

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 #1**

The manuscript implemented a major improvement to the representation of secondary CO production in the CO-only simulation. The improved CO-only simulation resolves the discrepancy between the full chemistry and CO-only simulations in both the magnitude and spatial distribution. It also includes a new capability of source-region tagging of secondary CO produced from oxidation of non-methane volatile organic compounds. These two improvements help us get a better understanding on CO sources and transport over the remote regions. In general, I found the main points and the structure of this manuscripts are clear. But some of discussions are weak and unclear. Below are my comments for making the manuscript more concise. I recommended the paper to be published with revision.

Main comments:

#### Section 4: global evaluation with observations

I understand that the observational data used in this paper is the same data used to benchmark new version of the full GEOS-Chem model. However, it is not proper of using observed climatology to justify model improvements in reference to the original simulation and the standard chemistry simulation. The observed climatology is normally used to do an initial sanity check of simulated mean and seasonality. The authors argue that Figure 4 does not help illustrating the improvements of the new scheme because of the larger observation-model mismatch. To me, the failure of figure 4 is also partly because we are referencing orange to apple. The results would be convincing if the authors compare monthly mean of simulated CO in 2009 to that of GMD surface CO over the same year. What is the measurement frequency for GMD surface CO? The authors should also plot the continuous monthly mean of model output as well as the mean calculated from dates with GMD measurements.

The comparison would probably not change significantly using the observed monthly mean from 2009, but at least that gives us a quantity evaluation of CO simulation in GEOS-Chem full chemistry run and CO-only run.

We appreciate the suggestion. In digging through the standard benchmark dataset, we realised that the surface CO data included were not multi-year means as expected but rather monthly means for 2005 only. We now follow the reviewer's suggestion and compare the simulation to the observed data from the same year (e.g., 2009 in Fig. 4). The agreement does improve, although the message remains the same: the observation-model mismatch is larger than the differences between the three simulations. Due to different data availability in 2009, we replaced two sites shown in the original Fig. 4 (Bermuda and Palmer) with nearby sites (Key Biscayne and Syowa, respectively).

Figure 4 now shows the 2009 monthly means for the model. We only saved the global model output at monthly time resolution, so we are unable to also plot the mean calculated only from dates with GMD measurements.

We have also used this opportunity to update the CO data used by the GEOS-Chem Support Team for the standard full chemistry benchmarking, which uses model output from 2013.

Please modify Figure 4 as follows:

1) The title should be surface CO instead of CO vertical profiles.

Fixed.

2) The lon and lat of Crozet should be: (-46, 52). Please double check whether this is just a typo in the caption, or extracted from the wrong grid box (-46, -52).

Fixed. This was in fact extracted from the wrong gridbox – and appears to have been for all earlier versions of the GEOS-Chem benchmark as well. This has now been fixed in Figure 4 and reported to the GEOS-Chem Support Team to fix in the public benchmark as well.

3) It is hard to distinguish the blue and purple lines. please use another color, dark green?

It is a challenge to find colors that are distinguishable both in general and for a red-green colorblind reader. We have replaced the solid blue line with solid orange in Figs. 4 & 5. While the orange and red lines may look similar to some colorblind readers, the dashing will help distinguish these, and the lines are now more easily distinguished for the non-colorblind reader.

4) Use the monthly mean and standard deviation of GMD surface CO data for 2009 as discussed above.

Fixed as discussed above.

In addition, I suggest the authors use specific names (instead of original or improved CO-only simulation) for these two versions of CO-only simulation.

In response to a comment from Referee #2, we changed the name “original” to “base”. We use the terms “base” and “improved” to avoid any ambiguity in the figures and text. 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 “base” and “improved”, which we use consistently throughout the text and figures. We would be happy to consider any specific suggestions for clearer version names. To further improve the clarity of these names for the readers, we now include information about the differences between the two in the figure captions for Fig. 1 (first model-model comparison figure) and Fig. 4 (first model-observation comparison figure):

**“The main difference between the two CO-only simulations is the vertical distribution of the CO source from NMVOCs, which is 3-dimensional in the improved simulation but surface-only in the base simulation.”**

Detailed comments:

P4 Line 3: We use the GEOS Chem CTM version 9-01-03 as the based version.

Fixed: “We use the GEOS-Chem CTM version 9-01-03 as the base version...”

