Probing Black Carbon Mixing State with SP2

A. J. Sedlacek[1] and R. Subramanian[2]

[1]BNL; [2]DMT

ASR Atmospheric System Research

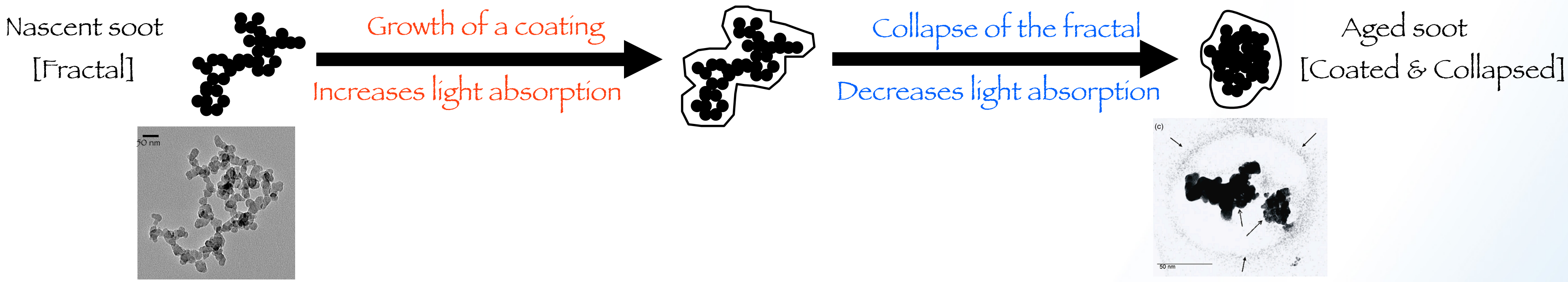
BROOKHAVEN NATIONAL LABORATORY

a passion for discovery

Office of Science U.S. DEPARTMENT OF ENERGY

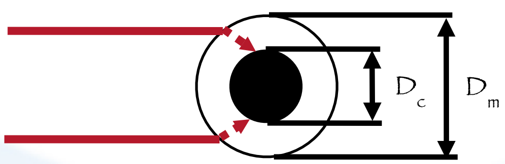
Black Carbon Mixing State

Particle mixing state greatly influences the radiative forcing contribution by BC



- Encapsulation can lead to significant increase in positive forcing
- Uncertainty regarding the contribution of soot to aerosol radiative forcing is unacceptably large

Encapsulation Effects



$$\text{Absorption Amplification} = \frac{\sigma_{abs}^{encapsulated}}{\sigma_{abs}^{uncoated}}$$

Black Carbon Mixing State

Being able to probe the BC mixing state will provide useful data towards:

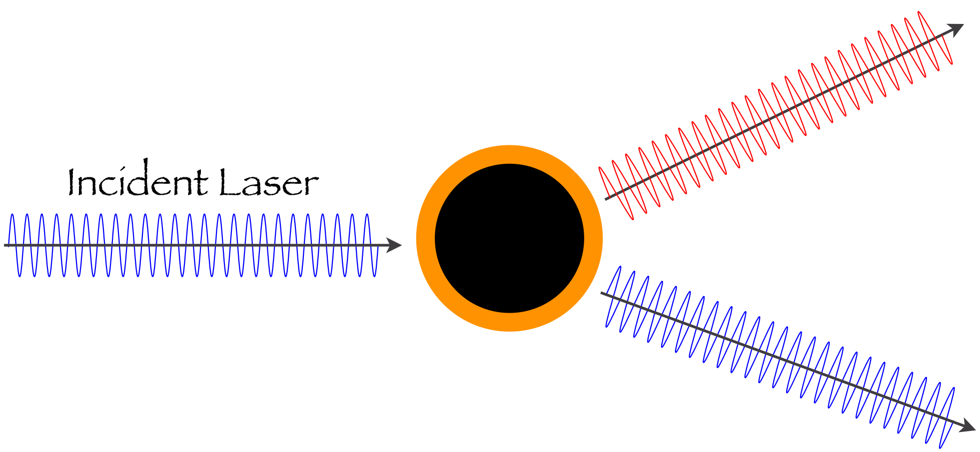
- Providing context for aerosol light absorption measurements (e.g., coating vs BrC)
- Providing particle-resolved data on BC mixing state evolution for comparison with models
- Probing the influence of mixing state (aging) on microphysical properties (e.g., CCN activity)

