Atmos. Chem. Phys. Discuss., 8, S5426–S5429, 2008
www.atmos-chem-phys-discuss.net/8/S5426/2008/

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

Received and published: 31 July 2008

Presented here are results from a handful of b-caryophyllene oxidation experiments designed to characterize the water-uptake characteristics of the resulting SOA. The approach of combining modeling with experimental measurements gave unique information about CCN activity, growth kinetics, and volatility. It is clear that the results and implications of observed CCN trends have been carefully considered, and discussion of observed phenomena is thorough, plausible, and supported by the data. Nonetheless, the limited number of experiments makes any sweeping conclusions about the nature of sesquiterpene oxidation (and the resulting SOA) rather tenuous, and language in the results section should reflect this uncertainty.

This reviewer agrees with reviewer #1, in that the manuscript would be significantly improved by a more detailed description of the experimental plan and objectives (i.e. what was the purpose of each experiment, how were instruments chosen for each experiment, why was the specific progression in experiments chosen, etc.) As much of this information as is appropriate should be included in an enhanced version of table 1.

One concern is that it was not clear to me which experiment(s) included filter sampling. I gather that a filter sample was only obtained during one experiment. Table 1 indicates that filter collection occurred during experiment 3, in which no OH was present. The text (p. 10113, lines 15-17), however, indicates that the filter was collected during an experiment in which oxidation was by OH and O3, suggesting that it was collected during experiment 2. This is an important distinction, since (as noted on p. 10109, lines 26-27), the nature of water-soluble oxidation products from the O3 and O3 + OH systems are likely different. If filter samples were indeed collected during just one experiment, the conclusions drawn later about WSOC may not necessarily apply to other experiments in the series. The authors found OH-experiments to produce less-volatile SOA, and it is entirely possible that there was significantly different (character and volume fraction) WSOC present in the non-OH experiments. This distinction about when filter samples were taken and the implications and limitations for conclusions about WSOC should be made clearer.

Next, chamber experiments are generally carried out at VOC concentrations above ambient. One consequence is that, at these higher VOC loadings, some semivolatile oxidation products that might otherwise remain predominately in the gas phase will partition to the aerosol phase as SOA. The presence of this more volatile SOA can significantly alter hygroscopic properties (Duplissy et al. GRL 2008). One would expect this more volatile fraction to be less polar, and therefore less hygroscopic. This is the opposite of what is observed in the present study, with more volatile species appearing to be more hygroscopic. Oligomerization is briefly mentioned in the manuscript, but it is not considered a reasonable explanation for the observed trends, nor is a real expla-

ACPD

8, S5426–S5429, 2008

Interactive Comment

nation for the trends advanced. While the reviewer feels that this finding is inherently valuable and understands that a thorough explanation would require much more detailed experiments, one is left without a compelling reason to believe that semivolatile hygroscopic material fully explains the observed discrepancies between the two instruments. For example, can one distinguish any changes in MW as time progresses? If not, the authors may more firmly rule out oligomerization.

The findings about semivolatility of the hygroscopic components, and the implications for measurements of CCN, are important. This is perhaps the strongest part of the paper, and the authors may consider emphasizing these findings further. Specifically, these results are significant for field measurements of CCN. It would be of value to emphasize the error that could be introduced by measuring CCN at elevated temperatures in the field. This is touched on (p. 10126, lines 23-24), but I envision a line reading something like: "If the volatility observed here is applicable to the real atmosphere, it is possible that CCN measurements made at 10 degrees above ambient could introduce an overestimation in CCN activity on the order of ____%". This inclusion certainly isn't necessary, but would be nice for framing the significance of this study's volatility findings.

Overall, the rest of the conclusions are nicely presented and explained (particularly the kinetic arguments).

One other minor suggestion is that the figures should include legends wherever possible, as opposed to descriptions of marker shapes and colors in the caption. It can be hard to move back and forth between text and figure, while at the same time trying to determine what the data show. This may be an issue of personal preference, but I think it would make the figures easier to follow.

Duplissy, J. et al., Cloud forming potential of secondary organic aerosol under near atmospheric conditions, Geophys. Res. Lett., 35, L03818, doi: 10.1029/2007GL031075, 2008.

ACPD

8, S5426–S5429, 2008

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 10105, 2008.

ACPD

8, S5426–S5429, 2008

Interactive Comment