Laboratory Studies of Carbonaceous Aerosols: Characterization and Atmospheric Processing

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Introduction and Methods

Carbonaceous aerosols affect climate by direct (e.g. absorption/scattering) and indirect (e.g. CCN/IN formation) processes.

Our current work involves the application of new laboratory techniques for generating particles as complex surrogates of ambient carbonaceous aerosols: secondary organic aerosol (SOA), oxidized primary organic aerosol (OPOA), and soot particles. A Potential Aerosol Mass (PAM) flow reactor simulates ~1 to 15 days' equivalent atmospheric processing (Lambe et al., 2011). Instrumentation is used to characterize particle chemical, physical, and optical properties over a range of oxidant exposures that is unattainable by smog chamber techniques and that match ambient observations.

Our goals are to identify correlations between associated chemical, physical, and optical properties that may help explain field measurements and enable more accurate climate modeling.

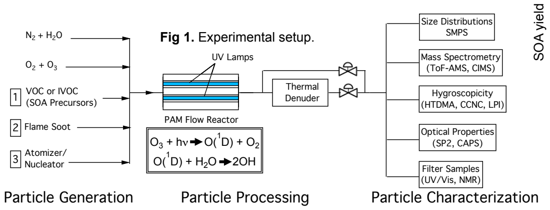


Fig 1. Experimental setup.

SOA Chemistry

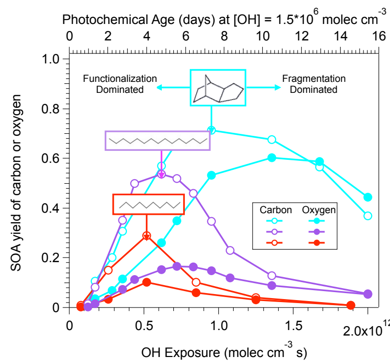
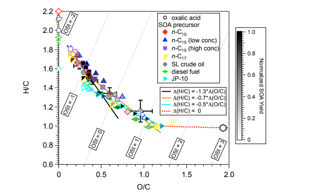


Fig 2. SOA yields of carbon and oxygen generated from oxidation of n-C_{10}, n-C_{15}, and JP-10 as a function of OH exposure in the PAM reactor (Lambe et al., 2012).

SOA Aging



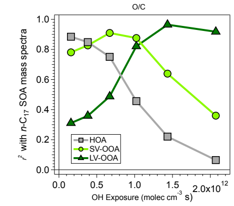
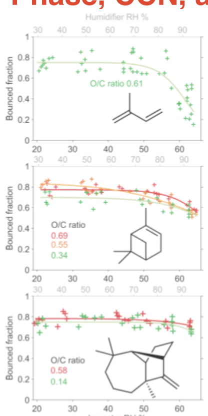
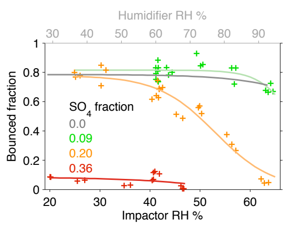


Fig. 3 (top). Van Krevelen diagram of laboratory SOA. **Fig. 4 (bottom).** Correlation of laboratory SOA with ambient HOA, SV-OOA, LV-OOA factors (Lambe et al., 2011, 2012).

Phase, CCN, and IN of SOA and OPOA





Figs. 5 (left), 6 (top). "Bounced fraction" (BF) of biogenic SOA particles measured with low pressure impactor technique (Saukko et al., 2012).

Liquids: BF~0; solids: BF~0.8. "Glassy" solid SOA at low RH become liquid-like at high RH or upon mixing with sulfate. Particle phase state impacts chemistry and CCN/IN formation processes.

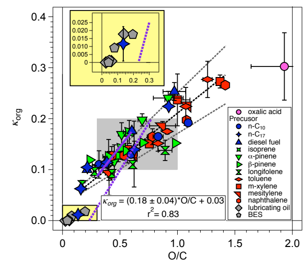


Fig. 7. CCN κ_{org} as a function of oxygen-to-carbon (O/C) ratio (Massoli et al., 2010; Lambe et al., 2011). Grey: typical ambient measurements. Purple lines: Chang et al (ACP, 2010; solid) and Jimenez et al. (Science, 2009; dashed) parameterizations of ambient OOA.

