

Response to Referee 1 (Referee's comments in blue, response in black)

This paper analyses CO simulations by a chemistry climate model in order to better understand possible reasons for reported low-biases in CO, especially on the NH. To this end, a rather complicated set-up is presented in which a kind of ad-hoc inverse modeling approach is combined with ad-hoc data-assimilation. Nevertheless, some nice insight is gained in the possible causes for model-data mismatches. I have two major comments, which I outline below. I think the authors should address these issues carefully.

We thank the Referee for the constructive comments. We have incorporated these comments into our revised manuscript, as described below.

1. Major comments:

Structure of the paper.

The structure is rather messy. I think the introduction, method, and results should be better separated to improve readability. For instance, section 2.2.4 clearly presents results, but is presented in the method section. Section 3.4 presents a section that starts with "The primary source of OH" that I would classify as "Introduction".

We reorganized some sections of the paper to improve readability. In particular, we moved the comparison of simulated OH fields from section 2.2.4 to the Results section. We moved some of the background information on OH from section 3.4 to the Introduction.

Section 3.3 suddenly ends with something about TCO, which seems more appropriate in section 3.4.1.

We discuss TCO in section 3.3 because this section discusses the impact of CO emissions on ozone. Section 3.4.1 discusses the impact of ozone on OH, so TCO is discussed again there.

The method description now centers around several "model options", which makes it hard to detect the strategy behind the paper.

We added the following paragraph to section 2.2 describing how we will alter CO emissions, OH, transport in a series of sensitivity studies, prior to our descriptions of the model options:

"We use a series of sensitivity studies to analyze the role of CO emissions, OH concentrations, and transport. Two methods are used to examine the sensitivity of CO concentrations to CO emissions from different sources: scaling up the CO emissions, and scaling up CO tracers tagged by source. We quantify the sensitivity of CO to OH concentrations by applying scaling factors to the OH field. We analyze the sensitivity to transport by comparing a free running CCM simulation with a

simulation that has prescribed meteorology. Several different chemistry options within the GEOSCCM framework are used to isolate specific processes.”

Also, the forward referencing to the sensitivity simulations (page 20311: “We conduct a sensitivity simulation, described in section 3.2”) makes the paper very sketchy. I would rather prefer to start with table 1 and slowly describe all the simulations in sequential order. The problem is, I guess, that the sensitivity simulations follow from the first results. In that respect, it would also be an option to follow that line and first present and discuss the reference simulations, motivate the sensitivity simulations, and then present those. Anyhow, the authors should improve the readability by severely reshuffling the paper.

We removed the forward referencing of the sensitivity studies from the Methods section. We follow the reviewer’s second option of first presenting results of the reference simulations and using these results to motivate the sensitivity studies. In particular, we combined section 3.2.3 with section 3.3 and use the results of section 3.2 to motivate the sensitivity study described in section 3.3. We state, “The tagged tracer results presented in Section 3.2.2 suggest that increasing CO emissions can improve the agreement with CO surface observations. However, increasing CO emissions will lead to feedbacks on OH, CH₄, and ozone that are not captured by the COonly chemistry option. We therefore conduct a sensitivity simulation with the GMI chemistry option called GMI-HiEmis that is identical to the RefGMI simulation except for an increase in CO emissions. “

Unclear choice of presenting the results

One might question the rather ad-hoc way in which the sensitivity simulations are set up. As the authors know, there are more formal “inverse modeling” methods to optimize emissions and OH to improve the match between model and observations that are based on the minimization of a cost function. I agree that the selected ad-hoc inversion is an attractive alternative. However, what I find disturbing is the poor definition of the “cost function”.

Our study is designed as a series of sensitivity studies conducted using a forward model rather than as an inverse modeling or data assimilation study. Consequently, we do not apply a cost function and instead treat the observations as “truth” to evaluate our reference and sensitivity simulations against. It is not our intention to optimize CO emissions, but rather to “investigate how attributing a CCM’s CO bias to CO emissions versus OH chemistry impacts ozone and methane lifetime”, as we now state in the introduction. We also added the following text in section 3.3 to clarify this: “The purpose of this experiment is not to calculate the optimum CO emissions to reproduce the CO observations, but rather to determine how a reasonable set of CO emission adjustments impacts the simulated concentrations of ozone and OH as well as CO.”

