Response to the reviewers on the manuscript (egusphere-2026-270): “Influence of Tropospheric Temperature on the Formation and Aging of Secondary Organic Aerosol from Biogenic Vapor Mixtures” by Gao et al.,

The authors thank the reviewers for their careful reviews of our manuscript and their helpful comments and suggestions. All comments are addressed point by point below, with the comment in black, our response in blue, and the corresponding revisions to the manuscript in red. All changes of the original manuscript are marked in the revised version.

Reviewer #1

This study did chamber experiments to investigate the impact of temperature on formation and ageing of SOA particles formed from isoprene and α-pinene mixtures. With the temperature ranged from 213 K to 313 K, the authors found that isoprene strongly suppressed α-pinene dimer formation at 213 K. Volatility and phase state of SOA particles at different temperatures were also estimated. With temperate-dependent SOA yields considered in a chemical transport model, predicted SOA concentrations increased. As chamber experiments done as low as 213 K for SOA formation and ageing, particularly for SOA formed from mixed precursors, are still limited, and the related mechanism is not fully understood, I recommend the publication of this study after a major revision.

Major Comments:

My major concern is the writing of this manuscript. Some key information is missing in method section or when describing figures.

(1) Please specify how to derive the mass-based stoichiometric yields (α) at the four volatility bins, that are key values used in chemical transport models for simulating SOA formation. How can four α values be derived if only one AMF value is achieved at each experiment (Fig. S2)? In addition, as the authors noted that phase state of SOA particles plays a role in SOA formation, when deriving stoichiometric yields at different temperatures listed in Table S2, is phase state considered? Furthermore, Line 177-181 in method section 2.4, using '*243 K ≤T< 273 K, parameters from Exp 2 (243 K)*' as an example, I am wondering how the simulated SOA concentration would change if you adopt parameters from Exp 3 (273 K) when 243 K ≤T< 273 K? Similarly for other temperature ranges. I don't mean to do further sensitivity simulations but at least, the authors should discuss how the chosen of stoichiometric yields would affect the predicted results of SOA concentrations.

The mass-based stoichiometric yields at the four volatility bins that were implemented into PMCAMx were estimated based on the molecular composition of the particles measured by the FIGAERO-CIMS before warming started in each experiment and not from the AMF value. In the experimental part of the manuscript, we utilized 9 volatility bins while for the model we re-distributed to four volatility bins to minimize the computational cost in the chemical transport model. The comparison of the estimated yields in Fig. S2 with the measured AMF of each experiment demonstrates the consistency of our approach. This more detailed explanation has been added to section 2.4 of the manuscript to avoid any confusion.

"The simulation chamber results are implemented to PMCAMx (Murphy and Pandis, 2009), a chemical transport model (CTM) which utilizes the SOA volatility bases set approach (Lane et al., 2008) to simulate the formation of secondary aerosol from biogenic and anthropogenic VOCs. A brief description of PMCAMx is provided in Section S2 of the supplement. The model-incorporated stoichiometric yields are based

on the molecular composition of the particles measured by FIGARO-CIMS. The volatility of the produced aerosol is determined following the approach described in Section 2.3, however, to minimize the computational cost the species are re-distributed to four VBS bins (10^0, 10^1, 10^2, 10^3 μg m^{-3}) rather than using the whole volatility range."

The phase state of the aerosol is indirectly considered when the mass-based stoichiometric yields are estimated, because the actual composition of the SOA is used and the estimated yields are consistent with the measured AMF. This point has been added to the section 2.4.

For the partitioning of the secondary organic species between the gas and the aerosol phase, PMCAMx assumes that there is equilibrium between the two phases and that the organic compounds form a pseudo-ideal solution. Specifically, in the model the partitioning of the organics between the gas and the aerosol phase depends on two parameters, the temperature and the total OA concentration of the simulated cell. Therefore, the model can replicate the concentration changes that occur in the atmosphere due to both warming and cooling. Utilizing the experimental data after warming rather than before warming commenced inside the chamber would result in the same predicted SOA concentrations by PMCAMx. Nevertheless, the warming stage of the experiments is better represented in the model by adopting the mass-stoichiometric yields derived from the initial temperatures. This discussion has been added to the section 2.4.

