Laboratory Studies on the Atmospheric Aging Processes of Combustion Soot

A.T. Lambe[1,2], T.B. Onasch[1,2], A.T. Ahern[1,2], J. P. Wright[1], D.R. Croasdale[1], D.R. Worsnop[2], and P. Davidovits[1]

[1]Boston College, [2]Aerodyne Research, Inc.

Introduction and Methods

- The climate forcing of particles containing absorbing material such as refractory black carbon (rBC) is complex because of the potential for positive and negative radiative forcing effects.
- To investigate the effect of chemical composition on CCN activity of rBC-containing particles, laboratory experiments were conducted in which ethylene flame soot particles were generated and exposed to OH radicals in a Potential Aerosol Mass (PAM) flow reactor.
- Our hypothesis is that OH oxidation of the soot will convert hydrophobic organic coatings to CCN-active hydrophilic molecules such as carboxylic acids.
- In a separate set of experiments, SOA was condensed on the soot particles and the enhanced CCN activity was measured.
- Implications of atmospheric aging of rBC-containing particles are discussed.

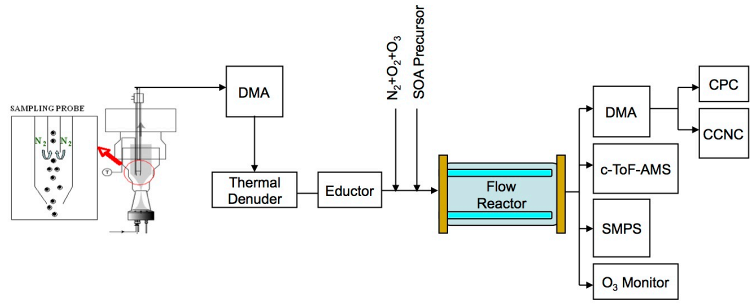

Figure 1. Experimental setup. Soot particles were generated using a flat burner flame (Cross et al., 2010). A mini-eductor pulls soot particles from the tip of the flame. Soot was exposed to OH radicals in a Potential Aerosol Mass (PAM) reactor (Lambe et al., 2011a) over OH exposures equivalent to approximately one day to ten days of atmospheric oxidation

Soot Aging

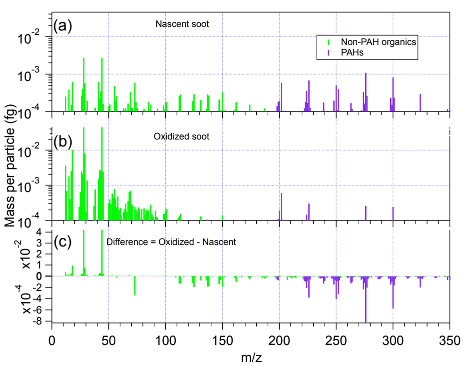

Figure 2. c-ToF-AMS spectra of nascent soot and oxidized soot following exposure to OH radicals in flow reactor.

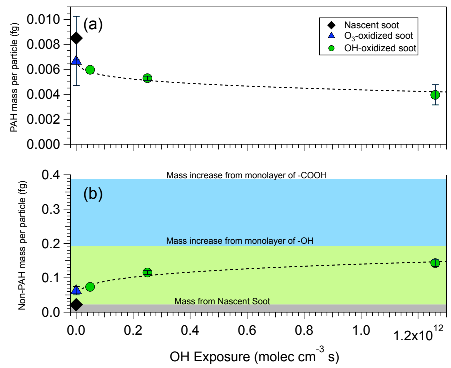

Figure 3. Mass of (a) PAH and (b) non-PAH organics as a function of OH exposure. Colored bars represent upper limits of calculated mass increase from addition of -OH and -COOH functional groups following heterogeneous oxidation of a surface organic monolayer.

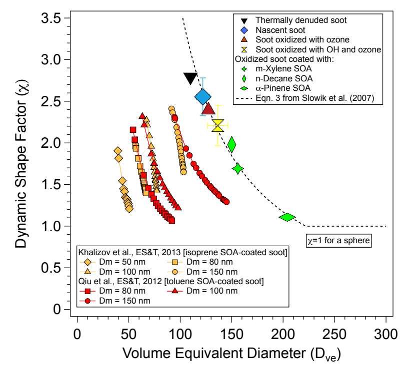

Figure 4. Shape factor of aged soot (D_m = 200 nm) as a function of volume equivalent diameter.

SOA CCN Properties

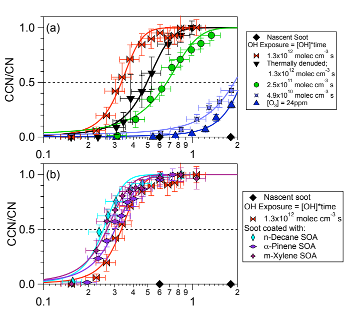

Figure 5. CCN activation curves of soot particles following (a) O_3 exposure, OH exposure, thermal denuding and (b) SOA condensation.

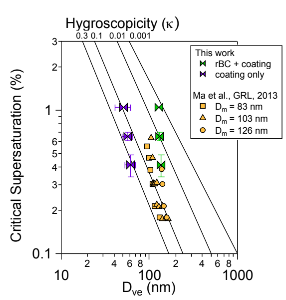

Figure 6. CCN critical supersaturation as a function of D_{ve} for aged soot.

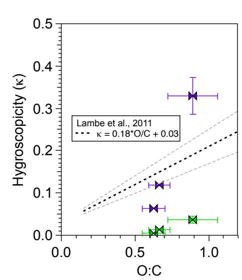

Figure 7. CCN κ-values as a function of O/C of aged soot. Dashed line corresponds to κ_{org} = (0.18 ± 0.04) × O/C + 0.03 parameterization from Lambe et al. (2011b).

Summary

- Heterogenous oxidation of soot leads to increasing CCN activation as a function of OH exposure.
- Coating soot with SOA changes particle morphology and increases CCN activity, consistent with previous studies (Qiu et al., 2012, Khalizov et al., 2013).
- Many climate models simulate aging of BC using overly simplified parameterizations (e.g. Cooke et al., 1999) that do not represent recent field measurements (Moffet & Prather 2009; Schwarz et al. 2010). Our results can be used as inputs to models to refine CCN predictions of BC-containing particles as a function of atmospheric age and SOA coating thickness.

Acknowledgements: DoE ASR #DE-SC0006980, #DE-FG02-05ER63995; NSF Atmospheric Chemistry Program #AGS-0904292, #ATM-0854916, #ATM-0837913, #AGS-0855135.

References: W.F. Cooke et al., *J. Geophys. Res.*, 104, 22137-22162, **1999**; R. Moffet and K. Prather, *Proc. Natl. Acad. Sci.*, 106, 11872-11877, **2009**; J.P. Schwarz et al., *Geophys. Res. Lett.*, 37, L18812, **2010**; E.S. Cross et al., *Aerosol Sci. Technol.*, 44, 592-611, **2010**; A.T. Lambe et al., *Atmos. Meas. Tech.*, 4, 445-461, **2011a**; A.T. Lambe et al., *Atmos. Chem. Phys.*, 11, 8913-8928, **2011b**; C. Qiu et al, *Environ Sci. Technol.*, 46, 9464-9472, **2012**; A. F. Khalizov et al., *Environ Sci. Technol.*, 47, 2254-2263, **2013**; Y. Ma et al., *Geophys. Res. Lett.*, 40, 6293-6297, **2013**.