The sensitivity studies described in Section 3.4 are designed to address specific hypotheses about the causes of OH asymmetry and bias. We realize that OH could

be adjusted through formal data assimilation. However, we believe that our approach of using existing reanalysis and assimilation products one at a time as inputs to our model provides an easily interpreted framework for isolating the effects of specific model biases on the CO-OH-CH₄ system. In this respect, our work complements the existing inversion and data assimilation studies in the literature.

Figure 3 (section 3.2.1) focuses on the latitudinal GMD CO observations between March and August, and for spring and summer individually. Why is the rest of the season left out? Figure 4 goes more quantitative, but now uses March through May (?). Figure 5 focuses on June through August, but again the rest of the year is left out from the analysis. It remains also unclear when the emission and OH changes are applied. Is this during the whole simulation (including Jan-March) or only in the months under investigation? This latter question is linked to section 3.2.3 in which emissions are changed in each season (table 3 (and not 2)).

We now explain in Section 3.2 that: “We focus on spring since it is the season with the largest bias in NH OH (Fig. 2), and contrast the spring results with those from summer, since the bias persists into summer despite seasonal differences in transport and chemistry.” We now clarify that this is the focus in the introduction as well, stating “Our focus is primarily on NH spring and summer, when the NH CO bias is large.”, and in section 3.2. We also clarify in section 3.2.1 that OH is reduced throughout the year. We apply the OH reduction over the whole year because the reduction is based on the methane and methyl chloroform lifetime results of Naik et al (2013), which are for the whole year. Similarly, we consider the whole year in our analyses in Section 3.4 and in Tables 2 and 4 since we are comparing methane lifetime to year-round observation-based estimates. We now state in Section 3.4: “We analyze the results for the entire year in order to compare our results to observation-based estimates of methane lifetime.”

This analysis now focuses on April 2007 (why?), and shows strange results in table 3. The zero increments in Sep-Dec are intriguing, as are the exact same increments that are required for Asian / EU emissions and Russian BB. These results look rather suspect to me. It becomes even more complicated when MOPITT results are further considered (section 3.2.4) when it turns out that some selected adjustments are incompatible with MOPITT (again focusing on April 2007 only).

We updated figures 6 and 7 to show the multi-year average of March-May to make the comparison to MOPITT more consistent with the time period discussed elsewhere. We did not adjust the emissions for September through December since the RefGMI simulation shows relatively little bias in Northern Hemisphere CO during these months, and our focus is on Spring and Summer. Since our emission adjustments are based on only two constraints (the global mean bias and the interhemispheric gradient), we do not attempt to independently adjust all the different sources. Instead, we apply the same adjustment to Asia anthropogenic, European anthropogenic, and Russian biomass burning emissions. We now clarify this in section 3.3:

“We do not alter the September through December emissions since the RefGMI simulation shows little NH bias in those months (Fig. 2) and our focus is on spring and summer. We choose the adjustment factors by solving for the linear combination of CO_{aa}+CO_{ea}+CO_{rub} and CO_{trbb} that minimizes the error in both the IHG and the global mean bias. Table 3 shows the emissions adjustments for each season. We apply the same adjustment to CO_{aa}, CO_{ea}, and CO_{rub} since these three sources show nearly the same slope (IHG bias / model bias) in Figures 4 and 5.”

We also combined the Russian biomass burning line with the European and Asian anthropogenic line in Table 3 to avoid confusion.

In summary, I can live with the fact that no formal inverse method is applied, but the lack of a clear cost function, target period, and optimization strategy makes the results non-robust and confusing. The authors therefore should work out a clear strategy and clear goals (is the focus on spring and summer biases only?). Having said this, I think the results that I see are quite interesting and provide a valuable contribution, once the strategy is better worked out.

As described above, we have clarified that our focus is on spring and summer, and that the goal is to examine the sensitivity of simulated methane lifetime and ozone to different assumptions about the cause of CO bias.

