

Interactive
Comment

Interactive comment on “CCN activity and volatility of β -caryophyllene secondary organic aerosol” by M. Frosch et al.

Anonymous Referee #1

Received and published: 13 October 2012

In the reviewed work, the authors present CCN activity and volatility results from the formation of beta-caryophyllene SOA experiments. The experimental procedure is similar to previously published works with the exception of the addition of HONO as a photochemical OH source. The results agree with previous studies and confirm that the role of OH in betacaryophyllene SOA experiments plays a significant role in the aerosol formed and its subsequent water uptake. The paper is written well and is appropriate for this journal. The figures are often too small and difficult to read. In some areas of the text, the justification of observations is vague and the arguments presented for the SOA behavior are not convincing to the reviewer. Overall, this paper does a nice job contributing supporting evidence of the CCN properties of betacaryophyllene SOA. However there are flaws in the interpretations and statements of observation. The reviewer has the following major and minor concerns.

C8112

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



MAJOR CONCERNS.

Like the work of Asa-Awuku et al, 2009 aerosol does not activate for several hours after nucleation. Critical dry diameters are presented in fig 3 but to what fraction do the larger particles (>100nm) grow? In this region, a critical diameter may not be obtained but particles can grow. The change in activated fraction may also be indicative of droplet growth that may be inhibited. Asa-Awuku et al, show that it takes considerable time for droplets of a given dry diameter to activate into droplets similar to that of ammonium sulfate at 100nm. Is this also true of the aerosol formed in this system? If so, the consideration that no kinetic inhibition exists may need to be revised. If kinetic inhibition does exist such that droplets less than 0.75 microns (the lower detection limit of the CF-CCNC OPC) are formed at the exit of the column, they will not be counted. This will bias all CCN/CN fractions and TDGA. The authors should reconsider their statement that no kinetic limitations exist.

Page 20753, line 27. I would also be hesitant to assume that the chemical composition of the gas phase in the SD-CCNC is similar to the gas phase of the flow coming directly from the smog chamber. Humidity conditions, condensation and adsorption of gas molecules on particles and the CCNC SS influences on gas-particle re-partitioning are not characterized.

Page 20758, line 28. This section of the text is somewhat confusing. For one, the authors do not give a specific residence time of the SD-CCNC, and the residence time in the TD (6.2 s) is similar to that in the CF-CCNC (8 s).

Page 20760, line 24~28. The stated comparison between the results of Alfarra et al (2012) and Tang et al. (2012) should not be made since the precursor concentrations are vastly different. Considering that Tang et al. also observed that there were no obvious variations of hygroscopicity when precursor concentration was above 50 ppb, the two results are actually consistent with each other.

Why does the hygroscopicity of aerosol measured with the SD-CCN decrease in Fig

4a? Little discussion is provided on this interesting phenomenon.

Page 20758, line 27: After the TD, are the particle and stream temperatures at the inlet of the SD-CCNC the same as those without passing a TD? The authors need to make sure that the particle and stream temperatures under different experimental conditions are the same at the CCNC inlet to make comparisons. In addition, the reviewer is not entirely convinced that Fig 5 provides sufficient evidence of the following, "L9. Observations indicate that SOA generated under light exposure is not volatile or that compounds contributing to CCN are not highly volatile". How does the volatility profile of the SOA change with time? In Fig 5b. The aerosol becomes significantly more hygroscopic with time; especially during the first 10 hours shown in Fig 6. Since, TD and Bypass data are not preformed simultaneously it is difficult to the compare the temporal trends. Is it not also plausible that the thermal denuded aerosol of a more hygroscopic particle at 2.5 hours can be more hygroscopic than the particle a bypassed particle at 2 hours (sometime before)? The last few points after 7 hours (once composition stabilizes?) in the CF-CCNC may suggest that the aerosol hygroscopicity is susceptible to temperatures when formed with light. The reviewer would appreciate clarification.

Lastly, how much does temperature in the chamber change from the addition to lights? This could also effect the SOA formation processes. Perhaps the authors could also show this in Fig. 6.

OTHER CONCERNS: Page 20759, line 22. What experimental uncertainties?

Page 20759, line 19. Tang et al, 2012 were able to show that reaction s with OH produced aerosol of higher hygroscopicity and increased oxygenated content (O/C)

Fig. 4. The symbols are too small and it is difficult to see the difference between open and closed symbols. In 4b, it is difficult to distinguish the squares from the circles.

Page 20762, line 13. How significantly with dilution?

Interactive
Comment

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)

[Discussion Paper](#)



Page 20762, line 27. Heterogeneous chemical reactions may also play a role in chemical aging under dark ozonolysis conditions

Fig. 8. What is the significance of a fit to the -1.2987 dependence? What will a forced fit with exponent -1.5 look like?

REFERENCES: Tang et al citation should be updated .

Asa-Awuku, A., Engelhart, G. J., Lee, B. H., Pandis, S. N., and Nenes, A.: Relating CCN activity, volatility, and droplet growth kinetics of α -caryophyllene secondary organic aerosol, *Atmos. Chem. Phys.*, 9, 795–812, doi:10.5194/acp-9-795-2009, 2009.

Tang, X., Cocker III, D. R., and Asa-Awuku, A.: Are sesquiterpenes a good source of secondary organic cloud condensation nuclei (CCN)? Revisiting β -caryophyllene CCN, *Atmos. Chem. Phys.*, 12, 8377-8388, doi:10.5194/acp-12-8377-2012, 2012.

Varutbangkul, V., Brechtel, F. J., Bahreini, R., Ng, N. L., Keywood, M. D., Kroll, J. H., Flagan, R. C., Seinfeld, J. H., Lee, A., and Goldstein, A. H.: Hygroscopicity of secondary organic aerosols formed by oxidation of cycloalkenes, monoterpenes, sesquiterpenes, and related compounds, *Atmos. Chem. Phys.*, 6, 2367– 2388, doi:10.5194/acp-6-2367-2006, 2006 .

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 12, 20745, 2012.

ACPD

12, C8112–C8115, 2012

Interactive
Comment

[Full Screen / Esc](#)

[Printer-friendly Version](#)

[Interactive Discussion](#)

[Discussion Paper](#)

