Black Carbon Particle Measurement Studies

Timothy B. Onasch[1,2], Eben S. Cross[1,2], Adam Ahern[1,2], William Wrobel[1], Jay G. Slowik[3], Jason Olfert[4,12], Daniel A. Lack[5,14], Paola Massoli[2,5], Christopher D. Cappa[6], Joshua Schwarz[5], J. Ryan Spackman[5], David W. Fahey[5], Arthur Sedlacek[4], Achim Trimborn[2], John T. Jayne[2], Andrew Freedman[2], Leah R. Williams[2], Nga L. Ng[2], Claudio Mazzoleni[7,13], Manvendra Dubey[7], Benjamin Brem[8], Greg Kok[9], R. Subramanian[9], Steffen Freitag[10], Antony Clarke[10], Dwight Thornhill[11], Linsey Marr[11], Charles E. Kolb[2], Doug R. Worsnop[2], and Paul Davidovits[1]

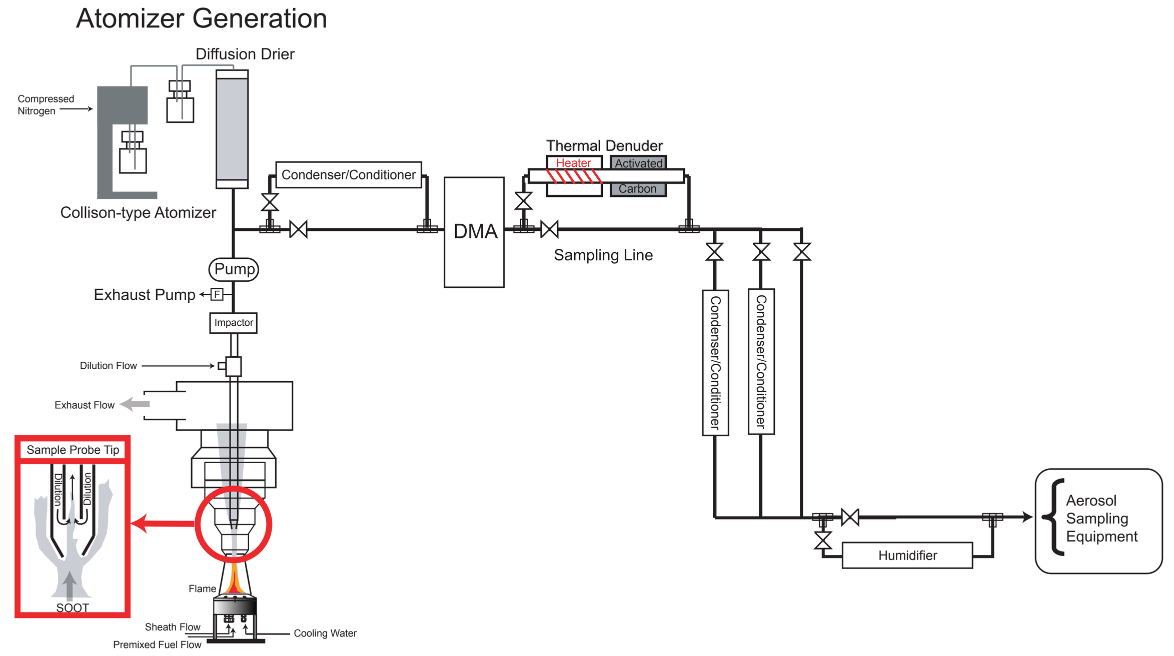
[1]Chemistry Department, Boston College, Chestnut Hill, Massachusetts, USA
[2]Aerodyne Research, Inc., Billerica, Massachusetts, USA
[3]Chemistry Department, University of Toronto, Toronto, CA
[4]Brookhaven National Laboratory, Upton, New York, USA
[5]NOAA Earth System Research Laboratory, Chemical Sciences Division, Boulder, Colorado, USA
[6]Department of Civil and Environmental Engineering, University of California, Davis, California, USA
[7]Los Alamos National Laboratory, Los Alamos, New Mexico, USA
[8]Department of Civil and Environmental Engineering, University of Illinois, Urbana, Illinois, USA
[9]Droplet Measurement Technologies, Boulder, Colorado, USA
[10]Department of Oceanography, University of Hawaii, Honolulu, Hawaii, USA
[11]Department of Civil and Environmental Engineering, Virginia Tech, Blacksburg, Virginia, USA
[12]Department of Mechanical Engineering, University of Alberta, Edmonton, Alberta, Canada
[13]Department of Physics, Michigan Technological University, Houghton, Michigan, USA
[14]CIRES, University of Colorado, Boulder, Colorado, USA

