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Atmospheric
Chemistry
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Discussions

Interactive comment on “Increased cloud activation potential of secondary organic aerosol for atmospheric mass loadings” *by* S. M. King et al.

S. M. King et al.

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We thank Referee #2 for the positive and insightful comments, which are shown below in bold. Author responses are shown in normal font, and any modifications to the manuscript are presented in italics.

The authors of the manuscript address an important concern in the prediction of organic aerosol to act as CCN: that the composition of semivolatile organic aerosol changes as the partitioning of the gases and condensed materials with the mass concentration. This is important because the much of our knowledge about organic CCN has been taken from laboratory experiments, which usually require high aerosol concentrations due to instrumental limitations. The authors

overcome this issue by generating the aerosol in a constant-flow mode, so that while the total aerosol mass concentration is at an atmospherically-relevant concentration, the aerosol mass concentration is also maintained to provide enough time for signal-to-noise averaging. The authors find that as the aerosol mass concentration decreases, the aerosol becomes more CCN active in comparison to predicted values. The authors suggest that this is due to a decrease in the surface tension of the aerosol at lower mass concentrations.

Overall, the manuscript is written well and concise. The figures are clear and appropriate. The manuscript represents an important step forward for understanding how to connect laboratory measurements with atmospheric observations and the manuscript is a worthy publication.

Regarding the conclusion, that a decrease in surface tension is reasonable mechanism for changes in CCN activation with mass concentration: is it also chemically reasonable that the shift in the composition of CCN at low mass concentrations leads to species with lower surface tension? The authors cite the work of Grieshop et al. and Shilling et al. which are evidence of shifts in the overall chemical composition at low mass concentrations. If the authors do not agree that surface partitioning causes the decrease in surface tension, the authors should postulate which chemical species could this change in surface tension. And are there known chemical species with lower surface tension that are consistent with the increase in %m/z 44? Is it possible to measure the surface tension of the condensed phase at various mass loadings by traditional means as a confirmation? However this is addressed, authors should make an attempt to connect the observation of a decrease in surface tension with a plausible mechanism for differences in the species distribution (either chemical or physical location within the particle) of the condensed phase.

We agree that the emphasis currently placed on surface tension as the explanation for

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the observed increase in CCN activity may lead to further questions that are beyond the scope of this study. It should be possible to measure the surface tension of the condensed phase at various mass loadings, but these experiments were not conducted as part of this study. Other studies have shown a reduction in surface tension due to certain carboxylic acids (Tuckermann, 2007 and references therein); however, the acids studied are only a few of the large number of condensed-phase organic compounds resulting from α-pinene oxidation.

To provide the requested clarification, the text is modified as follows and as presented in the responses to other comments on this manuscript:

[Page 1685, lines 5+] "Signal at *m/z* 44 includes fragments of oxo- and di-carboxylic acids, and the increase for lower organic mass loadings plausibly indicates a corresponding increase in the mass fraction of these oxygenated, polar compounds, representing the types of organic molecules normally considered CCN active. Possible causes for the increase in oxygenated fraction at low loadings are discussed in Shilling et al. (2009). *Notably, Capouet and Müller (2006) report that the volatility of carboxylic acids may be amongst the lowest of all the products of α-pinene ozonolysis."*

[Page 1686, end of text]: *"Other factors, such as decreased molecular weight, increased density, or increased van't Hoff factor, can contribute to the explanation but are quantitatively insufficient as the full explanation. As a hypothesis, an enrichment of carboxylic acids at low loadings, as suggested by Capouet and Müller (2006) and as consistent with the mass spectra reported by Shilling et al. (2009) for these experiments, anticipates a decrease in surface tension because carboxylic acids have been shown to reduce surface tension (Shulman et al., 1996; Hyvärinen et al., 2006; Topping et al., 2007; Tuckermann, 2007)."*

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 1669, 2009.

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