

Interactive
Comment

Interactive comment on “Loading-dependent elemental composition of α -pinene SOA particles” by J. E. Shilling et al.

J. E. Shilling et al.

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We thank the reviewer for their helpful comments and suggestions. We have made changes in the final ACP paper based on the reviewers comments as described below.

Reviewer Comment: It was discussed in the introduction that SOA was substantially underestimated in models, which usually use parameterizations that were derived based on yields determined in chamber studies conducted under high SOA loadings. In this study, Fig. 4 clearly shows an increase trend of yields vs. SOA particle mass loadings, which may suggest that the model prediction of SOA will be lower even more if yields from the low OA loading experiments are used. Comments on this point will be interesting.

Response: The experimental parameterizations used in models are indeed based on

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data collected at high SOA loadings. However, the yield measured at high loading is not directly used in the models; rather semi-volatile partitioning theory is employed to extrapolate the yields to the appropriate atmospheric loading (Chung and Seinfeld, 2002). Donahue and coworkers have pointed out that extrapolation of high-loading data underpredicts yield relative to yield directly measured at lower loadings (Presto and Donahue, 2006). Yield measurements from this study are described and discussed in a separate publication (Shilling et al., 2008), but we generally measure yields that are larger than those reported in previous literature studies (Griffin et al., 1999; Cocker et al., 2001; Presto and Donahue, 2006; Pathak et al., 2007). Thus, use of our measured yields in models would increase predictions of SOA loading. Further details and discussions of yield are available in our ACP paper at the following url: <http://www.atmos-chem-phys.net/8/2073/2008/acp-8-2073-2008.pdf>

Reviewer Comment: Regarding the discussions on the similarities in the mass spectra, it needs to be stressed that the SOA from this study was produced from one VOC while ambient OOA/SOA is the product of hundreds or thousands of VOCs involving more reactions. In addition, ambient OOA/SOA is probably more aged than the SOA mixtures obtained from this study. It will be interesting to see how the SOA mass spectra change as a function of reaction time, for instance the evolution pattern of the 4 base sets similar to that displayed in Fig. 4.

Response: The reviewer is correct; atmospheric SOA probably contains many more compounds and is likely to be more aged than the SOA produced in our chamber. Similarity of the mass spectra does not indicate that the respective SOAs samples contain identical compounds, but it does imply that the relative distribution of functional groups is similar in both samples. We have clarified this point in the text. We also agree that the aging of SOA is an interesting topic, but, unfortunately, we currently have no data on this subject.

Updated text: The implication is that the electron-impact fragmentation patterns of the organic material in the two types of particles are similar, suggesting that the relative

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distributions of the organic functional groups are also similar. Nevertheless, within this similarity there are undoubtedly important differences that are not revealed by the comparison shown in Figure 6 because of the myriad molecular assemblies possible from similar distributions of functional groups. Such different assemblies can be expected on the basis of the large number of VOC precursors involved in atmospheric SOA production compared to the single precursor employed in this study.

Reviewer Comment: The tag lines of the peak labels in Fig. 1 and 2 are sometimes hard to differentiate from the peaks. Please either remove the tag lines or make them more distinguishable from the peaks.

Response: We thank the reviewer for pointing out his difficulty. To improve the figure clarity, we have changed Figures 1 and 6 so that the peaks are a different color from the tag lines and have increased the spacing from tag lines to peaks in Figures 1, 2, and 6.

Reviewer Comment: Why Fig. 5 shows the effective density data only up to $40 \mu\text{g m}^{-3}$ of SOA loading? What at the densities for SOA at higher mass loadings?

Response: We are unfortunately unable to determine the SOA density at loadings above $40 \mu\text{g m}^{-3}$. During experiments, the scanning range of the SMPS was set to 10 to 495 nm. At the highest loadings, the mode diameter of the SOA particles was larger than the upper limit of the SMPS scan; therefore, we can not directly determine particle density in these experiments. We have noted this point in the text. Other researchers have measured particle densities of 1.19 to 1.6 g cm^{-3} for this system (Bahreini et al., 2005; Kostenidou et al., 2007; Song et al., 2007), in agreement with our measurements at loadings above ca. $15 \mu\text{g m}^{-3}$.

Updated Text: Densities for higher loadings could not be determined from our measurements because the mode of the particle size distribution exceeded the limit of 495 nm for our operating conditions of the SMPS.

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Reviewer Comment: No H₂O peaks are shown in Fig. 1, 2 and 6i. Were they included in O/C ratio analysis?

Response: H₂O peaks were included in the O/C ratio analysis. Because our study was conducted at 40% RH, we are unable to conclusively separate the contribution of particle-phase water and organics to the H₂O peaks. H-TDMA studies measured growth factors of 1.02 to 1.04 at 50% RH (Cocker et al., 2001; Varutbangkul et al., 2006). In light of these experiments, a small amount particle-phase water is expected to be associated with the SOA under our experimental conditions of 40% RH. Therefore we follow the method of Aiken et al. in assigning the signal at m/z 18, 17, and 16 to water produced by dehydration of organic species (Aiken et al., 2008).

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