Introduction

- Black Carbon (BC) particles
  - Absorb light efficiently, contributing to radiative forcing
  - Significant anthropogenic sources
- BC particles generated by incomplete combustion
  - Inhomogeneous chemical compositions (including refractory and nonrefractory components)
  - Irregular shapes (agglomerates, mixed-phase)
  - Variable optical properties (scattering and absorption)
- BC particles properties modified by atmospheric processes
  - Coatings of primary combustion products that may evaporate down wind of emission source
  - Secondary organic and inorganic condensates
  - Fates are not well known: coagulation, CCN activity, deposition, etc.
- Need for instrumentation capable of measuring the physical, optical, and chemical properties of BC particles
  - Variable BC properties complicate in situ measurement capabilities
- This work focuses on experiments designed to characterize BC particle properties under controlled laboratory conditions and the intercomparison of current, state-of-the-art instrumentation for measuring BC particle properties
  - Morphological and optical changes of BC particles during coating/denuding experiments
  - Mass-based and optical-based instruments

BC Particle Generation

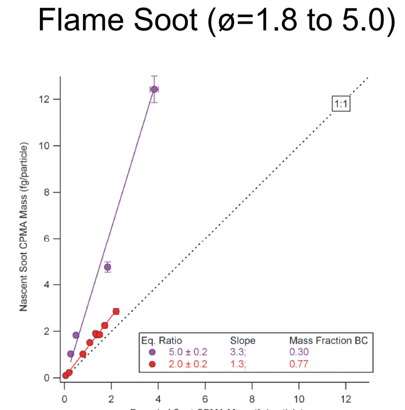
Atomizer Generation



Sooting Flame

BC Particle Types

Pre-mixed Ethylene Flame Soot (ø=1.8 to 5.0)



Atomized BC particles

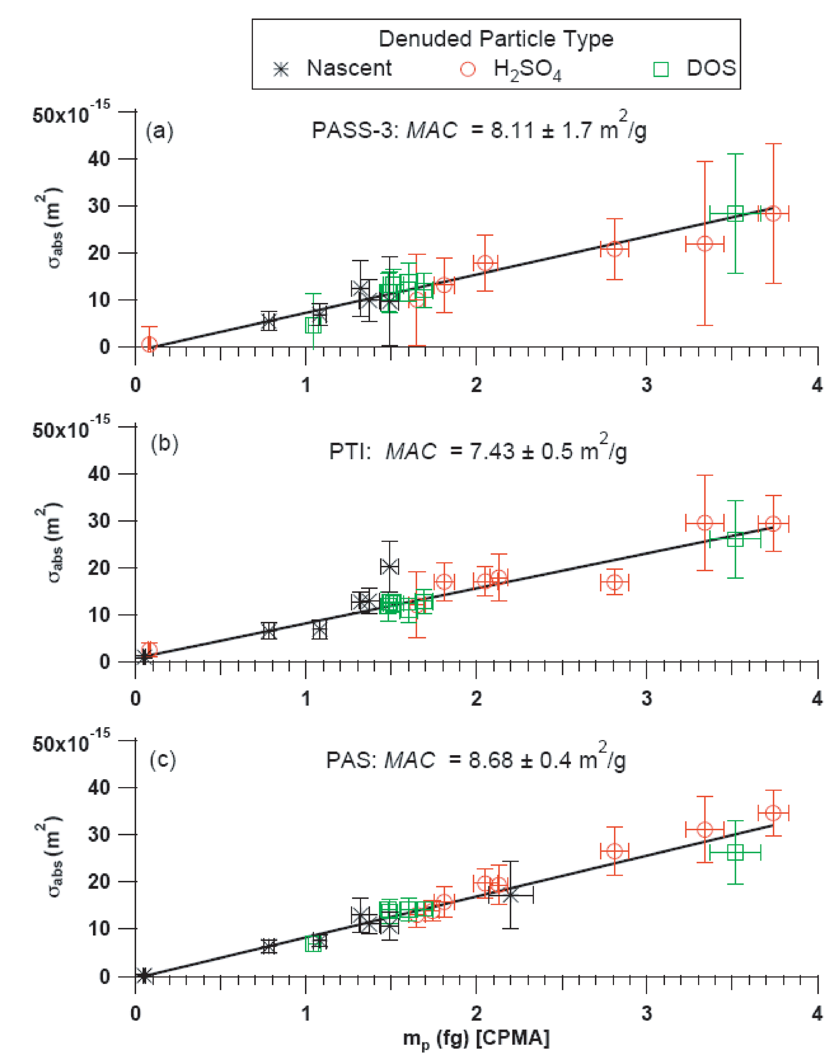
- Aquadag
- Oxidized Flame Soot (LBNL)
- Fullerene Soot
- Regal Black
- Activated Charcoal
- Carbon Nanopowder
- Glassy Carbon Spheres