P4 Line 6: Goddard Earth Observing System, Version 5

Fixed: “Goddard Earth Observing System, **version 5** (GEOS-5).”

P6 Line 11: It is not surprise to see the total CO in improved CO-only simulation improves significantly when comparing to the full chemistry CO simulation, since the  $P(\text{CO})$  are same in these two runs.

Yes precisely – the goal is to quantify how large the discrepancy was in the original (standard) simulation. We have added the following to the text when we first discuss the improved agreement with the full chemistry simulation:

P8 line 33-P9 line 5: “While some differences remain between the improved CO-only and full chemistry simulations, these are much smaller (less than 6 ppbv / 5% everywhere, and less than 3 ppbv / 3% outside the African plume) and show a greatly diminished spatial extent. **The improved agreement is expected, as the spatial distribution of CO production is now the same in the two simulations.** Lingering differences likely reflect use of scaled monthly mean OH concentrations and  $P(\text{CO})$  fields in the CO-only simulation, which may introduce some offset between the timing and locations of CO production and CO loss.”

P7, Line 19: China and India also show large overestimate in the surface in the original CO-only run. What causes that?

Although it’s not visible on the color scale used in the figure, these overestimates are smaller. They come from anthropogenic and/or biomass burning VOCs. We now add this to the text:

P8, lines 9-14: “At the surface, the base CO-only model greatly overestimates CO compared to the full chemistry simulation over the continents, with differences of more than 100 ppbv (60-80%) in major biogenic VOC source regions (e.g., Amazon, central Africa, Indonesia, and the Southeast US). **Similar, smaller (up to 50 ppbv / 10-15%) effects are present in regions with elevated VOC emissions from anthropogenic and biomass burning sources (e.g., China, Alaska). These overestimates** reflect the assumed instantaneous CO production from VOCs in the surface layer in the base model, whereas in the full chemistry and updated models this production happens more gradually.”

P7, Line 23: For the remote region: dose CO from NMVOC represent 1) CO produced locally through NMVOC transported from source region or 2) transported CO from source region which was produced there or 3) a combination of them?

We are unsure what text the referee was referring to, as P7, line 23 does not discuss CO from NMVOC. We now clarify this issue earlier, after Equation 5:

P7, lines 28-30: “**In other words,  $CO_{NMVOC,j}$  from region  $j$  represents the CO that was produced within the boundaries of region  $j$ , regardless of the origin of the NMVOC (i.e., local vs. transported). As most precursor NMVOCs have short atmospheric lifetimes, most  $CO_{NMVOC,j}$  will derive from NMVOCs emitted in region  $j$ .**”

P7 line 31: similar overestimates seen over Tibetan Plateau, probably related to the deep convection storms in summer time

We have added this region to the discussion. In the improved version of the figure, the overestimate appears centred over southern China. Precursors in this region are likely to include anthropogenic emissions, and so we have modified the accompanying text accordingly:

P8 lines 19-21: “In a few regions (central Amazon, Indonesia, **and southern China**), the base

CO-only simulation actually overestimates the full chemistry simulation at 500 hPa by 7-8 ppb (10-12%). These are regions where **large NMVOC sources** are coupled with frequent deep convective activity.”

P11, line 2: largest differences between two CO-only simulations

Fixed: “The largest differences **between the two CO-only simulations** are seen...”

P11 Lin32: Not sure if it is necessary to discuss the comparison with IASI here.

We disagree – while we found that the impact was small, one of the motivations for the work was the possibility that the original treatment of secondary CO would degrade comparisons with satellite instruments that are most sensitive in the mid-troposphere (such as IASI). We prefer to include this text as it may be relevant to other readers who have the same concern.

P12 Figure 4 caption: specify the year range of multi-year monthly mean

See earlier comments – we now instead show the 2009 data and specify this in the caption.

P15 Figure 6: For overlapping period, the light purple and dark purple circles look same to me.

Fixed

P16 Line 7-8: Can you be more specific on the NMVOCs setting in these two version runs? I understand how CO from NMVOCs emitted by biomass burning or anthropogenic emission is in the NMVOCs contribution in the new scheme. But I am confused with the setting of these sources in the old scheme.

This is explained in Section 2.2.1, and we now also re-iterate it 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.**”

P18 Figure 8: adding labels for the locations of three stations in Figure 7.

Fixed