Aerosol mass spectrometry of particles containing refractory carbon

T.B. Onasch[1,2], E. Fortner[1], P. Massoli[1], L. R. Williams[1], A. Lambe[1,2],
A. M. Trimborn[3], John T. Jayne[1], Paul Davidovitz[2], Douglas R. Worsnop[1]

1 Aerodyne Research, Inc.; 2 Boston College; 3 AeroMegt GmbH

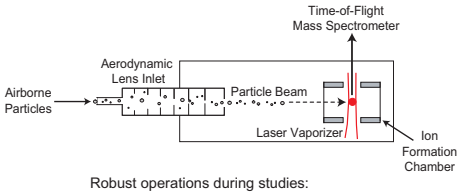
Introduction

Refractory black carbon (rBC) containing particles, including energy-derived combustion particles, can affect climate directly by absorbing (and scattering) incoming light depending on particle composition and morphology, and indirectly by influencing cloud droplet formation and lifetimes. The uncertainty in the climatic effects of rBC is driven, in part, by several important factors: poorly understood formation, poorly quantified atmospheric processing (e.g., hydrophobic into hydrophilic), and limited information on the UV-VIS-IR optical properties. These uncertainties are complicated by the lack of measurement techniques that can unambiguously measure properties of refractory carbon containing particles. To help address these uncertainties, we have developed a Soot Particle Aerosol Mass Spectrometer (SP-AMS) instrument capable of measuring the mass, size, and chemical composition of rBC particles.

Provides real-time *in situ* measurements of
- rBC carbon ion mass spectra which may be related to underlying carbon structure and bonding
- coat-to-core mass ratios for understanding optical properties
- mass and chemical composition of all nonrefractory and rBC components for source characterization

SP-AMS Instrument

- Intracavity CW Nd:YAG laser vaporizer (λ=1064 nm) for absorbing refractory Black Carbon particles
- Resistively heated tungsten vaporizer for nonabsorbing particles
- Electron ionization (70eV)
- High Resolution Time-of-Flight Mass Spectrometer
- Measures size-resolved, chemical composition of nonrefractory and refractory Black Carbon particles



Robust operations during studies:
- in laboratory (BC2, BC3)
- at ground sites (FLAME3, CalNex, ClearfLo)
- on R/V Atlantis (CalNex)
- on ARI Mobile Lab (NYC, Mexico)
- on DOE ARM G1 (Biomass Burning 2013)

Acknowledgements

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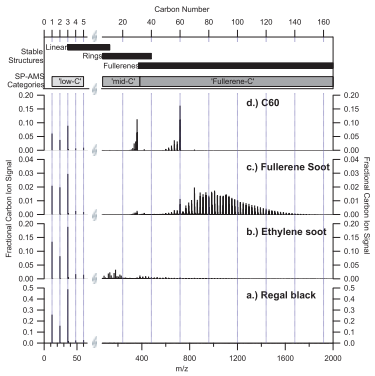
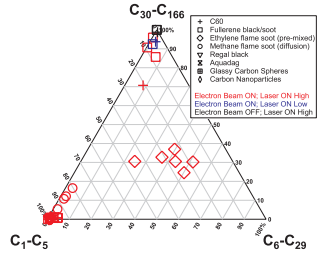
EPA STAR

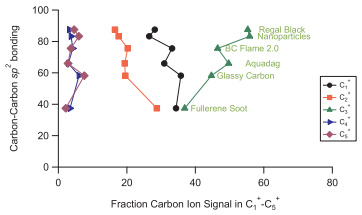
Carbon Ion Mass Spectra: Connection to Structure/Bonding



Normalized positive ion mass spectra for four particle types: (a) Regal black, (b) ethylene flame soot, (c) fullerene soot, and (d) pure C60.

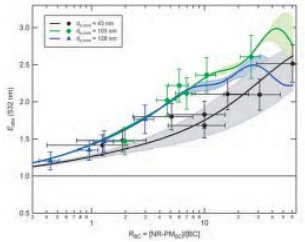


The integrated carbon ion signal fractions for three carbon ion cluster regions: C_1^+ to C_5^+, C_6^+ to C_{29}^+, and C_{30}^+ to C_{166}^+ are displayed in the ternary diagram.

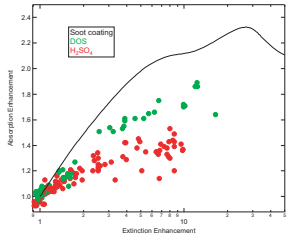


The fraction of carbon ion signal for a given carbon cluster in C_1^+-C_5^+ versus the carbon-carbon sp^2 bonding fraction as measured by XPS for six different rBC particle types.

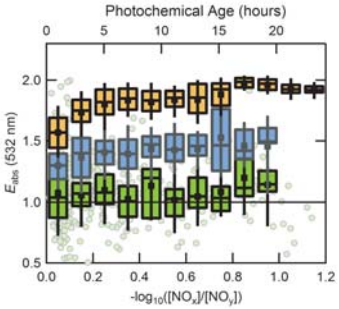
Black Carbon Optical Properties: Lab (BC2, BC3) and Field (CalNex)



Measured (symbols) and calculated (lines) Measured absorption enhancement as a function of coating abundance (which is proportional to the extinction enhancement) for DOS-coated soot during BC2, generated from a McKenna flat flame burner (Cross et al., 2010; Cappa et al., 2012).

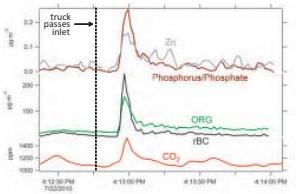


Measured (symbols) and calculated (lines) absorption enhancement as a function of extinction enhancement for DOS- and H2SO4-coated soot generated from an inverted diffusion flame burner. The calculated line uses core-shell Mie theory.

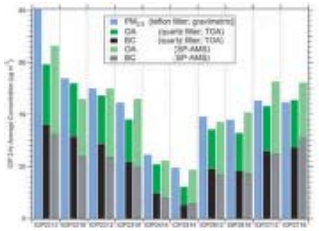


Measured absorption enhancement (green) for ambient BC during the 2010 CalNex campaign as a function of photochemical age, compared with core-shell Mie theory calculations assuming bare cores (orange) and accounting for residual non-BC material (blue) (Cappa et al., 2012).

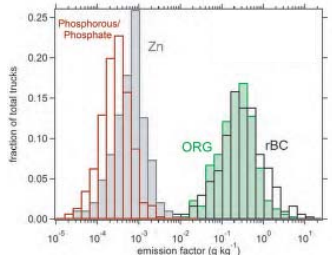
Source Characterization: Caldecott Tunnel Diesel Trucks



SP-AMS mass concentration measurements for an individual truck plume event, including rBC and organics as well as the trace elements Zn and Ph from lubricating oil.



Comparison of average PM2.5, OA, and BC concentrations measured during 2 hr intensive operating periods (IOPs).



Emission factor distributions for major and minor chemical components of exhaust particulate matter from diesel trucks.

Collaborators

UC Davis - C. Cappa and S. Forestieri
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UC Berkeley - R. Harley and T. Dallmann
VT - L. Marr and A. Tiwari
BNL - A. Sedlacek and E. Lewis
DMT - G. Kok and C. Hare