Acknowledgements

This research was supported by the Department of Energy (DOE) Office of Science (BER), Atmospheric Science Program grants No. DE-FG02-05ER63995, DE-FG02-05ER84268, DOE DE-FG02-05ER63982, DOE SBIR grant No. DE-FG02-07ER84890 and the National Science Foundation (NSF) Atmospheric Chemistry Program grant No. ATM-0525355.

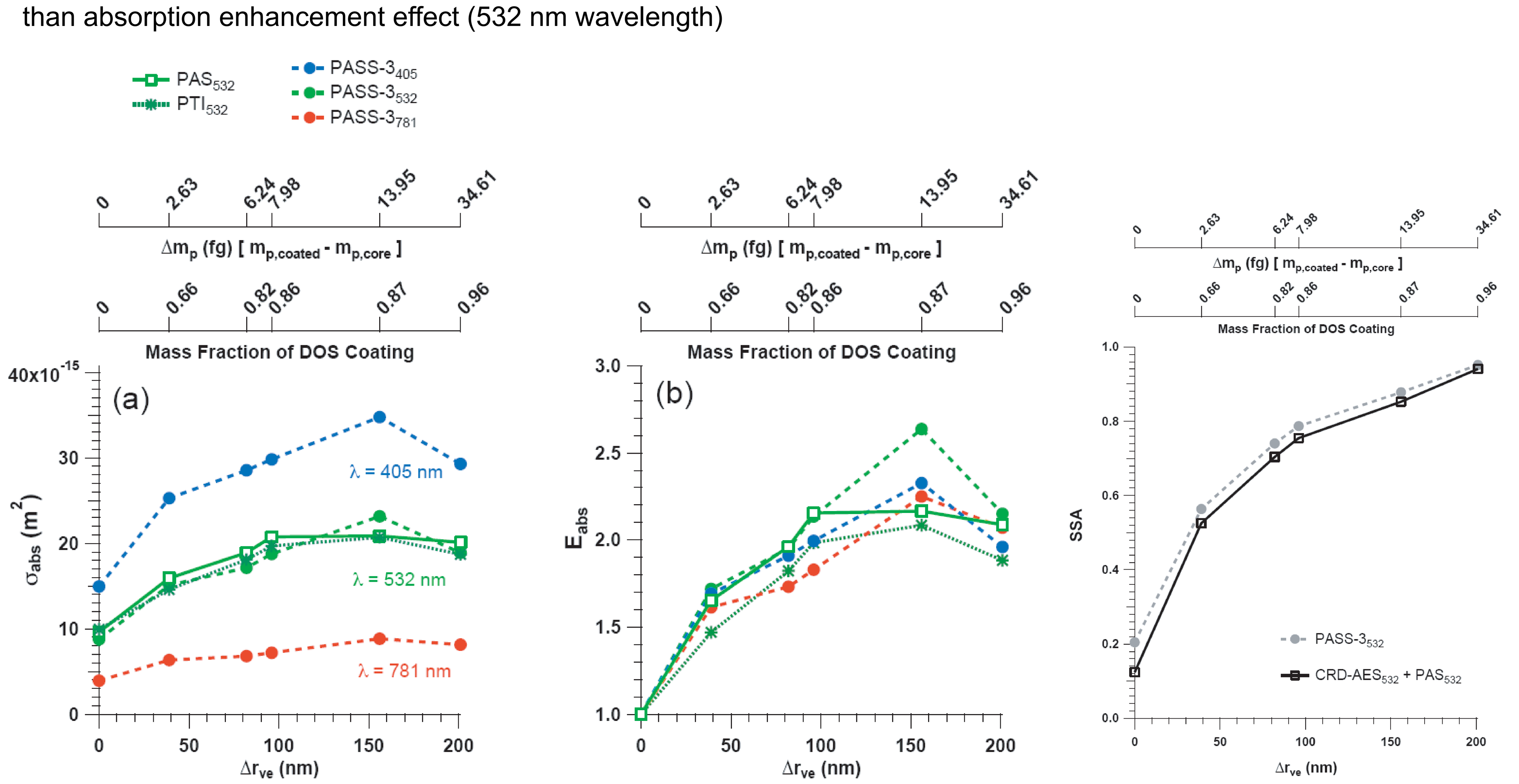