The single particle soot photometer (SP2) can probe the BC mixing state

Probing BC Mixing State

Schwartz et al., 2006; Moteki & Kondo, 2007, Subramanian et al., 2010

- Particle-by-particle instrument (number conc; mass conc; $dN/dNlogD_p$; $dM/dlogD_p$)
- High specificity towards 'refractory' black carbon (rBC)



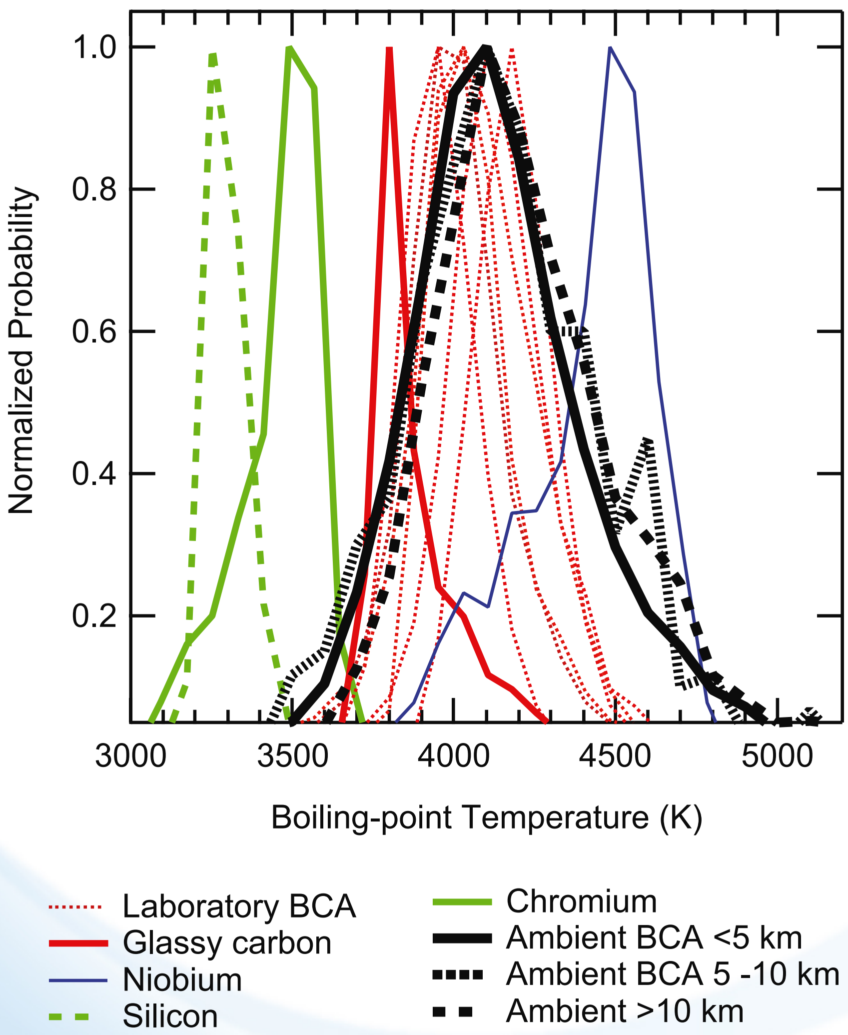
Incandescence
(BC diameter: ~60 — 600 nm)

Scattering
(particle diameter: ~175 nm - 325 nm)

Mixing state probe:

- Examine temporal profiles of the scattering and incandescence signals
- Probe coating thickness: optical (scattering) and BC mass equivalent (incandescence) diameters

