

Reply to comments of Anonymous Referee #2

This paper presents modeling studies of several aspects of the strong Indonesian biomass burning in 2015, including the impact of VOC emissions, the in-cloud oxidation of VOCs, and the transport of VOCs to the lower stratosphere by the Asian monsoon. This work uses the ECHAM/MESSEy model for a series of sensitivity tests, including two horizontal resolutions (1 deg and 2.8 degrees), and a comparison of aqueous-phase mechanisms. A new satellite product, HCN from IASI, is described in the appendix and used for model evaluation. A model experiment where all VOC biomass burning emissions are excluded is the basis for much of the analysis in the paper. There are a number of interesting results and new results, such as the aqueous chemistry impacts, presented, but I agree with the other reviewer that a major revision of the paper is required before publication.

Thank you very much for the helpful comments and seeing the value of our work to the community. Following the comments of both referees, we revised the manuscript significantly focusing on the following points:

- The title of the manuscript was adjusted to better represent the structural changes performed during the revision.
- We moved the comparison of the emission footprint of the Indonesian peatland fires to other biomass burning regions from the introduction to a new dedicated section (now Sect. 3).
- In addition to updating the section on the comparison of EMAC's representation of HCN to IASI satellite retrievals, we now also include a comparison to IASI CO retrievals.
- We restructured the reporting of our findings. It now follows a more systematic manner, in which we first report the impact on the troposphere (now Sect. 5), followed by the impact on the lower stratosphere (now Sect. 6). In both sections, we now focus on hydrocarbons, oxygenated organics, nitrogen containing compounds, and radicals in separate subsections. Afterwards the impact of in-cloud OVOC oxidation on the changes in the troposphere and stratosphere is discussed in a separate section (now Sect. 7).
- We extended the tropospheric analysis to include further hydrocarbons and OVOCs.
- Following the concerns of both referees, we removed the trend analysis on the influence of the Indonesian peatland fires on lower stratospheric O₃. Instead, we broadened the discussion on the lower stratosphere (hydrocarbons, oxygenated organics, nitrogen containing compounds, and radicals). In addition, we now focus on the change in the importance of the O₃ loss by phenoxy radicals in the UTLS and its potential to contribute to the lower stratospheric O₃ variability.

Please find in black the original comments and in red our replies.

The paper includes a broad set of topics - tropospheric composition, health impacts, aqueous chemistry, UTLS impacts - which make the paper seem a bit disjointed.

We understand that the original structure of the manuscript might seem a bit disjoint. Based on the comments, we revised the structure of the result sections and changed it such that it follows a more systematic structure. We first report the impact on the troposphere (now Sect. 5), followed by the impact on the lower stratosphere (now Sect. 6). In both sections, we focus on hydrocarbons, oxygenated organics, nitrogen containing compounds, and radicals. Afterwards, the impact of in-cloud OVOC oxidation on the tropospheric and stratospheric composition is discussed in a separate section (now Sect. 7).

As the other reviewer pointed out, the impact on stratospheric ozone appears to be in-significant and I recommend leaving that out.

Following the concerns of both referees, we significantly revised the section on the lower stratospheric composition. In the revised manuscript, we decided to remove the trend analysis and the related claims for the total O₃ loss in the lower stratosphere. Even though we agree that the changes

in lower stratospheric O₃ are small and localised, we still think that it is an interesting finding that aromatic emissions from the Indonesian fires potentially contribute to the lower stratospheric O₃ variability. Therefore, we now focus on the change in the importance of the O₃ loss by phenoxy radicals in the UTLS and its potential to contribute to the lower stratospheric O₃ variability. We find that in the upper northern tropical troposphere, this loss pathway contributes up to 40 % to the total chemical O₃ loss. When aromatic emissions from the Indonesian peatland fires are considered, this contribution further increases in the upper southern tropical troposphere by up to 20 % (from about 20 % to 40 %). Additionally, we broadened the discussion on the lower stratosphere, which now follows the same structure as the tropospheric analysis (hydrocarbons, oxygenated organics, nitrogen containing compounds, and radicals).

The paper discusses VOCs quite generally in many places, but does briefly summarize in Section 4.1 various categories of VOCs, and describes their differences. A bit more focus is placed on aromatics, and particularly phenols, which are certainly significant in biomass burning, but a number of other HCs and OVOCs are also important and should be discussed.

Following your recommendation, we expanded the discussion of hydrocarbons and OVOCs.