BC Mass Specific Absorption

- Denuded and coated/denuded pre-mixed flame BC particles exhibit a consistent mass-specific absorption coefficient of 8.1±1.8 m^2/g at a wavelength of 532 nm
- Three different absorption instruments participated (2 photoacoustic and 1 photo-thermal interferometer)



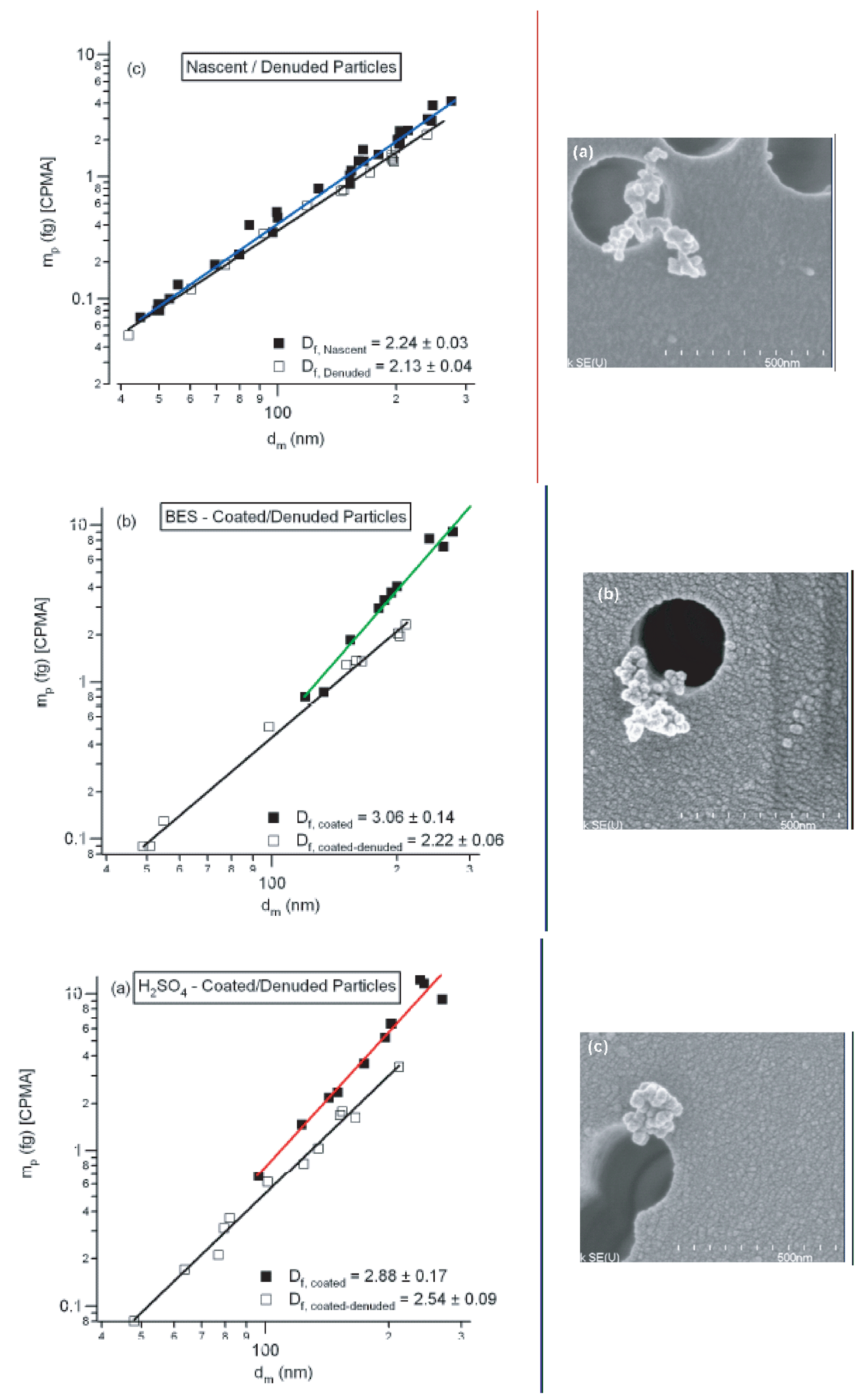
Absorption Enhancement due to Nonabsorbing Coatings