"For the partitioning of the secondary organic species between the gas and the aerosol phase, PMCAMx assumes that there is equilibrium between the two phases and that the organic compounds form a pseudo-ideal solution. Specifically, in the model the partitioning of the organics between the gas and the aerosol phase depends on two parameters, the temperature and the total OA concentration of the simulated cell. Therefore, the model can replicate the concentration changes that occur in the atmosphere due to both warming and cooling. Utilizing the experimental data after warming rather than before warming commenced inside the chamber would result in the same predicted SOA concentrations by PMCAMx. Nevertheless, the warming stage of the experiments is better represented in the model by adopting the mass-stoichiometric yields derived from the initial temperatures. Values for the Base and New cases are provided in Table S1 and S2. Utilizing the stoichiometric yields of Table S2, Figure S2 depicts the secondary organic aerosol mass fraction derived as a function of the total organic aerosol mass together with the experimentally measured values."

(2) There is only one sentence describing Fig. 1 in the main text. Please expand the description. Are the SOA concentrations shown in Fig. 1 wall-loss corrected? Concentration variations of VOC precursors are hard to spot for experiments at 213 K and 243 K. Adjust the maximum values of y-axis. From Fig. 1, I suppose the authors use time-varied AMF values deriving stoichiometric yields shown in Table S2 (Pathak et al., 2007)? I have major concerns in Fig. 1, and Fig. S2 with Table S2 commented

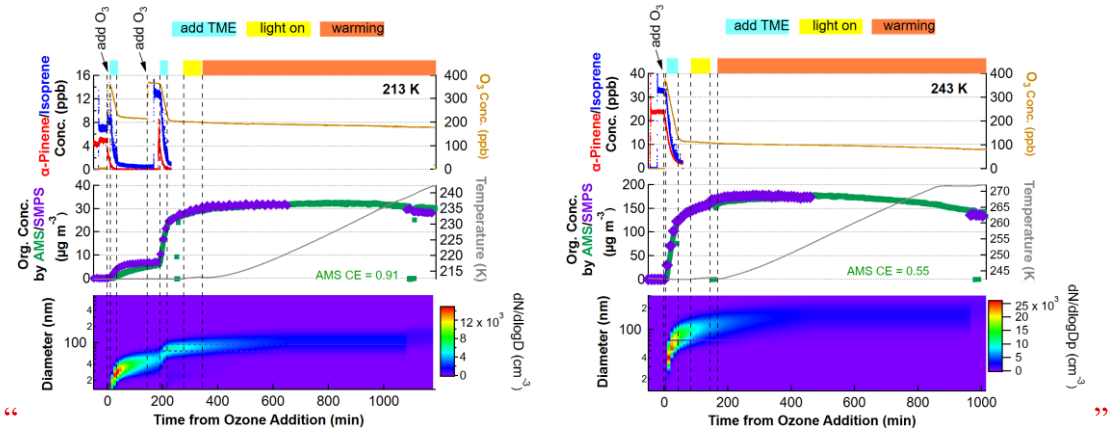
above, because these updated stoichiometric yields varied with temperatures can be applied by chemical transport modeling community to improve SOA simulations.

We rescaled Figure 1 and added more description on Figure 1 as follows:

"The beginning of the ozone addition is considered time zero for each experiment. The addition of ozone and the subsequent production of OH radicals, resulted in the complete depletion of the initial α-pinene and isoprene. The SOA particle diameters increased typically to 65-100 nm and the mass concentrations of the newly formed SOA ranged between 33-133 μg m^{-3} (mass yield: 9-52 %) depending on temperature. Subsequently, the fresh SOA was exposed to simulated solar radiation for 1 h. This photochemical aging resulted in no significant chemical change (cf. Figure S9). This aging age was followed by 10-12 h of warming, which will be discussed in Section 3.2."

