

Response to Referee Comment 2 (RC2) on ‘Effect of temperature on the formation of Highly-oxygenated Organic Molecules (HOM) from alpha-pinene ozonolysis’ by Lauriane L. J. Quéléver et al.

“The authors studied the formation of HOM (highly oxygenated organic molecules) by ozonolysis of α -pinene (and β -Pinene) and determined molecular HOM yields as a function of the temperature. Experiments were performed in the AURA chamber. HOM were measured by NO₃-CIMS. The findings are an interesting and important puzzle piece in finding out about the importance of HOM for SOA formation and atmospheric chemistry in general, as the temperature range was extended to -15°. Background for the study is that HOM formation should be hampered at low T, as the key formation step the rearrangement of peroxy radicals should be strongly dependent on temperature. The thorough study was a challenge and has some “weak points“, the main being the quantification of the HOM: even comparison of relative HOM pattern maybe difficult at different temperatures. As consequence the findings have some uncertainties. However, the authors addressed these issues and dealt with them quite clear and openly. Thus manuscript provides a new and original contribution for understanding HOM. The manuscript is highly interesting and very well written. I have only minor comments dealing with the quantification and some small remarks. I suggest to publish this very interesting manuscript in ACP. It could be even publish as it is, but the authors may want to consider the comments below.”

We thank the reviewer for the positive feedback and will answer the comments point-by-point below:

Comment 1: “line 185: “. . .the calibration factor $C = 1.65E9$ molecules cm^{-3} “, the reference to it is pretty vague. Do I understand correctly that you detect about 70 ppt HOM per normalized ct? Can you add your detection limit? “

Reply to comment 1: The reviewer is correct that we detect ~70 ppt HOM per normalized count, and we added this also to the manuscript for clarity. Reporting a calibration factor of the type used here has become customary for the CI-API-TOF, although most other similar instruments report sensitivities as cps/ppt. We also added our detection limit estimate.

Comment 2: “line 269: Did you try to verify your transmission function? Ehn et al. 2011 and Heinritzi et al. 2016 show quite different transmission behavior.”

Reply to comment 2: Unfortunately, we did not have the chance to do such a measurement. The transmission can indeed vary considerably based on the specific tuning of the instrument. In our study the calibration factor C was lower than typically reported, which may be an indication of relatively low transmission at the reagent ion masses. As we mainly look at the relative changes of HOM, the transmission function is ultimately of less concern in our study.

We also note that the transmission curves of Heinritzi (Fig 5a) and Ehn (Fig. 2d) look different mainly due to the selected curve type by which they are represented. The data points in these two plots are in fact fairly consistent, with the exception of the overall higher transmission in the instrument used by Heinritzi et al.

Comment 3: “line 270: How did you specifically estimate the uncertainty of tens of percent? From the variation of the RI 5-7E4 cps? (I guess you meant “several times 10%”?)”

“We conclude from the above investigations that changes on the order of tens of percent occurred in our instrument as the AURA chamber temperature was varied, and that only signal changes larger than this should be attributed to actual perturbations in the chemistry taking place in the chamber.”

Reply to comment 3:

This is correct, it was based on the RIC variation, and we have now added this more clearly into the main text.

Remarks:

(1) “line 142: “In practice, the experimental plan focused on α -pinene oxidation. . .”, sounds a little skewed. Maybe better “The experiments focused on α -pinene oxidation...”

(2) “line 373 and line 378: I suggest to reformulate this sentence. Despite your large un- certainty you still “calculate” the yields. They maybe only estimates because of the uncertainty. . . Accordingly you should replace “absolute estimates” by “molar yields” in line 378. “

(3) “Figure 3, legend: I suggest to replace “HOM spectrum” by “selected HOM”. It is easier to grip.”

Reply to remarks: Thanks for the suggestions, the text was modified accordingly.

References:

Ehn, M., Thornton, J. A., Kleist, E., Sipilä, M., Junninen, H., Pullinen, I., Springer, M., Rubach, F., Tillmann, R., and Lee, B.: A large source of low-volatility secondary organic aerosol, *Nature*, 506, 476, 2014.

Heinritzi, M., Hansel, A., Simon, M., Steiner, G., Wagner, A. C., Kürten, A., and Curtius, J.: Characterization of the mass-dependent transmission efficiency of a CIMS, *Atmos. Meas. Tech.*, 9, 1449-1460, 2016.