Response to Reviewer #2

We would like to thank the reviewer for the helpful comments on the manuscript. It helped a lot to improve our manuscript and to increase the readability. Please find below our replies in italics and indented.

1 General Comments

This paper presents an improvement to an existing and already implemented scheme, describing in detail the "tagging" of OH, HO_2, and H, by source sector. This is certainly a useful exercise. However, I found this paper confusing, as well as containing numerous errors. In its current form I am unable to recommend its publication in GMD.

> *Based on the reviewer's comments, we thoroughly revised and restructured the manuscript. We agree with the reviewer that the case "tag" as baseline is better suited and we adapted the manuscript accordingly. In addition, we modified the presented figures and put the contour levels on a common scale, as suggested by the reviewer. We think that the method and results are now represented in a better way.*

2 Specific Comments

I found the description of the tagging (Section 3.4) very confusing - the terms "explicit tagging" and "specific tagging" are used, and seem to mean different things. Longer-lived species are also tagged by source region, but the paper does not make clear the difference in this tagging and the explicit or specific tagging mentioned. It is clear that this process is complicated and requires careful consideration, but it is not explained in a way that I could easily understand. Perhaps some sort of graphical description would be helpful here?

> *Obviously the text was misleading. There is no explicit or specific tagging. There is only one tagging method which we are using. This tagging method along with different assumptions based on the lifetime of the regarded species leads to different implementations. We changed the corresponding wordings to clarify this.*
> *We added further explanations on how the tagging of the short-lived and long-lived species influence each other in the introduction. As suggested by the reviewers, we also inserted a sketch explaining the interaction between long-lived and short-lived species (Fig. 1). The implementation of the long-lived tagging is explained in detail in Grewe et al. (2017).*
>
> ***Changes in manuscript:***
> *The contributions to long-lived and short-lived species are closely linked. For example, the reaction*
> *$OH + O_3 \rightarrow HO_2 + O_2$*
> *involves the long-lived species O_3 and the short-lived species OH and HO_2. Hence, this reaction is considered in the implementation of the tagging method for long-lived and short-lived species. The contribution of, for example, shipping emissions to O_3 influences the contribution of shipping emissions to HO_2: the higher the contribution to O_3 is the more HO_2 is attributed to shipping emissions. Furthermore, OH from shipping emissions destroys O_3 and thus reduces the contribution of shipping emissions to O_3.*

When extending the tagging scheme to include more reactions (listed in Table 1 of the paper), reactions 19 (H_2O_2+hv->2OH), 28 (HOCl+hv->OH+Cl), and 30 (HOBr+hv->

OH+Br) are highlighted as being unable to be considered in the tagging scheme. However, the authors then include these reactions in Table 2 ("reduced - V1.1 all") and also in the line plots in Figure 1. They seem to make-up around 9% of the OH production rate, so I can see why they should be mentioned, but I was frustrated that they were given prominence over the "reduced - V1.1 tag" scheme, which is what was actually implemented in the model. Indeed, in Table 2 the OH loss and HO_2 production and loss rates are given alongside the "all" row and not the "tag", which I personally do not think is correct. I would see the "tag" scheme presented as the baseline, and the "all" is an extension to this. There is discussion in Section 3.3 about how good the "all" scheme is, but given it can't be used, why discuss it at all in this context?

> *Thank you for this recommendation. We changed the manuscript and set the case "tag", which is finally implemented in EMAC, as baseline of the manuscript. The moved the explanation about omitting certain reactions in the appendix. We hope the manuscript gained more readability.*

I was also confused about the rest terms introduced in Section 3.5. I appreciated that closing the budget is desirable, but I do not believe that the text in Section 3.5 justifies or explains their introduction sufficiently, and they seem very artificial. Can the authors please expand on this justification and the necessity for having these terms?

> *Thank you for this comment. We agree that the justification was not comprehensive. The steady-state assumption is the basic principle of the tagging method for short-lived species. As we consider a reduced HO_x reaction system, the steady-state between production and loss is not fulfilled. To re-establish steady-state, we introduce the rest terms.*
> *We restructured the Sections "Steady-state assumption" and "Closure of the budget" in the manuscript and merged them together. We also added the above explanation to better justify the rest terms.*
>
> ***Changes in manuscript:***
> *Thus, the state-state for the reduced HO_x and H reaction system (Tables 1 and 2) is not completely fulfilled.*
> *But steady-state between production and loss is crucial for the tagging method for short-lived species. To re-establish steady-state, it would be necessary to include the complete HO_x and H chemistry in the tagging method. However, this is not possible as the tagging method does not apply to all reactions of the HO_x and H chemistry (for examples see Appendix A). Consequently, we introduce rest terms resOH, resHO2 and resH for OH, HO_2 and H to compensate for the deviations from steady-state.*

Significant work is required by the authors to refine and clarify the manuscript. I suggest much more proof reading and editing are necessary prior to any resubmission.