The effect of particle wall losses was minor based on our previous studies in this large aluminium chamber (Huang et al., 2018; Gao et al., 2022; Gao et al., 2023; Gong et al., 2024) for which we did detailed aerosol dynamic modelling with the COSIMA model (Saathoff et al., 2009). To quantify the impact of dilution we used CO_2 as a tracer and estimated an overall average dilution during warming of less than 4% for all experiments.

The y-axis for 213 K and 243 K is adjusted:



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The yields of each VBS surrogate were not derived from the measured AMF but from the FIGAERO-CIMS measurements as explained in our response to the previous comment. We have updated Section 2.4 of the manuscript to better describe how the mass-based stoichiometric yields utilized in the model were derived from the experimental data.

(3) Similar to Comment 2, there is only one sentence describing Fig. 3 in the main text: *'However, by comparing the particle volatility distribution at different temperatures based on the gas- and particle-phase measurement at each temperature and based on the Clausius-Claepery equation (Figure 3), the strong temperature dependence on the ISO-AP dimers to AP-AP dimers between 213-273 K is suggested to be chemistry-driven.'* Such a long sentence is very hard to follow. I suggest describing the two

methods deriving volatility distributions in separate sentences. Is the Clausius-Clapeyron equation applied after the chemical-composition-based volatility estimation for 298 K (Li et al., 2016)? Is C^* shown in Fig. 3 for the volatility at experimental temperatures? Also for Line 255-256, at low temperatures, why is the rate coefficient of isoprene + OH higher? Better split the long sentence for readers to follow.

We agree that the long sentences are hard to follow. We have separated them into several sentences for better understanding:

"Figure 3 shows the comparison of the particle volatility distribution at different temperatures. This comparison integrates two approaches: gas- and particle-phase measurements at each temperature (i.e., $C^* = C_{OA}\frac{G_i}{P_i}$ (Gkatzelis et al., 2018)) and the Clausius–Clapeyron equation. $C_{14\text{-}15}$ dimers span in the less volatile bins from Clausius-Clapeyron equation at temperatures below 273 K, exhibiting a strong temperature dependence, compared with the volatility based on the measured organic mass. This dependence is suggested to be chemistry-driven rather than governed solely by phase partitioning."

The Clausius-Clapeyron equation was applied for the estimation of volatility for other temperatures after the chemical-composition-based volatility estimation for 298 K. In Figure 3, the volatility is at experimental temperatures. We clarify this by adding a sentence to Figure 3:

"The volatility of the bins is at their experiment temperatures."

Previous studies (Campuzano-Jost et al., 2000; Campuzano-Jost et al., 2004; Dillon et al., 2017) have demonstrated a negative correlation between the rate coefficient of isoprene + OH and temperatures, because of a negative activation energy. Therefore, we suggest a higher potential of the production of C_5 RO_2 leading to the stronger production of C_{15} dimers rather than C_{20} dimers at lower temperatures.

Specific Comments:

(1) Page 3, Line 119-120: Why for the experiment at 213 K, the injection of VOC precursors and O3 were done two times, different from other experiments? Was it because the SOA concentration formed at thus low temperatures was too low? In addition, '*213 K experiment*' in Table 1 seems not match the description here on Line 119-120. Exp. 6 and 7 in Table 1 are not described in Method section.

After the first oxidation of the VOC mixture, the SOA concentration was too low for the longer warming experiment. Therefore, we repeated the SOA formation with about twice the VOC concentrations. To clarify, we modified the sentence in lines 119-120 (now lines 118-121):

"At 213 K, the initial concentrations of isoprene and α-pinene of 6.7 ppb led to a relatively small amount of SOA mass. To generate sufficient SOA mass for the longer

warming experiment we generated more SOA mass in a second oxidation step with about twice the VOC concentrations of 13.5 ppb."

Furthermore, we added the following information to Table 1:

"*Total amount of O_3 of 366 ppb is summed from two rounds of injections: 253 ppb at the first injection, and 113 ppb at the second injection."

The description of Exp.6 and 7 has been added to the Method section:

"Additionally, to investigate the cross-dimers formed from the oxidation of α-pinene and isoprene, we used ^{13}C-labelled isoprene (>98%, Merck) in Experiments 6 and 7 (Table 1). This isotopic labelling enabled us to identify products with a shift of one nominal mass-to-charge unit (i.e., m/z +1), which unambiguously marks those dimers containing one skeleton from the labelled isoprene."

