

Interactive comment on “Global tropospheric hydroxyl distribution, budget and reactivity” by J. Lelieveld et al.

J. Lelieveld et al.

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This paper discusses the global OH atmospheric chemistry and analyzes the levels and chemical properties and the recycling of OH and HO₂ using the global modelling system EMAC. The paper reads smoothly at the most part, providing a lot of information on the chemistry of OH, and an abundance of results both in the main manuscript and the supplementary material.

General Comments The abstract should be more precise on what the main findings of this work are. The predecessor model as well as the previously assumed amounts of secondary sources should be specifically mentioned here.

Reply: The abstract is perhaps too brief. In the revised manuscript we will include some more information about the main findings, OH recycling and secondary sources.

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The terms “buffering” and “buffered” are used throughout the manuscript without proper definition given. Even after reading the entire manuscript it remains uncertain what the OH buffer actually is. The entire manuscript is based on the calculations made using an unpublished chemical mechanism (MOM) which is an update of a previous mechanism (MIM) using as reference a manuscript that is in preparation. Even though the full mechanism is included in the Supplementary material of the manuscript, a comparison of the model results using the updated mechanism to results of the previous mechanism and a more detailed comparison to measurements is needed. Also a better more complete budget analysis as well as a comparison and highlight of the differences between the two versions is clearly missing, especially since the authors give relative results such as “higher”, or “compared to predecessor models”.

Reply: In the revised manuscript we will remedy this. In the predecessor paper of Lelieveld et al. (2002) we presented sensitivity simulations with NO_x and CH₄, which showed that at a recycling probability of 60% or higher the OH concentration is insensitive to perturbations (i.e., the perturbations that can realistically be expected on Earth). We will use this to define the buffering concept. We will also include a more elaborate OH budget analysis, including a comparison based on methyl chloroform measurements, a discussion of measured and modeled OH over forested areas, and new sensitivity simulations to show the importance of the three main recycling mechanisms (NO_x, O_x and OVOC), which will also illustrate the OH recycling in biogenic VOC chemistry introduced into MOM. In the near future we will submit a separate manuscript (by Taraborrelli et al.) to provide details about MOM. This level of technical detail would not fit the format of the present paper.

The model description is rather short and feels incomplete. The EMAC modeling system is a complex system with a variety of options. The specific sub models used as well as the input used for the present study (i.e. emissions) should be clearly mentioned in the manuscript even if there is a small analysis in the supplementary material. The choice of RCP8.5 that suggests no further emission control also seems strange as it is

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often used to simulate the worst-case scenario. Even if in the year of interest (2013) the differences from the other scenarios are small, it still is an interesting choice and one that normally should be justified.

Reply: In the revised manuscript we will provide additional model description. Listing the submodels does not help unless a setup is applied in which prognostic routines deviate from the standard (e.g., Jöckel et al., 2010), which is the case here only with the chemistry routine MECCA and the emissions (both provided in the supplement). Actually, the main issue is the namelist used. This can be a problem with a complex model, and we will need to discuss in our EMAC community how to deal with it. One important aspect is that the model version used in the publication is frozen and publicly available. Page 17 of the supplement provides a rather detailed list of emission fluxes applied in the model. While the RCP8.5 scenario is a business-as-usual scenario for CO₂ emissions, it is much more conservative for reactive trace gases and generally conceived as more realistic than the other scenarios. For the year of interest (2013) these scenario aspects are hardly relevant.

Finally I would suggest that a label is added by the colorbar of all figures, indicating the depicted property/substance and the units. This would make the interpretation of the figures quite easier.

Reply: In the revised manuscript we will add the labels to the figures.

Specific Comments P1, L10: ...may be significant... ! change to something more precise, or explain thereason that they might not be significant. In all the reactions: Add the radical sign (dot) where necessary.

Reply: In the revised manuscript we will phrase this more precisely. While adding dots to radicals is generally good practice in chemistry, it is unusual in atmospheric chemistry. We will mention it in the text.

P3, L25: R6 does not directly produce OH, hence a more clear explanation of how OH

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is produced is needed.

Reply: We will add the complete list of reaction channels for R6, and change

$\text{RO}_2 + \text{HO}_2 \rightarrow \text{ROOH} + \text{O}_2$ (R6) Into

$\text{RO}_2 + \text{HO}_2 \rightarrow \text{ROOH} + \text{O}_2$ (R6a)

$\rightarrow \text{RO} + \text{O}_2 + \text{OH}$ (R6b)

$\rightarrow \text{ROH} + \text{O}_3$ (R6c)

and on p.3 line 25 we will refer to R6b.

P3, L26: While in polluted air peroxy.... ! While in polluted air, peroxy...

Reply: will be done

P4, L15: By interconnected, do you mean coupled? If yes, the more used (and easier to understand) term should be used. If not, please give a definition of what an interconnected sub model is.

Reply: we will replace with coupled.

P4, L27/28: Since only one year of results is presented (2013), why is a range of emitted quantities provided?

Reply: We have performed a 4-year simulation and only show the last year. In the 4-year period the online calculated emissions vary somewhat. We will specify this in the revised manuscript.

P5, L24/25 and elsewhere in the manuscript: Add the 105 term to the first number of all ranges.

Reply: will be done

P5, L27: Give the numbers calculated by Patra et al., since the discussion is based on them.

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Reply: will be done

P7, L4: Reaction R1 of the manuscript should be referenced here.

Reply: will be done

P7, L7/8 and figure 2 caption: scaled down by a factor of 20.

Reply: will be mentioned

P7, L28/29: O₃ from the stratosphere and O₃ from photochemically... ! O₃ from both the stratosphere and photochemically...

Reply: will be changed

P12, L15: The Physical-chemical tele-connections is here used without prior definition. Please give a clear definition.

Reply: In the revised manuscript we will refrain from using the work tele-connection.

Figure 5: Add the OH reactivity zonal means (latitudinal) since the height distribution is mentioned during the discussion in section 5.

Reply: This is provided in the supplement. In the revised manuscript we will mention this.

Figure 6: Enlarge the third panel of the figure since it is quite difficult to read the numbers in it. Also review the percentages given here since the numbers (as they are provided now) do not add up: e.g. for the OVOCs (red) the FT is 12% and the BL 19%. Multiplied by the 86% and 14% ratios respectively it gives a total of 13% (12.98) in the troposphere, where you present 12%. Maybe give the numbers with at least one decimal point so that the math comes out correct.

Reply: will be done and corrected where needed.

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