inter-annual variability of the production and loss rates saved from the full chemistry model influence the results of the 'improved CO only' simulation. Especially as the author state that results from one simulation year are sufficient for the improved model. For interested users it would be very important to see, how critical these pre-calculated production and loss rates are. If dynamics and pre-calculated production rates stem from different years they might not fit to each other and errors/biases are introduced. Please discuss these simplifications. Especially when thinking about sensitivity studies with changed biogenic emissions (e.g. to check the influence of additional source) a new calculation of the secondary CO production (with the full model) seems crucial to me.

This is an important point. We have added to the Supplement a section (new Section S1) and a figure (new Fig. S2) to explain that the interannual variability in the improved CO-only simulation is dominated by meteorology and emissions, with only a very minor contribution from the pre-calculated chemical production rates. We agree with the referee that some specific applications (e.g., evaluating differences in biogenic emissions) would require the user to re-calculate the secondary production fields, and this is why we originally noted in the text that an interested user could easily run the standard chemistry simulation as required. We now explicitly provide examples of when this step may be necessary. The updated text now reads:

P6, lines 23-29: “For many applications, use of a single model year (e.g., that used in the 1-year benchmarking process) will be sufficient—as **discussed in the Supplement, interannual variability in simulated CO is dominated by variability in the meteorology and primary emissions with only a very minor contribution from variability in the CO production rates (Sect. S1, Fig. S2)**. However, the code is designed in such a way that an interested user could easily re-run the standard chemistry simulation to save the CO production fields for a specific year of interest. **This extra step would generally only be necessary for changes likely to significantly impact chemical production (for example, different biogenic emissions or meteorological fields).**”

We would also note that in the past, the CO-only simulation has been applied to a broad range of problems (see for example references in the introduction), and for many of these the small errors introduced by using a single year for the chemical production will not be important.

In this context it would also be interesting to show how large the changes with the new OH fields instead of the OH field used in previous version of the CO-only simulation are.

We have re-written this paragraph to add detail, and have added an extra figure to the Supplement (new Fig. S1) to show the magnitude of the changes due to OH:

P5, lines 15-21: “Previously, the CO-only simulation used OH from earlier versions of the model, **typically v5-07-08** (e.g., Kopacz et al., 2010; Fisher et al., 2010; Jiang et al., 2011, **2017**), to mitigate a known OH high bias in more recent versions of GEOS-Chem. Here, we use OH from v9-01-03 in both our base and improved CO-only simulations to maintain consistency with the full chemistry model and to ensure all changes are due to the new representation of chemical production rather than differences in OH. **Global annual mean OH is about 5% larger in v9-01-03 ($11.4 \cdot 10^5$ molecules cm^{-3} ; http://wiki.seas.harvard.edu/geos-chem/index.php/Mean_OH_concentration) than in the default v5-07-08 ($10.8 \cdot 10^5$ molecules**

cm^{-3} ; Evans and Jacob, 2005; Kopacz et al., 2010). As a result, CO is systematically lower when using the updated OH fields, as shown in Fig. S1.”

One problematic thing about the described update is that the amount of needed simulations are doubled. What is the benefit of using first the full model and then the CO-only model? In this context a discussion about the influence of the used CO production rates is important to see if results for different years are dominated by meteorological variability or by variability of the CO production (which of course is also influenced by the meteorological conditions). Especially if users are interest in one specific year it would be interesting to add the source attribution capabilities to the full chemistry model. The source attribution can then be calculated directly during the full model run and the second run (CO only) would not be necessary.

As described in the comment above, we now explicitly show that the interannual variability in simulated CO is dominated by meteorology and emissions, with only a very minor contribution from secondary CO production. This means that many users who are not specifically focusing on e.g. biogenic emissions or biogenic source regions will still be able to run only the CO-only simulation. Relative to the full chemistry simulation, the CO-only simulation is 6 times faster and requires only ~40% as much memory, so there are significant advantages to the GEOS-Chem community to maintaining a CO-only simulation (e.g., for use in forecasting, source attribution, interpreting multi-model ensembles, etc.). This is particularly useful for the four-dimensional variational data assimilation scheme in GEOS-Chem, which can require 20-40 iterations of the model simulation when inverse modeling CO emissions. We agree that adding CO source attribution capabilities to the full chemistry model would also be a useful future addition (that could be maintained in parallel to the CO-only simulation) – but that would require additional code development that is outside the scope of this work.

The comparison of the 'original CO' and 'improved CO' to the results of GEOS-Chem using the full chemistry clearly show the improvements of the new version. The authors might discuss shortly that the good agreement between the full chemistry and the 'improved CO only' runs can be expected, as same dynamics, chemical tendencies, emissions and OH-field from the full model run are used by the 'improved CO only'.

We now clarify that the good agreement between the full chemistry and improved simulations is expected (see also response to Referee #1):

P9, lines 2-3: **“The improved agreement is expected, as the spatial distribution of CO production is now the same in the two simulations.”**

Given the small difference between all three model simulations compared to the observations presented in this Section it can be condensed a lot. Of course, a comparison to observation is important. As the differences between all versions are much smaller as the difference between observations and model most of the figures might be moved to the Supplement and the text of this Section can be condensed to the most important findings.

We believe it is important to give the readers a thorough (and visual) understanding of the

comparison to observations. From Figs. 1-3, one would expect to see a large change in the model-observation agreement between the old and new simulations. The fact that this isn't the case (shown in Fig. 4) is an important result from this work – showing that the GEOS-Chem benchmark data are not sufficiently representative to constrain current model uncertainties. The aircraft are useful for showing the magnitude of the change in regions with enhanced biogenic emissions that are not captured in the surface data.