(2) Page 8, Line 237: Change '*shown as Figure 2g and 1h*' to shown as Figure 2g and 2h.

This was corrected accordingly.

(3) Figure 4: It is hard to follow the trend of these small symbols. Using solid vs open symbols for warming start and end may be clearer than current version. Page 12 Line 320, I did not see slope of 3.25 in Fig. 4.

The slope of 3.25 refers to the ratio of H:C increment and O:C increment after warming. Since the H:C values (from 1.69 to 1.82) and O:C values (from 0.36 to 0.4) have been reported in the text above, we do not show the slope of 3.25 in Figure 3. To clarify this, we modified the sentences as follows:

"According to the HR-AMS measurements, the bulk O:C and H:C ratios of SOA particles formed at 243 K (SOA_{243K}) increases from 0.36 to 0.4 and from 1.69 to 1.82, respectively, during gradual warming to 273 K ($SOA_{243K\rightarrow273K}$). Although the incremental O:C change is small, the online HR-AMS measurements showed a significant trend during the warming process (Figure). The ratio (=3.25) of the H:C increment and O:C increment in the Van-Krevelen diagram indicates hydration reactions during warming…"

We tried to improve the Figure 4 by using solid vs open symbols. However, since the symbols are very small, it is not significantly distinguishable between solid and open symbols. Therefore, to make the Figure 4 better visible, we re-arranged the two sub-plots and zoom in more to H:C 1.3-2.0 and O:C 0.3-0.7 ranges. The updated Figure 4 shows as follows:

