Comparison of Cloud Condensation Nuclei Activity of Secondary Organic Aerosols Derived from Hygroscopic Growth Factor And Direct CCN Measurements

A.T. Lambe[1,2], A.T. Ahern[2], P. Massoli[2], L. R. Williams[2], W.H. Brune[3], M. Ehn[4], P. Miettenen[5], E.S. Cross[1], T.B. Onasch[2], J.T. Jayne[2], M. Kulmala[4], A. Laaksonnen[5], D.R. Worsnop[2,4,5], and P. Davidovits[1]

[1]Department of Chemistry, Boston College, Chestnut Hill, MA, USA
[2]Aerodyne Research Inc., Billerica, MA, USA
[3]Dept. of Meteorology and Atmospheric Sciences, The Pennsylvania State University, University Park, PA. USA
[4]Division of Atmospheric Sciences, University of Helsinki, Helsinki, Finland
[5]Department of Physics, University of Kuopio, Kuopio, Finland

Introduction and Methods

• Hygroscopic growth factors (HGF) and cloud condensation nuclei (CCN) activity were measured for secondary organic aerosols (SOA) generated from a-pinene and m-xylene. These species were used as surrogates for gas-phase biogenic and anthropogenic emissions.

• Precursors were oxidized in a laboratory aerosol flow reactor as a function of OH radical concentrations varied from 4×10^{8} to 1×10^{10} molec cm^{-3}.

• Corresponding OH exposures ranged from 4.8×10^{10} to 1.2×10^{12} molec cm^{-3} s, or 0.5-14 days' atmospheric exposure at an OH concentration of 1×10^{6} molec cm^{-3}.

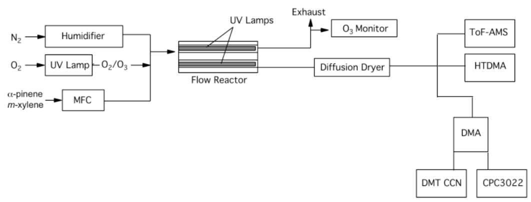


Figure 1. Experiment schematic showing aerosol flow reactor and particle characterization experiments. OH radicals were generated from the $O(^1D) + H_2O$ reaction, with $O(^1D)$ produced via O_3 photolysis and H_2O introduced by passing N_2 through a humidifier. α-pinene or m-xylene were introduced to the reactor using compressed gas mixtures in N_2 and were regulated with a mass-flow controller. Particle physical and chemical properties were chacterized with an HTDMA, a Droplet Measurement Technologies CCN counter, and an Aerodyne ToF-AMS.

• An Aerodyne time-of-flight aerosol mass spectrometer measured aerosol chemical composition.

• A hygroscopic tandem differential mobility analyzer (HTDMA) measured growth factors by passing dry aerosols through water-humidified air (90% RH) and measuring the wet-to-dry particle diameter ratio.

• A CCN instrument (Droplet Measurement Technologies) passed dry aerosols through water-humidified air (0.1-1% supersaturation) and measuring CCN with an optical particle counter.

Hygroscopic Growth Factor and CCN Measurements

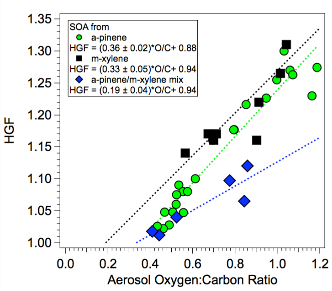
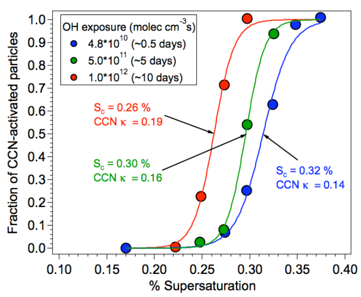


Figure 2. HGF for SOA generated from α-pinene and m-xylene. As aerosol oxygen-to-carbon ratio increases from OH exposure, particles are proggressively more water-soluble and have higher HGF. Best-fit lines are added to guide the eye.



Figures 3. Representative CCN activation curves for a-pinene SOA at different OH exposures. For a given supersaturation, a larger fraction of particles activate to form CCN at higher OH exposures. 50% of particles activate to form CCN at the critical supersaturation S_c.

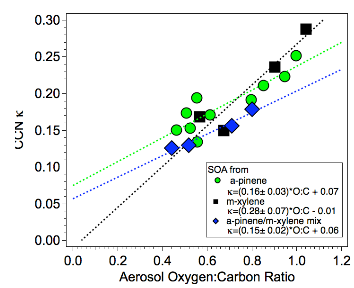


Figure 4. CCN-derived hygroscopicity paramter k (see Equation 1 below) for α-pinene and m-xylene SOA. As aerosol oxygen-to-carbon ratio increases, surface tension decreases, enhancing CCN activity. Best-fit lines are added to guide the eye.

Conclusions

• HGF and CCN activity for α-pinene and m-xylene SOA increased with OH exposure and were linearly related to the aerosol oxygen:carbon ratio.

• HGF- and CCN-derived κ values were linearly related but in poor agreement.

• HGF measurements at subsaturated conditions cannot reliably extrapolate CCN activity at supersaturated conditions.

Comparison of HGF- and CCN-Derived κ

• For a given species, the hygroscopicity parameter κ (Petters and Kreidenweiss, 2007) relates dry particle diameter (D_d) to critical supersaturation and can be applied to direct CCN measurements made at supersaturated conditions:

$$CCN\ \ \kappa = \frac{4A^3}{27D_d^3 \ln^2 S_c}; \qquad A = \frac{4\sigma_w M_w}{RT\rho_w} \qquad [1]$$

• Where σ_w, ρ_w, and M_w are the surface tension, density, and molecular weight of water. k can also be extrapolated from HGF measurements at subsaturated conditions (Petters and Kreidenweiss):

$$HGF\ \ \kappa = 1 + \frac{1}{RH} \times \frac{HGF^3 - 1}{e^{\left(\frac{A}{D_d \times HGF}\right)}} - HGF^3 \qquad [2]$$

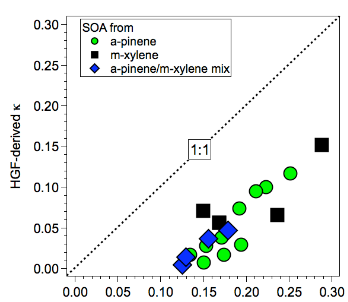


Figure 5. HGF- and CCN-derived κ for α-pinene and m-xylene SOA.

Acknowledgements

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References

Ehn, M., Petaja, T. et al. *Atmos. Chem. Phys.*, 7, 211-222, **2007**.

Petters, M.D. and Kreidenweiss, S.M.. *Atmos. Chem. Phys.*, 7, 1961–1971, **2007**.