

We thank the reviewer for reviewing the revised manuscript and giving us some more feedback. Below we address the comments point-by-point. The original comments are in black, and our responses are given in red and modifications in the revised manuscript are given in blue.

Reviewer: 1

The authors gone to some length to address the comments raised by both reviewers.

My main concern was about the VOC estimation and the related uncertainties. The authors provide good supporting evidence in their reply and added a Appendix Section about the specific and overall uncertainties for all relevant aspects of the SOA yield calculations. While the absence of direct VOC measurements is still not ideal, this is the best that can be done and sufficient for a robust study.

Response: We appreciate the comment from the reviewer.

My second major concern (the interpretation of the HOM results) has been resolved as well by reducing the prominence of the HOM related parts and making it clear that these are only “bonus findings” that support the main topic of the manuscript. That is perfectly fine.

Response: We appreciate the comment from the reviewer.

I found a few mostly technical details in the new sections of the manuscript that the authors should clarify before publication.

- 1) Why have the formed SOA mass concentrations changed? In Table 1: n-decane old: 1.8-7.3 ug/m³ , new: 4.8-9.3. These are measured values that should not be affected by any of the additional uncertainty calculations and the like. The authors state in one of the replies that particle transmission was already corrected in the original manuscript. SO that cannot be the reason for the higher values. What made all these values increase in the revision?

Response: Indeed, the particle transmission was already corrected for in the original manuscript. However, the reason for changed SOA mass concentrations is the new correction from the model for estimating the fate of low-volatility organic compounds (LVOCs) by Palm et al. (2016). As described in Palm et al. (2016), “The correction, hereafter referred to as the “LVOC fate correction”, is applied by dividing the amount of SOA mass formed by F_{aer} ” (where F_{aer} is the modelled fraction of LVOCs that are condensed on the aerosol). Therefore, all SOA mass concentrations increased when this correction was applied. To clarify this, we have now mentioned the LVOC fate correction in the revised manuscript:

Line 137-140: “The mass concentration of the SOA was calculated by combining the total particle volume from the SMPS and the SOA density calculated from the elemental ratios. The density was calculated for each step according to the equation in Kuwata et al. (2012), yielding in densities from 1100 to 1400 kg m⁻³. Mass concentration was also corrected by the “LVOC fate correction”, according to Palm et al. (2016), as described in the section above.”

As we also applied more averaging (for clearer plots and better statistics) in the revised manuscript, i.e., data points with same amount injected VOC are combined to one data point, there will be some changes in the data points plotted in the graphs and written in the table. For example, for n-decane (as the reviewer pointed out), the original the range changed from 1.8-

7.3 ug/m³ to new 4.8-9.3 ug/m³. For the lower end, we have three points (1.83...; 2.61...; 3.17... ug/m³), therefore the lower end of 1.8 ug/m³ in the original manuscript. The modelled F_{aer} for the averaged points is 0.53..., so taking the average of the original SOA mass concentrations and dividing it by F_{aer} , we get the new lower end of 4.8 ug/m³. Same thing with the higher end, it has two points (7.27... + 7.06... ug/m³) and F_{aer} is 0.77..., from that we get the new higher end of 9.3 ug/m³.

- 2) Fig 2: I really like the use of shading in this Figure. But the authors should double check the plotted values. Symmetric errors in linear space (e.g. +/- 30%) will not look symmetrical in log spacing. But some of the shading looks suspiciously symmetrical around the data points. IN the attached document, I picked an example (brown point at ~0.06). I added two black bars of equal length to highlight the similarity of the width of the positive and negative error band. I added an example for 0.06 +/- 40% (red point on the right)

Response: The reviewer is correct; the errors are not symmetrical in Fig 2. We separately calculated the positive and negative errors, by choosing the “worst case scenario” for the two cases. I.e., to find the lower error-band, we minimized the SOA-yield function (that consists of several variables with different uncertainties) and to find the upper error-band, we maximized the SOA-yield function. Thereby, the errors are not symmetrical as the described method for calculating the errors gives different upper and lower values. This is now also described in the Appendix when describing the uncertainties:

Line 413-417: “This includes the uncertainty of *both* particle phase instruments, as well as the uncertainty from the LVOC fate model (described in Section 2.1). *When calculating the SOA yield errors in Fig. 2, we calculated separately the upper and lower error values. This was done by altering all the variables within their uncertainty ranges in the SOA yield function to find the minimum and maximum values. This method will not result in symmetrical upper and lower errors, as seen in Fig. 2.*”

- 3) Original Specific Comment #1 Line 48 “bimolecular reaction”: From the authors reply, I understand what is meant by the phrase “when more bimolecular reactions took place”. But looking only at the phrase in the manuscript, it is still not clear which bimolecular reactions are meant in this context. There are many other bimolecular reactions that do not convert RO₂ to RO, e.g., the quenching reaction RO₂+HO₂. The authors need to make this more specific in the manuscript as the general reader will not look at the review replies.

Response: To clarify the paragraph, we have now removed “bimolecular reaction” and rephrased it as:

Line 46-51: “Wang et al. (2021) did not only measure HOM yields, but showed that the oxygen content in oxidation products generally increased when more *peroxy radicals (RO₂) were converted to alkoxy radicals (RO)*, even though not always reaching six or more O-atoms. Much of the O-atom incorporation was attributed to RO₂ *reactions* with other RO₂ radicals or NO, forming RO able to isomerize and thus allow reactions with molecular O₂.

This is in contrast to many monoterpenes where the RO₂ radicals themselves can undergo isomerization reactions (autoxidation), owing to suitable structures in the monoterpene-derived radicals which are less common in alkanes.”

- 4) Line 240 in Marked Manuscript: “...but as the particle mass increases, more volatile(, less oxidized and longer photochemical aging”. Is LONGER photochemical aging really correct here? This is opposite to what is shown in the Figures (E.g. Fig 1 in manuscript). The higher SOA masses were achieved by increasing the VOC concentrations and leaving the oxidant production the same which means that the OH exposure (equivalent photochemical aging time) is lower for the high SOA masses. So it should be “more volatile, less oxidized and SHORTER photochemical aging” here.

Response: The reviewer is correct: “longer” should be replaced with “shorter”. This is a mistake from our side. This is also easy to see in Fig. 3b where the colorbar (white to black) shows the photochemical age; we have shorter photochemical age at higher SOA mass concentrations.

This is now corrected for in the revised manuscript as:

Line 241-242: “...but as the particle mass increases, more volatile (, less oxidized and *shorter* photochemical aged) compounds can condense...”

NOTE: Markes types are modified in Figures 2, 3, 4 and A9 to ensure readers with color vision deficiencies to interpret them correctly. No data is changed, only the visual look is changed.

Kuwata, M., Zorn, S. R., and Martin, S. T.: Using Elemental Ratios to Predict the Density of Organic Material Composed of Carbon, Hydrogen, and Oxygen, *Environmental Science & Technology*, 46, 787-794, 10.1021/es202525q, 2012.

Palm, B. B., Campuzano-Jost, P., Ortega, A. M., Day, D. A., Kaser, L., Jud, W., Karl, T., Hansel, A., Hunter, J. F., Cross, E. S., Kroll, J. H., Peng, Z., Brune, W. H., and Jimenez, J. L.: In situ secondary organic aerosol formation from ambient pine forest air using an oxidation flow reactor, *Atmospheric Chemistry and Physics*, 16, 2943-2970, 10.5194/acp-16-2943-2016, 2016.

Wang, Z. D., Ehn, M., Rissanen, M. P., Garmash, O., Quéléver, L., Xing, L. L., Monge-Palacios, M., Rantala, P., Donahue, N. M., Berndt, T., and Sarathy, S. M.: Efficient alkane oxidation under combustion engine and atmospheric conditions, *Communications Chemistry*, 4, 10.1038/s42004-020-00445-3, 2021.