Atmospheric Chemistry and Physics
An Interactive Open Access Journal of the European Geosciences Union

Atmos. Chem. Phys., 9, 771-782, 2009
www.atmos-chem-phys.net/9/771/2009/

Loading-dependent elemental composition of α-pinene SOA particles

J. E. Shilling[1,*], Q. Chen[1], S. M. King[1], T. Rosenoern[1], J. H. Kroll[2], D. R. Worsnop[2], P. F. DeCarlo[3,4,], A. C. Aiken[4,5], D. Sueper[2,4,5], J. L. Jimenez[4,5], and S. T. Martin[1,6]**

[1]School of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138, USA
[2]Aerodyne Research, Inc., Billerica, MA 08121, USA
[3]Department of Atmospheric and Oceanic Sciences, University of Colorado, Boulder, CO 80309, USA
[4]Cooperative Institute for Research in the Environmental Sciences (CIRES), Univ. of Colorado, Boulder, CO 80309, USA
[5]Department of Chemistry and Biochemistry, University of Colorado, Boulder, CO 80309, USA
[6]Department of Earth and Planetary Sciences, Harvard University, Cambridge, MA 02138, USA
[*]now at: Pacific Northwest National Laboratory, Atmospheric Sciences and Global Change Division, Richland, WA 99352, USA
[**]now at: Paul Scherrer Institut, Laboratory of Atmospheric Chemistry, Villigen-PSI, 5232, Switzerland

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 μg/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 μg/m^3 the unit-mass-resolution *m/z* 44 (CO_2^+) signal intensity exceeded that of *m/z* 43 (predominantly $C_2H_3O^+$), suggesting more oxygenated organic material at lower loadings. The composition varied more for lower loadings (0.5 to 15 μg/m^3) compared to higher loadings (15 to >140 μg/m^3). The high-resolution mass spectra showed that from >140 to 0.5 μg/m^3 the mass percentage of fragments containing carbon and oxygen ($C_xH_yO_z^+$) 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 μg/m^3) employing products having empirical formulas of $C_1H_{1.32}O_{0.48}$, $C_1H_{1.36}O_{0.39}$, $C_1H_{1.57}O_{0.24}$, and $C_1H_{1.76}O_{0.14}$. 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 μg/m^3) to modeling applications relevant to the atmosphere, for which loadings of 0.1 to 20 μg/m^3 are typical. For the lowest loadings, the particle mass spectra resembled observations

reported in the literature for some atmospheric particles.

Final Revised Paper (PDF, 1793 KB) Supplement (1104 KB) Discussion Paper (ACPD)

Citation: Shilling, J. E., Chen, Q., King, S. M., Rosenoern, T., Kroll, J. H., Worsnop, D. R., DeCarlo, P. F., Aiken, A. C., Sueper, D., Jimenez, J. L., and Martin, S. T.: Loading-dependent elemental composition of α-pinene SOA particles, Atmos. Chem. Phys., 9, 771-782, 2009. Bibtex EndNote Reference Manager