Physics behind the SP2: Incandescence Temperatures



Incandescence unique to material

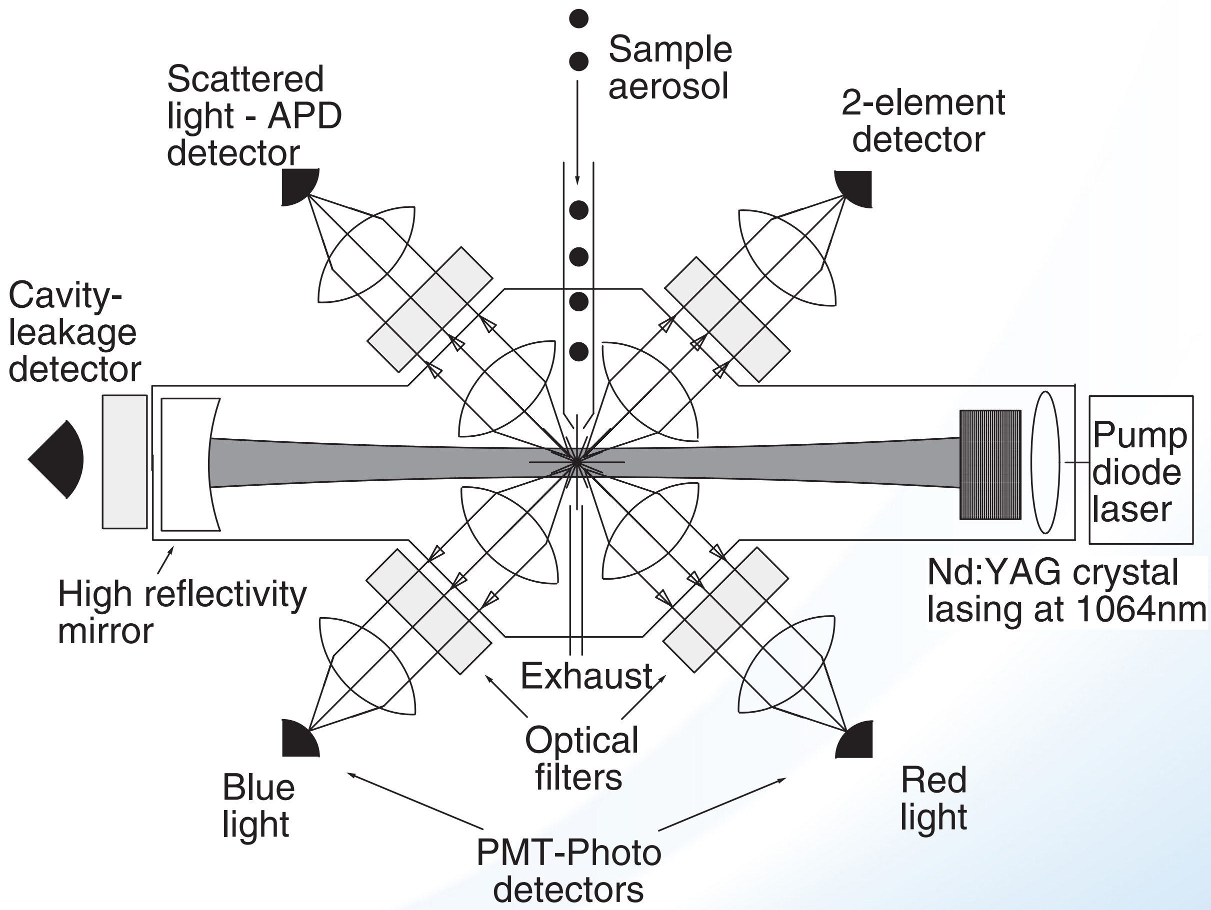
Intensity of signal ∝ amount of material

Careful choice of 'narrowband' and 'broadband' filter provides high selectivity towards BC