- Coating BC particles with nonabsorbing liquid organic or inorganic material increases the mass specific absorption coefficient
- Absorption enhancement increases with coating thickness to an apparent asymptote or maximum around 2X the denuded absorption
- Absorption enhancements are similar across a wavelength range from 405-781 nm
- Single scatter albedo measurements indicate that scattering due to size increase for coated BC particles (100-200 nm) increases faster than absorption enhancement effect (532 nm wavelength)



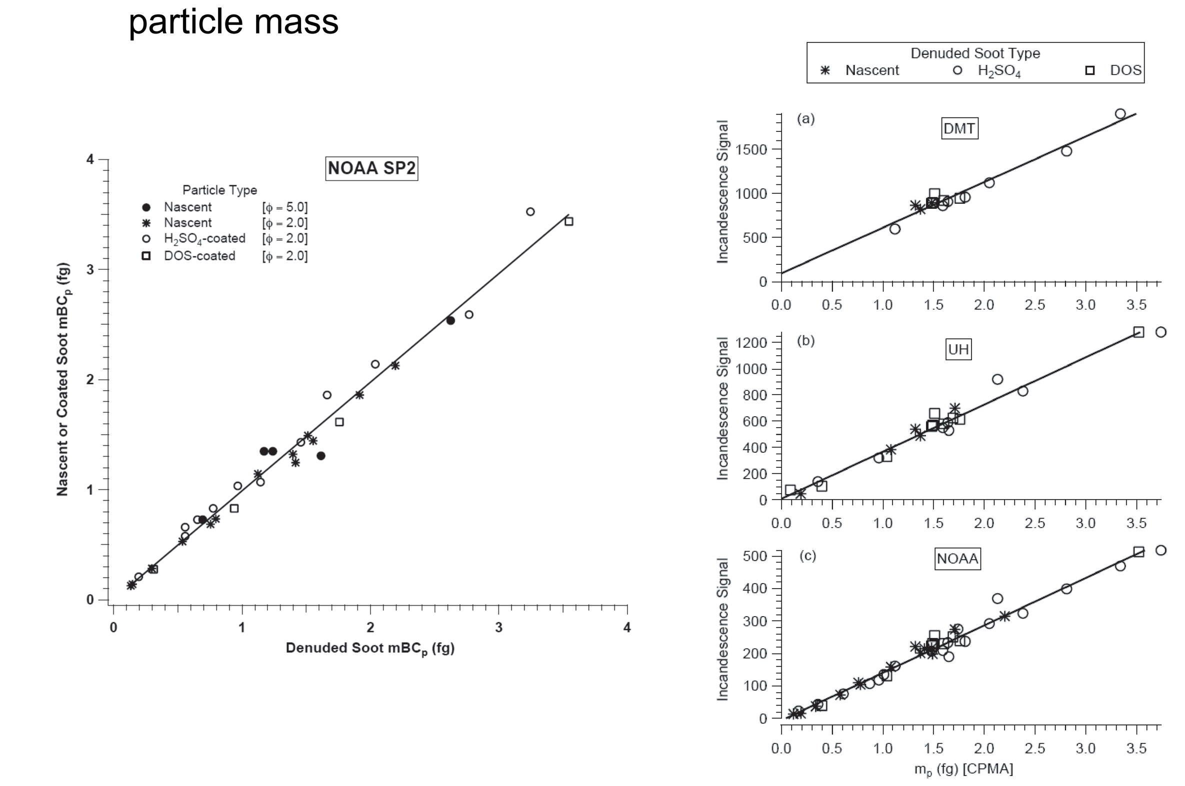
Morphological Changes due to Coating/Denuding