The discussion of HCN does not really fit with the reactive chemistry analysis and I do not find the use of it to evaluate the biomass burning emissions in the model very convincing. This paper seems to be an opportunity to describe the new IASI retrievals of HCN, but it seems like that discussion might be appropriate for a short AMT paper, with comparisons to other satellite retrievals of HCN and validation with insitu aircraft observations. Evaluation of the model with the established and validated CO satellite retrievals, from IASI and other platforms, would provide greater confidence in the model simulations.

As explained in the manuscript, the HCN satellite data have been retrieved from IASI observations using a general framework that has been fully described in dedicated publications and that has already been applied successfully to the retrieval of ammonia and of several VOCs (we provide the references in the text). Therefore, describing again the entire methodology, e.g., in an AMT paper, would be redundant with previous works, and we believe it is more appropriate to limit the explanation in an appendix to the few elements specific to the HCN retrievals (e.g., spectral range, post-filtering). A comparison of the IASI HCN columns with independent measurements is foreseen as part of a general study dedicated to the validation of the different IASI VOC products. The 2015 Indonesian peatland fires led to substantial emissions of VOCs, in particular of nitrogen-containing species. As shown with the satellite measurements in Fig. A3, it is particularly true for HCN, during this event by far the highest HCN columns and the largest plume captured on record throughout the IASI operational time series (since late 2007). We therefore believe HCN is an interesting tracer to represent the exceptional amplitude and extent of these peatland fires.

On the other hand, we agree that CO is a typical, widely used biomass burning tracer. Therefore, following your suggestion and the comment of the other referee, we revised the HCN comparison and extended the analysis to include also a comparison of EMAC's CO to IASI retrievals in Sect. 4. Here, we find that EMAC tends to slightly underestimate CO over Indonesia and overestimates CO in South America. We attribute the underestimation in Indonesia to a too low emission coefficient used by EMAC. The overestimation in South America is related to an overestimation of biogenic emissions in this region. Following both analysis we are certain that EMAC represents the Indonesian peatland fires in a reasonable manner, especially when considering the exceptional strength of the 2015 Indonesian peatland fires.

Here are some of the technical corrections that are needed:

l.110: in the resolution label, L90 apparently indicates 90 levels; explain 'MA'.

Exactly, L90 indicates that 90 levels are used. Here, MA indicates the Middle Atmosphere (MA) version of the model, i.e. that the 90 levels focus on the lower and middle atmosphere. In the original submitted version this is explained in line 114 and 115. In the revised version, we adjusted the explanation of MA in L90MA.

l.123: 'capable to represent' should be 'capable of representing'

Done.

l.193: 'one simulation exists, in which all ...' → ' in one simulation all ...'

Done.

l. 339: 'destruct' → 'destroy'

Done.

l.348: "80's" → "1980s"

Done.

l.396: relative → relatively

Done.

l.405: rewrite "which even enhances by in-cloud ..."

Done.

l.407: 'in the same order to SOA formation' → "to SOA formation on the same order"

Done.

l.431: reference for La Nina strengthening AMSA?

Basha et al. (2020) report a higher extend of the AMSA during La Niña. Please note that this statement is not longer included in the revised manuscript.

l.431: 'strengthens' → 'has strengthened'

Please note that this statement is not longer included in the revised manuscript.

l.433: extend → extent

Please note that this statement is not longer included in the revised manuscript.

l.456: 'appoint' is not the right word here. suggest? hypothesize?

Please note that this statement is not longer included in the revised manuscript.

l.467: 'our study' - does this refer to this manuscript, or previous work? If this paper, where is this shown?

This refers to this manuscript. We do not show these findings in any figure. We now include "(not shown)" in the revised manuscript to clarify this for the reader.

l.506: deletion → depletion

Done.

Table 1: column 2 would be better labeled 'Dominant fuel type' (fire type implies to meflaming or smoldering, for example).

A very good point. In Table 1, column 2 is now referred to it as dominant fire type. Within the text, we also now refer to 'fuel type'.

Table 2: What does 'ScSta' mean? Also, define JAMOC here.

Here, ScSta refers to EMAC's standard aqueous-phase mechanism and JAMOC to the complex in-cloud OVOC oxidation scheme Jülich Aqueous-phase Mechanism of Organic Chemistry. We added an appropriate elaboration to the caption of Table 2 and refer to Sect. 2.1.1. for further details.

References

Basha, G., Ratnam, M. V., and Kishore, P.: Asian summer monsoon anticyclone: trends and variability, Atmospheric Chemistry and Physics, 20, 6789–6801, <https://doi.org/10.5194/acp-20-6789-2020>, 2020.