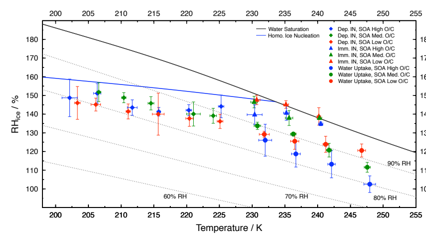


Fig. 8. RH threshold as a function of temperature for onset of ice nucleation via water uptake, immersion freezing and deposition freezing of laboratory-generated naphthalene SOA with low, medium, and high O/C ratios of 0.27, 0.54, and 1.0 respectively (Wang et al., 2012).

Black Carbon Aging and Optics

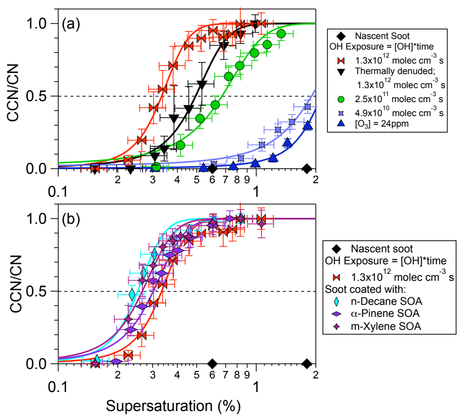


Fig. 9. CCN curves of soot particles (a) heterogenously oxidized by OH radicals in the PAM reactor and (b) oxidized and coated with SOA.

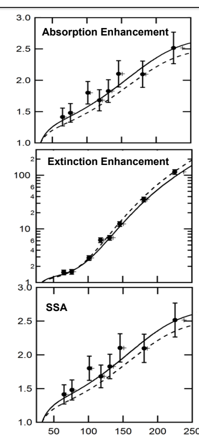


Fig. 10. Measured (symbols) and calculated (lines) absorption enhancement, extinction enhancement and single scatter albedo for $d_{p,VED}$ = 33 nm nascent soot particles coated with DOS (Cappa et al., 2012).

Ambient SP-AMS Measurements

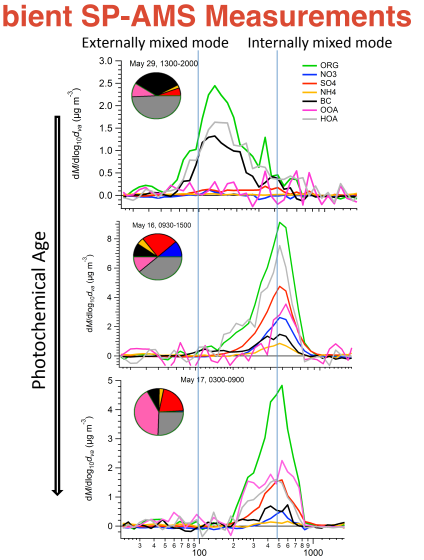


Fig. 11. Chemically-resolved mass distributions of black carbon particles measured by the SP-AMS during CalNEX 2010 for different outflow events from the LA Basin, showing increased NR-PM, O/C, and internal mixing with photochemical aging.

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References: J.L. Jimenez et al., *Science*, 326, 1525, **2009**; R.Y.-W. Chang et al., ACP, 10, 5047–5064, **2010**; P. Massoli et al., *GRL*, 37, L24801, **2010**; A.T. Lambe et al., *AMT*, 4, 445-461, **2011**; A.T. Lambe et al., *ACP*, 11, 8913-8928, **2011**; E. Saukko et al., *ACPD*, 12, 4447-4476, **2012**; A.T. Lambe et al., **2012**, Submitted; B. Wang et al., **2012**, Submitted; C.D. Cappa et. al, **2012**, Submitted.