> *We thoroughly edited the manuscript based on the reviewer comments. We hope that it now better suits the reviewer's expectations.*

3 Technical Corrections

1. I personally did not like the authors stating the species chemical formula after the name, without using either parentheses or parenthetical commas, e.g.
   The radicals hydroxyl OH and hydroperoxyl HO_2 are crucial for the atmospheric chemistry.
   rather than
   The radicals hydroxyl (OH) and hydroperoxyl (HO_2) are crucial for the atmospheric

chemistry.
or
The radicals hydroxyl, OH, and hydroperoxyl, HO_2, are crucial for the atmospheric chemistry.
This first format is used throughout the document (including the abstract). I would advise the authors to correct this to one of the others.

Thank you. We changed the notation to parentheses.

2. Page 1, line 16: remove "the" before "atmospheric chemistry".

Done.

3. Page 3, line 5: remove "the" before HO_x.

Done.

4. Page 4, line 6: could the authors please explain what a "cataster" is?

We changed the word to inventory.

5. Page 4, lines 20-21: I would suggest either "The mechanism in V1.0" or "The V1.0 mechanism".

We changed the wording.

6. Page 5, lines 6-7: I don't quite understand what the authors mean by "Each reaction occurring in a simulation was precisely added up" in the context of the paragraph. Could the authors please re-phrase this?

We have reformulated the corresponding sentences.

Changes in manuscript:
Most reaction rates used in the tagging method corresponds to the production and loss rates directly provided by the chemical scheme MECCA of EMAC.

7. Page 5, line 16: I would not use the phrase "boil down". I would suggest using "reduce" instead.

We changed the word.

8. There is discussion in Section 3.2 about the relative contributions of various reactions to the OH and HO_2 budgets. It might be helpful to also visualise this, perhaps using bar- or pie-charts, perhaps in the Supplementary Information?

Thank you for this hint. We added the amounts of the relative contributions of the mentioned reactions to the text.

Changes in manuscript:
The reactions which are important in the troposphere are indicated in Table 1. As stated above, reaction (1) of H and O_2 dominates the HO_2 production in the troposphere. It produces 49 % of tropospheric HO_2. In V1.0, only part of this HO_2 source was regarded (see Sect. 3.1). The most important HO_2 loss is the reaction with NO (reaction 14) followed by the reaction with itself producing H_2O_2 (reaction 3) which accounts for 32 % and 12 % of tropospheric HO_2 loss. The production via H2O and $O(^1D)$ produces about

21 % of tropospheric OH (reaction 2). The excited oxygen radical ($O(^1D)$) originates from the photolysis of O_3. Also reaction (14) of NO and HO_2 produces 32 % of tropospheric OH. OH is mostly destroyed by CO (reaction 11, 38 %) followed by NMHC (reaction 21, 25 %).
In the stratosphere different chemical reactions become important. Here, OH is mainly destroyed by O_3, producing 40 % of stratospheric HO_2. The reaction is partly counteracted by the reaction (14) which produces 21 % of OH and destroys 24 % of HO_2. Since large quantities of O3 are found in the stratosphere, O_3 or the excited oxygen radical ($O(^3P)$) destroys about 62 % of HO_2. Reactions with NMHC, CO and CH_4 play only a minor role in the stratosphere.

9. Page 13 equations 17, 18, 23, and Page 14 equation 24: Why does the term resOH=n appear in both equation 17 and 23, and the terms resHO2=n and resH=n appear in both 18 and 24. Looking at equations 15 and 16, shouldn't these terms appear only once each?

   Yes, this is right. We deleted them in eqs. (23) and (24).

10. I was slightly frustrated by the use of different scales in the various sub-plots in Figure 2 (and also 3). While I appreciate there are orders of magnitude differences between various sectors, it would be helpful to have these all plotted on the same scale (with different common scales between Figures 2 and 3). I think that it would be helpful, as these are contrasted with Figures A1 and A2, which do have a common scale for all the sub-plots of each figure.

    Thank you for pointing this out. We changed these figures and put them on a common scale.

11. In Figures 4 and 5, is the use of the 0.1 to 0.5 (and -0.5 to -0.1) band useful? The authors explicitly discount changes this small, and would changes on these levels even be significant?

    This is a good point. We deleted these figures and replaced them to a direct comparison with V1.0 as it was recommended by reviewer #1.

12. In Figures 4 and 5, could the authors explain the jagged feature seen in the OH biomass burning, the HO2 N2O decomposition, and to a certain extent, the HO2 lightning plots?

    We exchanged these figures to a direct comparison to V1.0 as it was recommended by reviewer #1. The jagged features resulted from divisions with small numbers.

13. Page 19, line 4: I believe the authors mean "no large changes", not "no changes", as this is the wording they use in two other places in the manuscript.

    Yes, we mean "no large changes". So we changed it. Thank you.

14. Page 19, line 11: "long-lived tracers".

    Thank you for this hint. We corrected it.

15. Page 19, line 11: I would not use "Exemplary", and would instead use "For example".

    We changed it.

16. Page 19, last paragraph: Is this referencing the plots in the Supplementary Information? If so, please say so.

Yes, indeed. We included the corresponding references.

17. In the Supplementary Information, I would suggest labelling the figures as S1, S2 etc., especially since these figures should be referenced in the main text in some way, and it would be confusing otherwise.

This is a good point. We changed the labels of the supplement.