We feel this section will be of interest to some readers and is already condensed, with only 4 paragraphs of text and all but two figures in the Supplement. We therefore prefer to leave Section 4 in place.

Specific comments:

Sect. 2.1: Please provide a table with the different performed simulations.
We now include a table with the simulation details (P4, new Table 1).

Instead of 'improved CO only' etc. I would recommend more handy names of the simulations. We have not been able to come up with a better succinct name that clearly communicates the updates made to this simulation, but we would be happy to consider any specific suggestions.

In general this Section might be moved to the end of Section 2, meaning that first model improvements and at the end the model set-up are discussed.
We prefer to retain the order as-is as it allows the text to flow directly from the description of the new simulation to the implications of the changes. We feel this flow would be lost by moving Section 2.1 to the end of Section 2.

Sect. 2.2.2: Please clarify which production rates are written out from the full chemistry model run. From my understanding only the total CO production and the methane loss rates are written out, from which the production of CO from NMVOC are then calculated off-line.

We now clarify which rates are saved from the full chemistry model and that the methane and NMVOC production are calculated offline.

P6, lines 30-31: “The total $P(\text{CO})$ saved from the full chemistry model is split **offline** into contributions from methane ($P(\text{CO})_{\text{CH}_4}$) and from NMVOCs ($P(\text{CO})_{\text{NMVOC}}$) by also saving the methane loss rates ($L(\text{CH}_4)$) from the full chemistry simulation.”

Meaning you solve the eqn:

$$d\text{CO}/dt = E + P\text{CO}_{\text{CH}_4} + P\text{CO}_{\text{NMHC}} - k[\text{OH}][\text{CO}] \quad (1)$$

If so, please clearly state the equation you are solving in the description of the improved model. We have also added a new Equation (2) that shows the equation solved in the improved model.

P7, line 9: $d[\text{CO}]/dt = E + P(\text{CO})_{\text{CH}_4} + P(\text{CO})_{\text{NMVOC}} - k[\text{OH}][\text{CO}] \quad (2)$

Figure 1: The figure could be improved a lot by using different color scales for the surface and 500 hPa.

Fixed – we now use a color scale of -50 to 50 ppb for surface CO and -10 to 10 ppb for 500 hPa CO.

Please indicate also the differences in percent in the text.

We now also state the differences in percent in the text everywhere differences are quantified.

Section 4: Why are you only evaluating values for the year 2009, while the years 2010 and 2011 are also simulated. Is the difference to the observations in these years similar or different from the values presented for 2009?

As we state at the start of Section 4, results from the three different years are indistinguishable in terms of model-observation comparisons. See P9, lines 7-8: “As in Sect. 3, we show model output from 2009, which is virtually identical to output from our other simulation years (2010 and 2011).”

The used color coding of Fig. 4, 5 and 6 is problematic. Especially the difference between the blue and the purple color is very hard to see.

Fixed – see response to Referee #1 – blue has been changed to orange.

P10L1ff: In my opinion it is very problematic to refer to additional graphs (and software) which are available somewhere on the internet. In some years these URLs might not be valid any more. If the graph/software are relevant for the publication then they should be part of the electronic supplement (or get a DOI in any other way).

We were hesitant to make the Supplement unreadably long (each yearly benchmark is 41 pages), and so chose to post the extra benchmark plots at a large online publishing company unlikely to disappear. Nonetheless, we are sensitive to the referee’s concern and will include all 3 benchmarks in the Supplement of the revised version.

Figure 4: Please indicate the years for which the observations are averaged. Again, I don’t understand why the observations are averaged over many years but the model results not.

Fixed – see response to Referee #1. Figure 4 now shows 2009 results for both model and observations.

Figure 6: The text and symbols are rather small.

We have now magnified the figure as much as can be done in the GMDD latex template (making the figure any larger within the template causes it and all subsequent figures to appear after the references). We expect a larger version of the figure to appear in the final version.

Are the model results sampled at the same time as the observations or are ‘simply’ daily average values of the model used when observations are available.

The model results are daily mean values for days when observations are available, as we now state in the figure caption:

P16, Fig. 6 caption: “Simulated **daily mean** CO from the base CO-only (red) and improved CO-only (purple) has been sampled from the relevant grid boxes only for days with available observations...”

Figure 7: Please clarify - Anthropogenic and biomass burning does only consider primary emissions of CO, right? All production of CO by NMVOC (anthropogenic, biomass burning and

biogen) are part of the NMVOC oxidation. If so, please clarify the sentence line P16L9ff.

As explained in Section 2.2.1 and now re-iterated here, in the base model secondary production from anthropogenic and biomass burning NMVOC was included as part of the primary source.

We now repeat this information here:

Pg 17, lines 9-11: “In the base simulation, these were included with the primary emissions **as they were calculated by increasing primary anthropogenic and biomass burning emissions by 19% and 11%, respectively (Sect. 2.2.1); whereas in the improved simulation**, they are part of the NMVOC contribution.”

P19L16ff: Please clarify this paragraph. Do you mean that the improved model should perform better as the full chemistry model or are you discussing the small difference between the original and the improved model?

We have reorganised this text to explicitly state that the differences between the simulations are smaller than the observation-model mismatch:

P20, lines 22-24: “Our comparisons showed that at the benchmark sites, **the differences between the base and improved CO-only simulations** are in most cases significantly smaller than the observation-model mismatch, despite the large differences between **the** simulations seen elsewhere.”

Technical comments:

P5L6: Please give a proper reference to the used JPL version.

Added.

Figure 4 Title: This figure does not show vertical profiles.

Fixed