- Liquid organic (bis-ethylhexyl sebecate) and inorganic (sulfuric acid) coatings on BC particles reduce asphericity
- Sulfuric acid coating/denuding (especially with RH) collapse BC morphology
- BES coating/denuding does not dramatically effect BC morphology



Incandescence Signal of BC Particles with Coatings

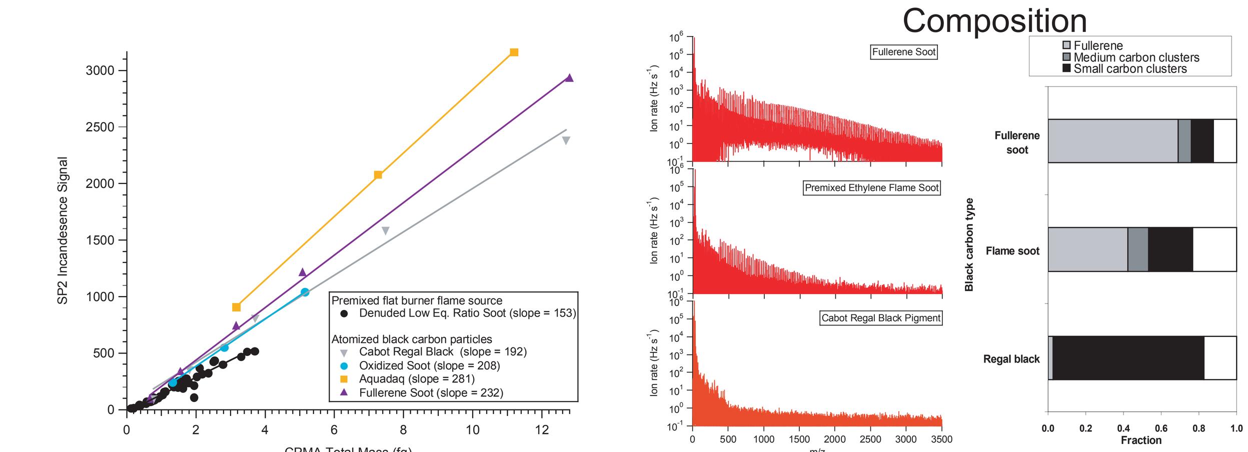
- SP2 incandescence signals are proportional to BC particle mass and insensitive to morphology and coatings
- Three different SP2 instruments participated and exhibited similar behavior, but with different sensitivities
- Tested the detection limits for SP2 instruments to measure BC particle mass



BC Chemical Composition Issues

- BC particles passing through a 1064 nm laser beam heat (up to their boiling point), incandesce, and vaporize carbon clusters
- SP2 Incandescence signal intensities vary as a function of BC particle type
- Vaporized carbon clusters distributions as measured by a SP-AMS vary as a function of BC particle type
- Complicates instrument calibrations and interpretations of ambient measurements

'Black Carbon' Chemical Composition



Summary and Outlook

- Mass-specific absorption coefficients (532 nm) for pre-mixed ethylene flame BC particles measured to be 8.1±1.8 m^2/g with three different absorption instruments
- Absorption enhancements observed for nonabsorbing organic and inorganic coatings on flame generated BC particles
- Flame generated BC particle morphology collapse during coat/denude with sulfuric acid (especially with RH), but little effect during coat/denude with BES
- SP2 (three different instruments) incandescence signals proportional to flame generated BC particle mass and independent of morphology or coating
- More controlled laboratory work is necessary to investigate different BC particle types and their influence on our capability to accurately measure the physical, optical, and chemical properties of ambient BC particles