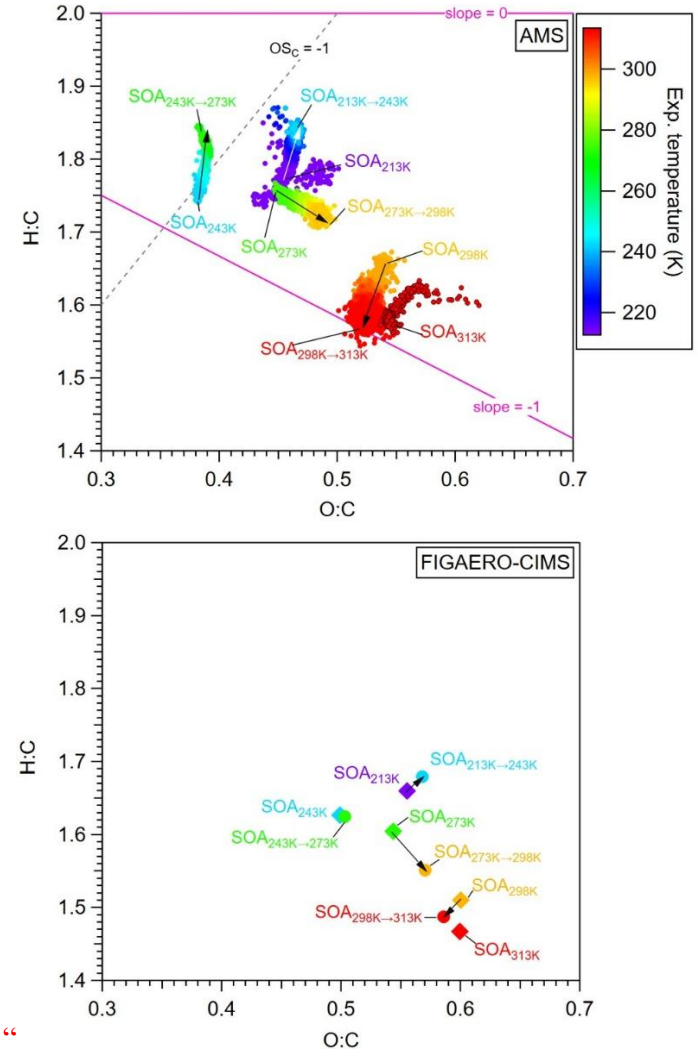

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Figure 4. Van-Krevelen diagram for SOA particles during the warming periods of Exp 1 (213 K to 243 K), Exp 2 (243 K to 273 K), Exp 3 (273 K to 298 K), Exp 4 (298 K to 313 K), and Exp 5 (313 K) from HR-AMS measurements (left) and FIGAERO-iodide-CIMS measurements mean values (right, symbols of diamonds and circles for warming start and end, respectively). Arrows are for guiding from the start to end of the warming periods. Symbols are coloured by temperatures. The carbon oxidation state (OS_C = 2 O:C – H:C) is shown with a grey dashed line. The pink lines with different slopes represent various reaction pathways: slope = 0 (formation of hydroxy/peroxy groups); slope = −1 (formation of carboxylic acids, or addition of both hydroxy and carbonyl groups).”

(4) Figure 5 and Line 323, Line 359, why the authors choose to calculate the compounds lost based on FIGAERO-CIMS instead of using AMS to calculate the mass loss after warming? In the Conclusion section Line 518, however, the authors use mass to describe the loss after warming.

We focused on the compounds lost based on FIGAERO-CIMS instead of AMS because we wanted to identify the molecular species that evaporated from the particle phase. In the Conclusion section Line 518, the “losing 72% of its mass” still refers to the loss of compounds based on FIGAERO-CIMS, which has been discussed in Line 357-358:

"During warming of SOA_{273K} to 298 K, 72.5 % of all particle-phase $C_xH_yO_z$ compounds are lost, with OS_C…". We are sorry for the confusing statement, and revise it as follows:

"…, losing 72.5% of all the particle-phase $C_xH_yO_z$ compounds detected by FIGAERO-CIMS."

Furthermore, we added the overall organic particle mass loss based on HR-AMS in lines 362-364:

"Correspondingly, the oxidation state (OS_C) of bulk SOA_{243K} decreases from -0.97 to -1.02 for $SOA_{243K\rightarrow273K}$ (Figure S10), while the organic particle mass decayed by 17% as measured by HR-AMS."

And in lines397-399:

"In contrast, from the HR-AMS measurement, the bulk SOA_{273K} loses 75% of mass, and show a significant increase of OS_C…"

And also lines 413-415:

"During warming from 298 to 313 K, 40.2 % of all particle-phase $C_xH_yO_z$ compounds and 71 % of organic particle mass were lost, by substantial evaporation."

(5) Page 13, Line 338-344, the *T*g calculation is better put in the method section for readers to follow. What is the value of hygroscopicity parameter used? Is the T_g estimated (e.g., Line 345) considering water uptake by SOA particles?

We added the T_g calculation to the method section:

"Based on the same dataset used for the volatility prediction, the glass transition temperature (T_g) of CHO compounds is estimated by the parameterization method expressed by equation (3) (Derieux et al., 2018):

$$T_g = (n_C^0 + \ln(n_C))b_C + \ln(n_H)\,b_H + \ln(n_C)\ln(n_H)\,b_{CH} + \ln(n_O)\,b_O + \ln(n_C)\ln(n_O)\,b_{CO} \quad (3)$$

where n_C, n_H, n_O are the number of molecular C, H, O atoms, respectively; n_C^0 is the reference carbon number; b_C, b_H and b_O refers to the contribution of each atom to T_g; and b_{CH} and b_{CO} are coefficients reflecting contributions from carbon–hydrogen and carbon–oxygen bonds, respectively. The values of all parameters used can be found in the published paper (Derieux et al., 2018)."

Water uptake and hygroscopicity were not included in the T_g estimation. This is because we did not measure the hygroscopicity parameter, therefore we are not able to calculate the mass concentration of water in the particles using the Gordon-Taylor equation (Derieux et al., 2018). We acknowledge that this is a limitation of the T_g estimation in this work. We added a sentence to the line 388-389:

"We note that the T_g values may be underestimated, as the water content in the particles was not taken into account due to a lack of measurements."

(6) Page 14, Line 401-403, during warming, as evaporation leads to lower-volatility compounds enriched, why does the entire VBS shift toward higher volatility? Is Fig. 6 and Fig. S11 for C^* estimated at experimental temperatures instead of 298 K?

