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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Molecular understanding of new-particle formation from alpha-pinene between −50 °C and 25 °C" *by* Mario Simon et al.

Anonymous Referee #2

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This manuscript investigates the effect of temperature on the molecular composition of α-pinene oxidation products and new-particle formation rates. As far as I know, this is to date the most systematic study of the temperature dependency of pure organic nucleation and the underlying chemical processes. The manuscript is generally well written. I think it can be accepted for publication after the authors address the following comments and suggestions.

1. Section 3.2: The authors show that the HOM yield decreases by about an order of magnitude from 6.2% to 0.7% from 25 degree C to -50 degree C. However, Quéléver et al. (2019), which is cited in this paper, showed a much stronger temperature dependence of HOM yields. They reported that the HOM yield decreased from 5.2% at 20 degree C to 6.3×10ˆ-3 % at -15 degree C. What causes this tremendous discrepancy?

Can you add a discussion of the reasons?

2. Figure 6: The authors mentioned that "the highest and lowest bin are overflow bins". How do you treat the yields in the volatility bins outside the range of this figure? Specifically, did you add the yields in all volatility bins < -11 to the bin of -11, or did you simply neglect these bins with the lowest volatility? I suggest that the author add some inserts or some more panels to illustrate the volatility distribution of ULVOC at even lower volatility bins. I speculate that at -50 degree C the volatility distribution of ULVOC could extend to a bin much lower than -12. This may help to explain why the NPF rates at -50 degree C are significantly larger than those at warmer temperatures even for the same ULVOC concentration (Fig. 7b). Also, why does PTR3 detect many ELVOC compounds under -10 degree C, but much fewer (or even none) under -25 degree C and -50 degree C?

3. Section 3.5, Line 588-589: The NPF rates under ion-free conditions still have a large spread. Particularly, the NPF rates at -50 degree C are still significantly larger than warmer temperatures. I guess the ULVOC distribution I mentioned in my last comment might be a possible reason.

4. Figure 7: In panels a and c, as temperature decreases, the nucleation rates remain roughly unchanged from 25 degree C though 5 degree C to -10 degree C, but increase sharply at lower temperatures. Any reasons?

5. As the author mentioned, this study covers a wide temperature range from ground level (25 degree C) to the upper-free troposphere (-50 degree C). However, the experiments are all conducted at a constant pressure of 5 hPa larger than surface pressure while the pressures in the real atmosphere can decrease from about 1000 hPa to about 100 hPa along with the temperature decrease. Will the pressure change significantly affect the temperature dependency of NPF rates? I would appreciate some discussions because this will affect how these results should be interpreted in the context of the real atmosphere.

6. Line 477-479: This argument can be moved to the next section.

7. Line 511: What are “other chemical systems”? Please clarify.

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