Schwarz et al., 2006

Single Particle Soot Photometer: The Instrument

- Particle-by-particle instrument

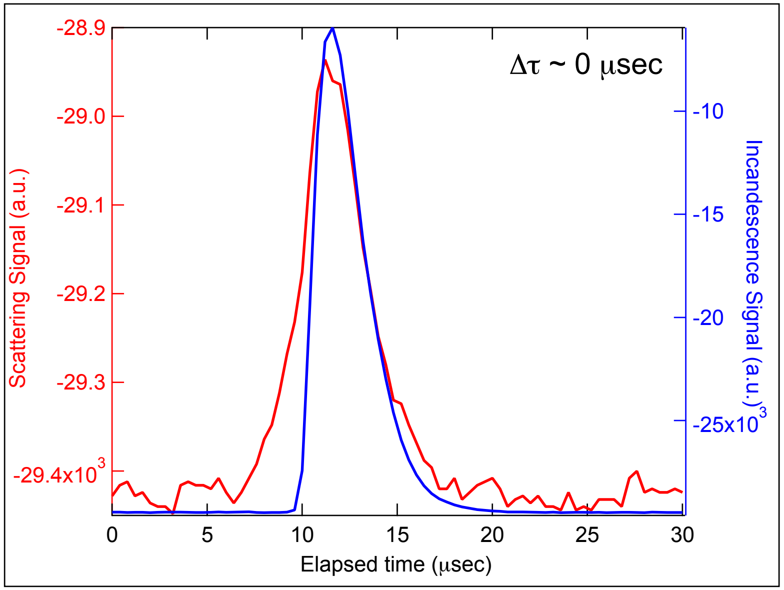


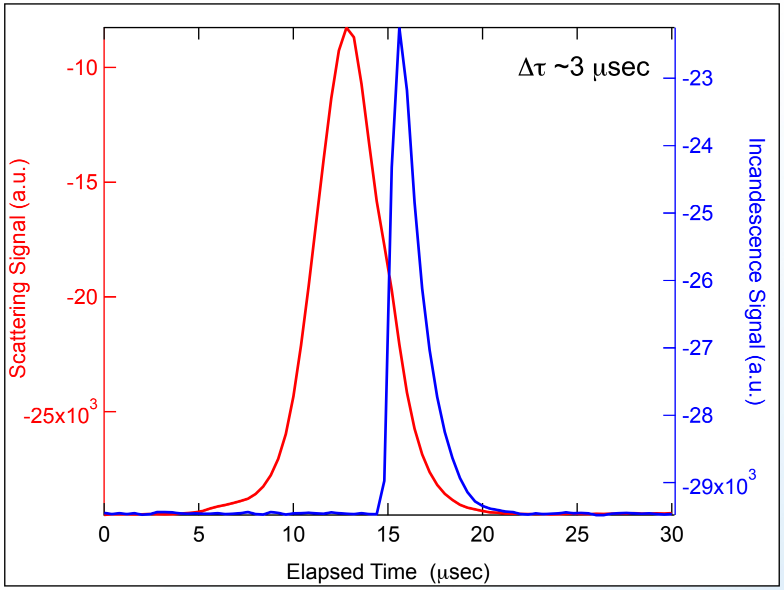
Schematic Schwarz et al., 2010

Black Carbon Mixing State: Lagtime

Moteki and Kondo, 2007; Subramanian et al., 2010

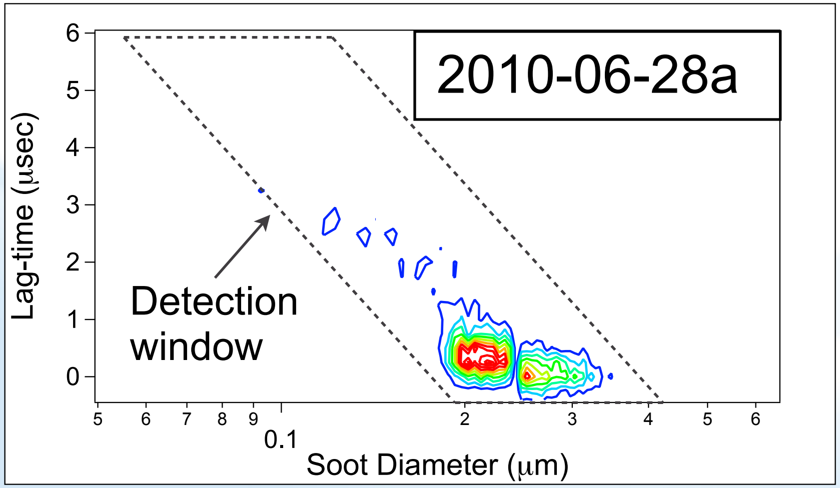
$\Delta\tau = \tau_{incandescence} - \tau_{scattering}$ = time to 'boil off' coating