During warming, evaporation leads to compositional changes that enrich the relatively lower-volatile compounds. Concurrently, rising temperatures shift the entire VBS toward higher apparent volatility, following the Clausius-Clapeyron relation. For instance, despite the evaporation of some volatile components during warming from 243 K to 273 K, the resulting $SOA_{243K\rightarrow273K}$ particles exhibit higher overall apparent volatility, containing only 35% of LVOC/ELVOC/ULVOC (Figure S9). The C^* of compounds in $SOA_{243K\rightarrow273K}$ in Figure S9 is at 273 K, while the C^* of compounds in SOA_{243K} in Figure S9 is at 243 K. Therefore, although the less volatile compounds are enriched after warming, some of them could be grouped into higher volatility groups due to the shift of the entire VBS according to the Clausius-Clapeyron relation.

The C^* values in Figure 6 are at the experimental temperatures, while the C^* values in Figure S11 (now S12) are at 298 K for better showing the change of 298K C^* at different temperatures.

(7) Page 16, Line 440-442, the authors stated that lower temperature enhanced the formation of both ISO-AP cross dimers and AP-AP dimers, which seems contrary to Line 260 and Line 508 that ISO-AP cross dimers suppress the formation of AP-AP dimers. Please explain this.

We thank the reviewer for pointing out this mistake. We have revised the sentence in Lines 440-442 (now in Lines 494-497):

"Thus, we emphasize that, besides promoting the condensation of condensable components, lower temperatures chemically enhance the formation of ISO-AP cross dimers, while higher temperatures…"

(8) Page 18, Line 519-520: it is stated that SOA formed directly at higher T is more volatile than SOA formed at lower temperatures and subsequently warmed, which seems contrary to Line 459-460 that higher volatility of SOA 273K→298K is observed than that of SOA298K.

We thank the reviewer for pointing out this imprecise statement. We have revised the sentences as follows:

"Except for particles formed at 273 K and subsequently warmed to 298 K, in all other temperature regimes, SOA formed directly at higher temperatures is both more oxidized and more volatile than SOA formed at lower temperatures and then warmed."

(9) Supplement: Section S3: Fig. S8 did not match the description in Section S3. Please double check the Figure number cited in SI. Furthermore, '*Based on this method to determine SOA compound volatility, all compounds in the α-pinene isoprene derived SOA particles at 213 K should have a low volatility*', 'this method' in the above sentence

refers to which method? Please specify the difference of 'this method' and the volatility estimated from chemical composition.

We added a new figure in the supplement to support the statement in Section S3, and revised all figure numbers in the supplement:

