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

Anonymous Referee #2

Review of "An improved carbon greenhouse gas simulation in GEOS-Chem version 12.1.1" by Beata Bukosa et al.

This paper simulated CO2, and CO2 production by oxidation of CH4 and CO using 3 different types of OH fields. The topic of research is important for better estimation of CO2 sources and sinks on the Earth's surface. The distribution of OH is heavily concentrated over the tropical region, where CO2 would be added to the atmospheric CO2 due to oxidation of CO and CH4. If ignored this CO2 chemical production, biased source of CO2 is needed from the tropical land and ocean regions by inverse modelling. I very much liked the idea of this research, but unfortunately wasn't able to read through the whole manuscript due to poor execution of the research idea, in my opinion. Thus I cannot recommend publication of this work in Geoscientific Model Development in the present form or anything close to this. It is better to rerun the model and submit a newly prepared manuscript.

We believe the reviewer has misunderstood both the aim of our research and the experimental design and execution. The reviewer's misunderstanding (and comments 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. We have substantially revised the manuscript to better clarify both the intent and the experimental design of our work. Here we summarise these points before addressing the reviewer's specific comments. (See also our response to Reviewer #1 for a more detailed description.)

The aim of our work was to compare the *existing* GEOS-Chem CH₄, CO, and CO₂ simulations (which are uncoupled) that are currently being used by the community to a new version of the simulation that couples these species together based on their chemistry. 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 the way they are currently being used in the GEOS-Chem community and 2) present the improvements introduced by the coupled simulation. The intent was not to focus on the differences in OH, and we now clarify this in the revised manuscript.

We also note that the original (uncoupled) simulations do not ignore CO₂ chemical production as implied by the reviewer. Rather, the CO₂ production in the uncoupled CO₂ simulation is inconsistent with the CO chemical loss in the uncoupled CO simulation (and the same is true for CO production from CH₄). It is this inconsistency our work aims to rectify.

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 CO$_2$ (and their chemical production terms) and remove parts of the CH$_4$ analysis.

Here are some of my major concerns:

Table 1: For eaxmple, "Coupled only" : I do not understand - are all CO are produced from CH4 oxidation ? If so you are going to underestimate CO. If not, is CO in L(CO) and P(CO)ch4 are different entities, then there is a good chance of double counting

Table 1 contains 3 sections:

- **"Fields used by both uncoupled and coupled simulations"**
- **"Uncoupled only" – which we will change to "Fields used by uncoupled simulation only"**
- **"Coupled only" – which we will change to "Fields used by coupled simulation only"**

The first section includes P(CO)$_{NMVOC}$, which is the other source of CO chemical production (CO produced from non-methane volatile organic compounds). We expect that by changing the table headings as described in the bullet points above it will become clearer that CH$_4$ oxidation is not the only source of CO chemical production.

Further, as stated in the caption and in line 87, Table 1 only provides the terms that impact the chemical production and loss fields (and vary between simulations). As stated in lines 87-88, the full list of all the flux terms (i.e., other emission or uptake fields) used by each simulation is presented in Table S1 in the Supplement (as these do not vary between the different simulations). In summary, all simulations include CO chemical production from CH$_4$, CO chemical production from NMVOCs, and direct CO emissions; hence there is no CO underestimation (excluding potential biases in the external emission fields). This is also shown in Equation (4), which shows that the simulations include CO emissions and total P(CO) chemical production, composed of the CH$_4$ and NMVOC terms as shown in Equation (5).

L(CO) and P(CO)$_{CH4}$ are entirely different terms. P(CO)$_{CH4}$ is defined in equation 5 (line 123) and represents the amount of CO produced when CH$_4$ reacts with OH (assuming a 100% CO yield from this reaction as explained on line 122). L(CO) represents the amount of CO lost via reaction with OH in the troposphere and from archived CO loss in the stratosphere. The L(CO) term is calculated at every model time step after all the emission and production fields (including P(CO)$_{CH4}$) are added to CO, hence there is no double counting. The calculated L(CO) (both troposphere and stratosphere) is then used to calculate the chemical production of CO$_2$. This simulation description is provided in detail in lines 110-141.

Formulation of Eq. 1 & 2 (also for CO): Not correct !!, I think Eq. 1 and Eq. 2 are not separable in a chemistry-transport model, except for "tagging". Please clarify or rectify errors

These equations are correct as written but were not adequately described in the text in the original manuscript. We now explicitly state in the revised manuscript that these equations only describe the changes in 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.

Note that the GEOS-Chem model dynamically calculates the tropopause height at every timestep, and uses this information (in all GEOS-Chem simulations) to assign each grid box to either the troposphere or the stratosphere. This happens before applying the appropriate source and sink terms as described in Equations (1) and (2) (and also for CO). We now clarify this in the main text.

Figure 2: Quite large differences in OH. Acceptable? May be you should run CH3CCl3 tracer of checking your OH.

We agree with the reviewer that there are large differences in OH values – which is one of the inconsistencies in the uncoupled simulations (currently used by the GEOS-Chem community) that our new coupled simulation is designed to eliminate.

