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Loading-dependent elemental composition of α -pinene SOA particles

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Abstract. The chemical composition of secondary organic aerosol (SOA) particles, formed by the dark ozonolysis of α -pinene, was characterized by a high-resolution time-of-flight aerosol mass spectrometer. The experiments were conducted using a continuous-flow chamber, allowing the particle mass loading and chemical composition to be maintained for several days. The organic portion of the particle mass loading was varied from 0.5 to $>140 \mu\text{g}/\text{m}^3$ by adjusting the concentration of reacted α -pinene from 0.9 to 91.1 ppbv. The mass spectra of the organic material changed with loading. For loadings below $5 \mu\text{g}/\text{m}^3$ the unit-mass-resolution m/z 44 (CO_2^+) signal intensity exceeded that of m/z 43 (predominantly $\text{C}_2\text{H}_3\text{O}^+$), suggesting more oxygenated organic material at lower loadings. The composition varied more for lower loadings (0.5 to $15 \mu\text{g}/\text{m}^3$) compared to higher loadings (15 to $>140 \mu\text{g}/\text{m}^3$). The high-resolution mass spectra showed that from >140 to $0.5 \mu\text{g}/\text{m}^3$ the mass percentage of fragments containing carbon and oxygen ($\text{C}_x\text{H}_y\text{O}_z^+$) monotonically increased from 48% to 54%. Correspondingly, the mass percentage of fragments representing C_xH_y^+ decreased from 52% to 46%, and the atomic oxygen-to-carbon ratio increased from 0.29 to 0.45. The atomic ratios were accurately parameterized by a four-product basis set of decadal volatility (viz. 0.1, 1.0, 10, $100 \mu\text{g}/\text{m}^3$) employing products having empirical formulas of $\text{C}_1\text{H}_{1.32}\text{O}_{0.48}$, $\text{C}_1\text{H}_{1.36}\text{O}_{0.39}$, $\text{C}_1\text{H}_{1.57}\text{O}_{0.24}$, and $\text{C}_1\text{H}_{1.76}\text{O}_{0.14}$. These findings suggest considerable caution is warranted in the extrapolation of laboratory results that were obtained under conditions of relatively high loading (i.e., $>15 \mu\text{g}/\text{m}^3$) to modeling applications relevant to the atmosphere, for which loadings of 0.1 to $20 \mu\text{g}/\text{m}^3$ are typical. For the lowest loadings, the particle mass spectra resembled observations

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