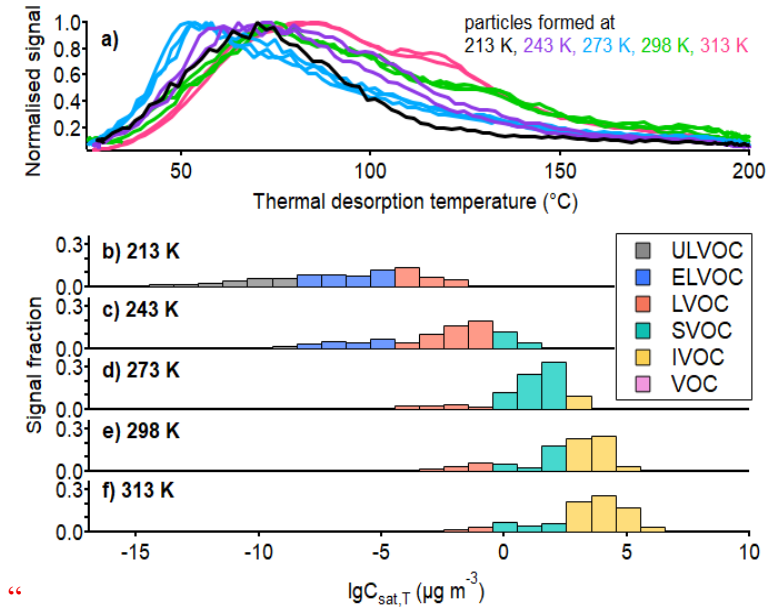

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Figure S1. Thermal desorption profiles for SOA formed at different temperatures (colors) panel (a). Two profiles in the same color are from subsequent deposited particles. Volatility distributions for particles formed at 213 K (b), 243 K (c), 273 K (d), 298 K (e), 313 K (f) with $C_{sat,T}$ shifting according to the Clausius-Clapeyron relation; colors indicate different volatility groups. The ULVOC, ELVOC, LOVC, SVOC, IVOC, and VOC are identified based on the saturation concentrations as described in the method section."

"This method" refers to the parameterization approach based on the molecular formula. We clarify this by revising the sentence in the Section S3:

"Based on the parameterization method using the molecular formula to determine SOA compound volatility (detailed description in the Method section), all compounds in the α-pinene isoprene derived SOA particles at 213 K should have a low volatility, …"

Reference:

Pathak, R. K., C. O. Stanier, N. M. Donahue, and S. N. Pandis (2007), Ozonolysis of a-pinene at atmospherically relevant concentrations: Temperature dependence of aerosol mass fractions (yields), J. Geophys. Res., 112, D03201, doi:10.1029/2006JD007436.

Reviewer #2

This manuscript presents a study investigation of the temperature dependence of secondary organic aerosol formation and aging from mixtures of isoprene and α-pinene at different temperatures. The authors use AIDA chamber experiments, detailed molecular composition, and implement the results in a chemical transport model. The

study addresses an important question: how tropospheric temperature influences SOA formation pathways and particle aging, particularly in systems involving multiple biogenic precursors. Most laboratory studies are conducted near room temperature, so the systematic exploration of temperatures spanning much of the troposphere is actually very good and very important.

Overall, the manuscript is very good, well written and very interesting for the scientific community. The combination of chamber measurements, molecular characterization, and modelling is a particular strength.

Overall, I believe the manuscript is suitable for publication in ACP.

Minor comments

The manuscript states that the PMCAMx simulations cover 5 June – 8 July 2012 during the PEGASOS campaign. However, Figure 7 appears to refer to measurements during June–July 2019, which is inconsistent with the PEGASOS campaign timeline. I believe that the right time is 2012.

Indeed, the correct year is 2012. It is now corrected.

The chamber experiments are conducted at relatively high precursor concentrations (tens of ppb VOC and several hundred ppb O3). That is very typical for chamber studies, however those conditions can change radical chemistry, oligomer formation and SOA yields. It would be nice to see a deeper discussion on how these conditions differ from typical atmosphere.

We agree that the relatively high precursor concentrations used in our chamber experiments (tens of ppb VOC and several hundred ppb O_3) may influence radical chemistry, oligomer formation, and SOA yields compared to typical atmospheric conditions. We have now added a dedicated discussion in the revised manuscript (lines 449-459) to address this limitation. Specifically, we note that while such concentrations are typical in chamber studies to achieve sufficient particle mass for detection and for >10 hours aging by warming, they can enhance the rates of bimolecular reactions, potentially favoring lower-volatility products and higher oligomer content than under ambient conditions. Conversely, the high O_3 levels may accelerate the oxidation aging process. We have also explicitly acknowledged that our observed volatility distributions and Tmax values represent an upper limit of the reactivity regime, and that extrapolation to atmospheric conditions should be made with caution.

"Here, we note that the precursor concentrations used in this study are substantially higher than typical atmospheric levels. While such conditions are typical in chamber experiments to generate sufficient particle mass for instrument detection and >10 hours aging by warming, they may influence the underlying chemical processes. Specifically, elevated VOC and O_3 concentrations can enhance the rates of bimolecular reactions, potentially favoring radical–radical recombination, accelerating oligomer formation (Zhao et al., 2023), and increasing SOA yields relative to ambient conditions. These

conditions may also shift the partitioning of semi-volatile species toward the particle phase. Consequently, the volatility distributions reported here likely represent an upper bound of reactivity and should be interpreted with caution when extrapolating to atmospheric conditions."

References

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