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Interactive Comment

Interactive comment on "Relating CCN activity, volatility, and droplet growth kinetics of β -caryophyllene secondary organic aerosol" by A. Asa-Awuku et al.

A. Asa-Awuku et al.

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We thank the reviewer for the positive feedback and comments.

The manuscript is generally well-written, and the introduction does a good job of discussing the key concepts in the field and extensively cover the existing literature. That said, the paper could be significantly improved if the authors would carefully lay out in the first two sections a more detailed account of the five experiments included in the study, and the motivations for choosing which instruments to include for each experiment. The use of a summary table (i.e., Table 1) is a practical one, but in this case many key details are missing and the logic behind the progression of experiments seems somewhat difficult to follow.





We have included an improved description of the experiments.

One significant concern is the lack of repetition for the experiments used to support the key findings for the study. Only two experiments are included for which data from both CCN counters are available, and only one experiment included the use of the thermal denuder. The authors use these limited results as the sole basis for the first of their "major findings". Their argument is supported by the data, but the limited amount of information leaves available alternative explanations, including something as unfortunate as an undetected instrument malfunction. It seems odd that such a large impact of semivolatile SOA on CCN activity would exist for b-carvophyllene where (as the authors indicate) there is no observed difference between the instruments for monoterpene SOA. The differences between the CCN counters when measuring the effect of the oxidation pathway is not sufficiently explained, again suggesting the possibility of instrumental variability. One possible check on the consistency of the SD CCN counter would be to compare the results of experiment 1 & 4, which were carried out at very similar conditions; the authors presumably have this data and should include it in the manuscript. Ideally, more replicates of the experiment would be added to the study, but the reviewer acknowledges that this is probably not feasible.

The experiments carried out are limited in number, but were repeated to confirm their robustness. For example, Experiment 1 was repeated at least two more times, and the discrepancy between the two CCN counters was consistent. The two CCN instruments agree very well for monoterpene SOA and $(NH_4)_2SO_4$ calibration aerosol. This, together with the support provided from the thermal denuder CCN experiments suggests that the effects are real, and not instrument malfunction.

Despite some skepticism that the presence of semivolatile hygroscopic material fully explains the observed discrepancy between the two instruments, the results of experiment 2 with the thermal denuder are quite exciting. Clearly the hygroscopic component is more likely to evaporate under dry conditions. This raises

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an interesting question: would this same material be continue to dominate the overall volatility under the humid conditions of the CCN counters and the natural environment? Within the CFSTGC CCN counter, the aerosol sample would be humidified and probably deliquesced before significant heating occurs; this is substantially different from the conditions within the thermal denuder.

This is a good question, but will be the subject of a future study as it requires substantially more experiments to deconvolute volatility from RH effects.

The remaining analyses are well developed, including the major finding that the less volatile material remaining after thermal denudation is connected to slower growth kinetics. In light of the limited nature of the available data, it would be preferable if the concluding statements were more nuanced, but the authors do not radically overreach in this regard.

Done.

In addition to the above major comments, there are the following minor comments: Page 10111, Line 25: What fraction of the SOA volatilizes in the thermodenuder?

Based on shifts in the size distribution, roughly 30% of the particle volume evaporates in the thermal denuder during the course of the experiment. We have included this in the text.

Page 10112, Lines 4-26: What are the measurement uncertainties for the SD CCN counter, and for the CFSTGC CCN counter operating in various modes? The SMCA technique would seem to have particularly high measurement uncertainties under atmospherically relevant aerosol concentrations. Note that we are not told the actual particle concentrations for the experiments described in this study.

The absolute supersaturation uncertainty was 0.03 at 0.61%, 0.05% at 1.02% super-

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saturation for the CFSTGC, and, 0.02% at 0.6% supersaturation for the SD.

The SMCA method is subject to the same concentration uncertainty as expected with an SMPS for the CCN-relevant sizes. Total CCN concentrations vary between a few hundreds to thousands of particles per cm⁻³, hence for the 2 minute scantimes used here, the counting statistics are more than sufficient for a reliable CCN/CN activation curve.

Page 10115, Lines 8-10: The density assumption is critical to the subsequent soluble fraction calculation. How sensitive are these subsequent calculations to the choice of density?

Köhler theory analysis provides molar volume (M_o/ρ_o), which is then introduced into Equation (4) for computation of solute volume fraction. This means density uncertainty does not affect the calculation.

Page 10119, Lines 1-3: A minor point, but to my eye the differences between Figs. 2 & 3 seem to be in slope rather than shape (i.e., Fig. 3 looks sigmoidal only because the transition occurs more rapidly than the Fig. 2 data).

We have modified the text to reflect this.

Page 10127, Line 5: I would hesitate to extend the observed results to sesquiterpenes generally.

Good point. The word "sesquiterpene" has been replaced with " β -caryophyllene".

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