2. Minor issues:

The title does not seem to cover the contents of the paper. The paper is only partly about the methane lifetime. Maybe: “Analysis of the carbon monoxide bias in chemistry climate models”.

We changed the title to “Implications of Carbon Monoxide Bias for Methane Lifetime and Atmospheric Composition in Chemistry Climate Models”

Page 20306, line 16: CO only contributes to O₃ formation at sufficiently high NO_x concentrations. Please add the role of NO_x.

We added “where sufficient NO_x is present”

Page 20307, line 25: “consistency of CO emission estimates with surface and satellite observations”: this now sounds as if emission estimates are measured at the surface and by satellites. Please make clear that there is a model in between.

We reworded this sentence to “Previous studies used models to examine the consistency of CO emission estimates with surface and satellite observations of CO concentration.”

Page 20311, line 17: What are these co-emitted NMHCs? It sounds rather misty that on page 20312, line 15 these co-emitted emissions are reduced. Is this a kind of fudge factor? Are these anthropogenic NMHCs or natural? Please clarify.

Both biomass burning and anthropogenic sources that emit CO typically emit Non-Methane Hydrocarbons (NMHCs) as well, and these NMHCs eventually oxidize to form CO. Since NMHCs are not explicitly modeled in the COonly chemistry option, we account for their effect on CO through the inclusion of a CO amplification factor. A detailed description of this process is provided in Duncan et al. (2007a), which we cite here and discuss in the introduction. The choice of CO amplification factor is chemical mechanism-dependent, since it is intended to account for processes that are left out of the chemical mechanism, which is why it differs between the CO-OH and COonly chemistry options. We now clarify that these are from both anthropogenic and biomass burning sources.

Page 20313, line 25: “during” NH spring.

Done.

Page 20316, line 28: table 3.

Done

Page 20317, line 1: and b shows → and b show

Done

Page 20318, section 3.2.5: I find these results interesting. Has a reason been identified for the different transport? Which transport patterns have been identified as the best? Presumably vertical transport differs in MERRA, and this is an assimilation product, which is consistent with observations, hence better? Could this important observation be mentioned more clearly, e.g. in the abstract?

We now mention in the abstract that the use of specified meteorology improves the comparison with surface observations: “We find that the use of specified meteorology alters the distribution of CO compared to a free-running CCM simulation, improving the comparison with surface observations in summer”. We expect the horizontal winds to be better in MERRA, which will influence tracer distributions. Vertical transport is not constrained by observations in MERRA, so it is not clear which is best in that regard. This is an interesting subject for future work, but is beyond the scope of the current study.

Page 20320, line 22: The increased OH → The decreased OH Page 20323, line 11 (and section 3.4.4 in general): Are there observational indications (e.g. from satellite data) that NO_x emissions are too high (a known bias)? Also, reducing the NO_x concentrations by 30% will affect not only the OH recycling (HO₂ + NO), but also the O₃ production. Do I understand correctly that these effects are ignored here, because O₃ columns are also prescribed in the RefCO-OH simulations? Please mention this clearly. Do you proportionally adjust NO and NO₂?

This sensitivity study is not based on observational evidence of NO_x bias, but rather on the known sensitivity of OH to NO_x concentrations and the hemispheric asymmetry in NO_x emissions. The reviewer is correct that the impact of changing NO_x on O₃ production is ignored here, since the O₃ field is prescribed. We now mention this in Section 3.4.4: “We note that in this experiment, ozone values remain the same as in RefCO-OH, rather than responding to the change in NO_x.” We also clarified that NO and NO₂ are both reduced by 30%.

Page 20325, line 14: [Linked to the above: “reducing the uncertainties in NO_x emissions”](#): this would call for a similar approach using e.g. NO₂ column observations from OMI/GOME-2, like Miyasaki, 2012.

We added the following sentence in Section 3.4.4: “Previous studies have estimated NO_x emissions based on satellite observations of NO₂ columns (Lamsal et al., 2011; Miyazaki et al., 2012a), but some uncertainty in NO_x emissions remains.”