To clarify, these different versions of the OH fields come from different historical full-chemistry simulations, which advanced from v5 (circa 2004) to v9 (circa 2013) due to developments and improvements to the full chemistry model. We try to highlight these differences and improve the simulation of all carbon gases by introducing the coupled simulation. Currently the GEOS-Chem community is using inconsistent OH fields across the uncoupled CH_4, CO and CO_2 simulations that will introduce biases in the modelled values. Introducing the new coupled simulation with consistent and more recently updated OH fields (as well the possibility to easily update the OH fields using future full chemistry model versions) eliminates these large differences. In our revised manuscript, we have removed the orig-OH simulation from the main discussion and reduced the discussion of the different OH fields. Where we do discuss them, we more clearly state that the v9 fields are the most up-to-date of the 3 fields used by the existing, out-of-the-box carbon gas simulations.

Although understanding and exploring the biases in the OH fields is an important task in atmospheric chemistry it is not the focus of our paper. The OH analysis in the paper is presented only for a better understanding of the changes in the chemical fields, and as such comparison with CH_3CCl_3 is beyond the scope of this work.

Figure 5 (left column): the P(CO)ch4 and P(CO2) are apparently not consistent with the OH fields in Figure 2.

If the reviewer is suggesting that changes between OH fields (Figure 2, strongest OH in v8 and weakest in v5) should be mirrored in the $P(CO)_{CH4}$ and $P(CO_2)$ chemical production fields, that is not correct.

While the differences in OH *contribute* to the differences between the simulations, they are not the only factor driving the variability in chemical production. For $P(CO)_{CH4}$, this term represents production of CO from CH_4 reaction with OH (equation 5) and therefore depends not only on [OH] but also on $[CH_4]$, which differs between the uncoupled and coupled simulations. Lower OH values will not necessarily lead to a weaker chemical production: if the CH_4 amounts in the full chemistry simulation (used by the uncoupled simulation) are higher than the CH_4 initial fields in the coupled simulation, this can lead to overall stronger $P(CO)_{CH4}$ even if the OH levels are lower. We discuss these additional terms in detail in Section S1 in the Supplement. We now explain this relationship in the text when we discuss Figure 5. In addition, we have added text to Section 2 (model description) to describe the different methods used to calculate $[CH_4]$ between the full chemistry (used for uncoupled) and coupled simulations.

For $P(CO_2)$, the situation is similar (the comparison between the uncoupled and coupled simulations will depend on both [OH] and [CO]). Here, however, we note that we only expect a very small change in $P(CO_2)$ between the coupled and coupled-origOH simulations. This is because, as shown in Figure 1, the ONLY difference between these simulations is the OH field that is used to calculate $P(CO)_{CH4}$. The latter represents only a fraction of the total CO source (which includes $P(CO)_{NMVOC}$ and direct CO emissions), and so this will translate to a smaller impact on $L(CO)=P(CO_2)$, consistent with the results shown in Figure 5. To state this another way, the v5 OH (turquoise) shown in Figure 2 is NOT used to calculate $L(CO)=P(CO_2)$ in the coupled-origOH simulation.

We recognise that this latter point is confusing and distracts from the main messages of our work. As stated previously, we have removed the coupled-origOH from the main body of the text, which we expect to make the comparison between the coupled and uncoupled simulations easier to understand. In addition, in the new subsection where we describe the coupled-origOH simulation, we more clearly explain that the v5 OH is ONLY used to calculate $L(CH_4) = P(CO)_{CH4}$ and not $L(CO) = P(CO_2)$.

Finally, we note that Figure 5 shows the total production summed over the full troposphere, while Figure 2 shows [OH] for specific model levels. We now state this explicitly in the caption to Figure 5.

In summary, we see no inconsistencies between Figures 2 and 5, and we expect that the changes outlined above will make this much clearer for readers. However, if we misunderstood the reviewer's comment we would appreciate further clarification as to what specifically is 'not consistent'.

This where I had to stop going forward or read the text carefully. I am extremely sorry, but this has to be solved first before interpreting the results.
As outlined above, there is no inconsistency to resolve. The revised manuscript has been reduced in complexity to make our aims, methods and results clearer.

You have about 20% higher OH for the red and purple lines, compared to blue, both at the surface and at 500 mb when averaged over a year (Fig. 2).
But here in Fig. 5, we find the blue line is close to purple than the red line for P(CO2), and also I cannot explain the relative values of P(CO)ch4 as expected from the OH fields.
Please refer to the detailed comments above. In particular, for $P(CO_2)$ the blue (coupled-origOH) and purple (coupled) lines SHOULD be closer to one another than to the red (uncoupled) line because they use the same OH for calculating $L(CO) = P(CO_2)$, and the only difference will come from the fraction of [CO] that originally derives from $P(CO)_{CH4}$.

As described above, we have made substantial modifications to the revised manuscript to make it easier to understand these subtleties, including removing the coupled-origOH simulation from the main text, better explaining these relationships where appropriate, and clarifying the differences in spatial scale between Figures 2 and 5.

I understand that the OH level is affecting the concentrations of CO and CH4 and then you get very mixed pictures for P(CO) or P(CO2). But these are not realistic, because we only have one state of CO and CH4 concentration distributions (strictly).
We agree with the reviewer that in the real atmosphere there is only one state of CO, CH_4, CO_2 and OH. Indeed, this was the main rationale for creating the coupled simulation, as in the uncoupled version there was a disconnect between these species that is not realistic. Our coupled simulation removes the disconnect, using only one consistent version of the OH and chemically linking all three carbon species distributions to one another.

If you are checking the effect of OH then design experiments accordingly, and so on. Please consider.
Please refer to our comments above. The focus of the paper is not to understand the effect of OH. We only presented the OH analysis to better explain the changes in the chemical production fields. As described in detail above, the modifications in the revised manuscript better streamline the aim and results of the paper.