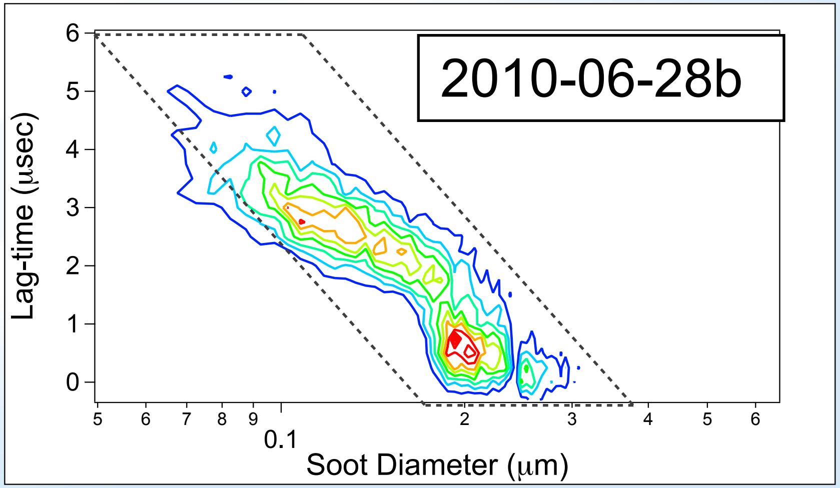




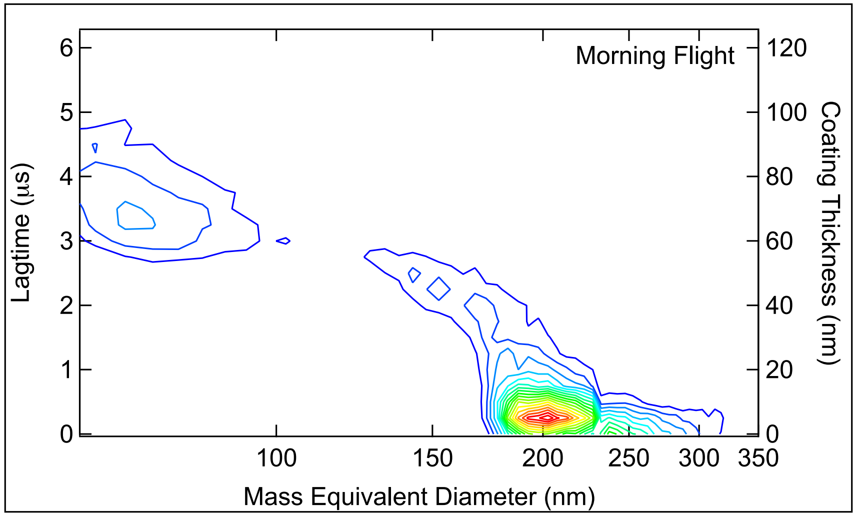
Thin coating ≡ $\Delta\tau \approx 0$

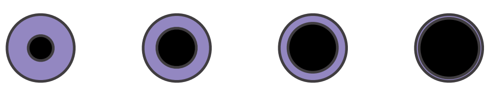
Thick coating ≡ $\Delta\tau \neq 0$

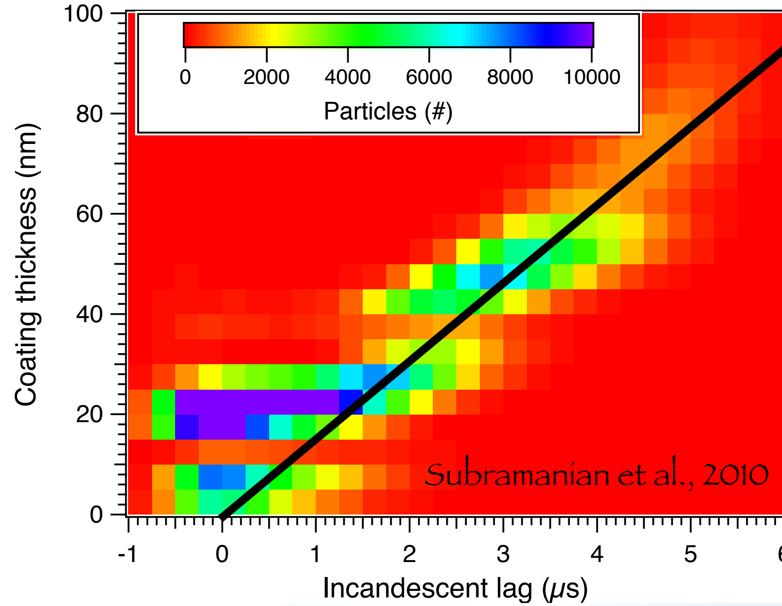


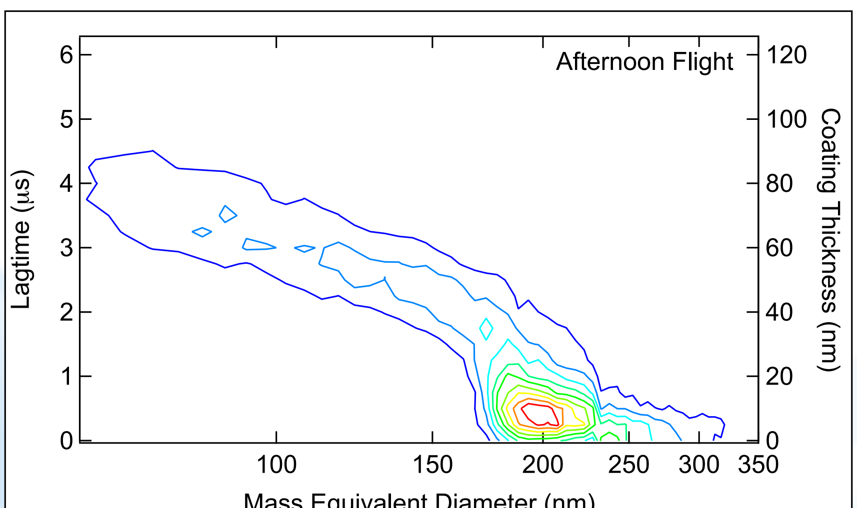


June 28, 2010 -- Evolution of rBC mixing-state





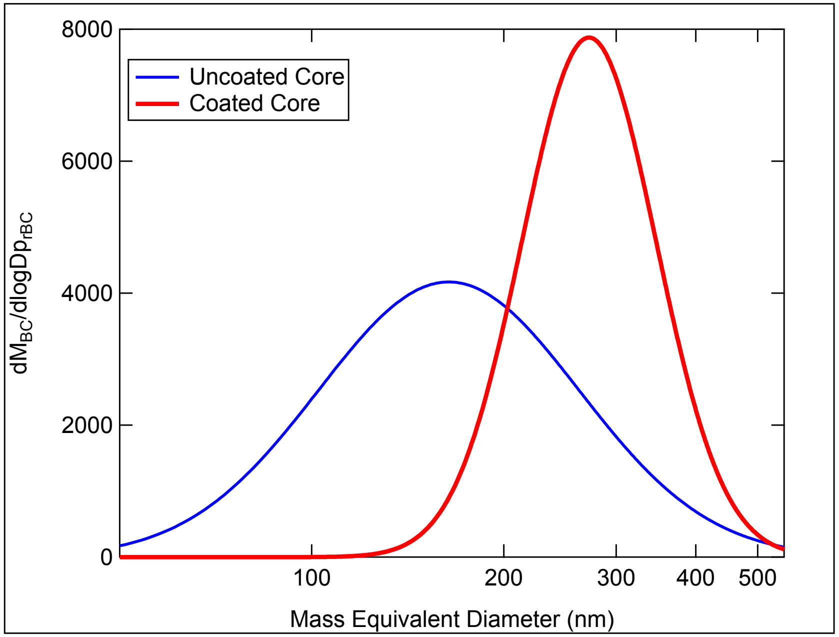




- Bimodal distribution in the morning (aged and nascent soot)
- Loss of small diameter, thickly coated BC
- Increase in larger diameter, thickly coated BC
- Appearance of 120 nm diameter BC with coating thickness ~55 nm in PM

Reconstructed $dM_{coated\text{-}core}/dlogD_p$

Apply simplified condensation model to uncoated BC distribution to reconstruct coated-core distribution



Key measurement (AM → PM):

120 nm BC core grow to D_p = 230 nm

Condensation Model:

- Condensable supplied until 120 nm core is coated to 230 nm
- Fuchs (Knudsen) correction

Int'l mixing state of BC:

$M_{coating}/M_{rBC}$ ~3

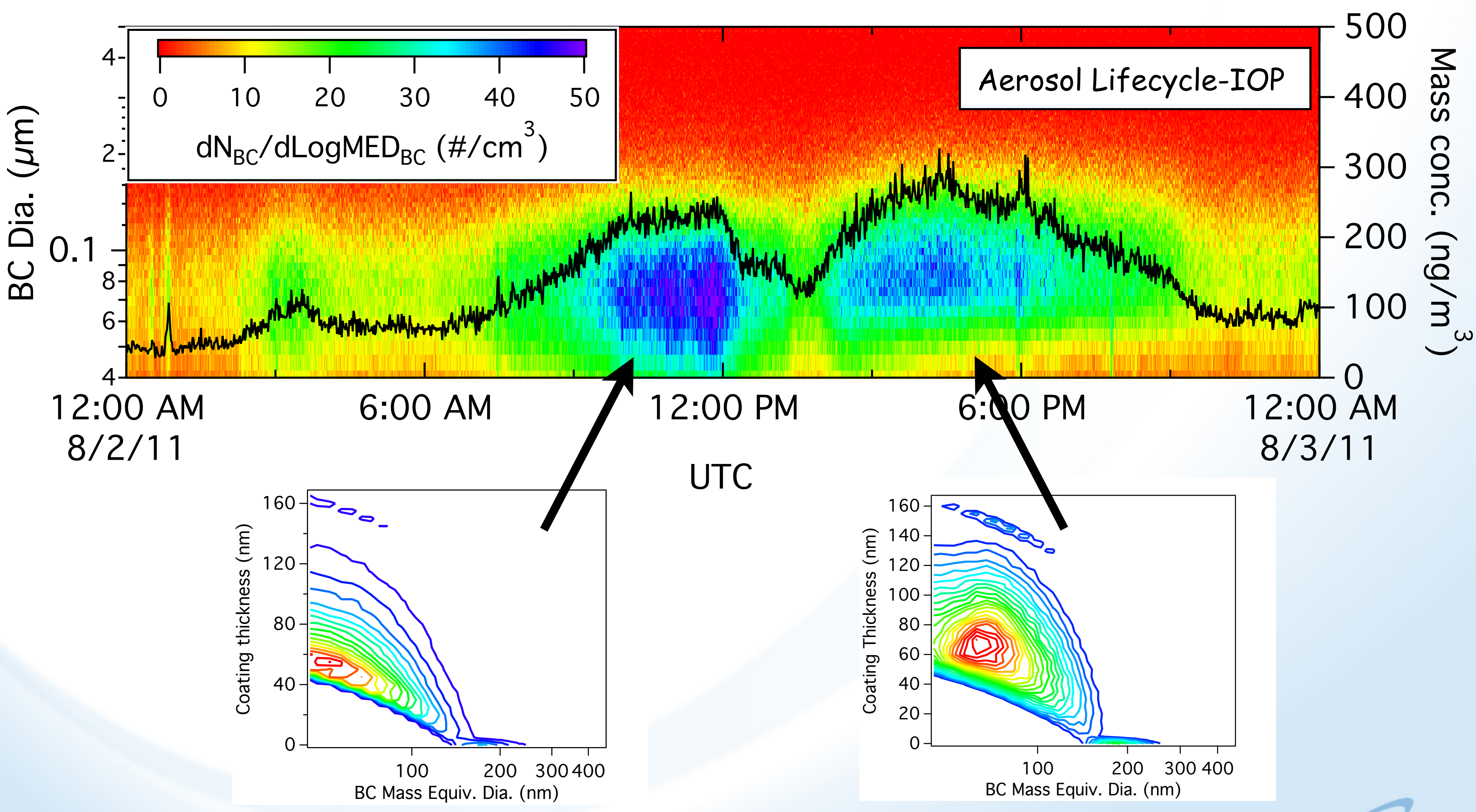
Reconstructed distribution enables estimate of coating mass & rBC mass

- ρ_{org} = 1.4 g/cc (organic; AMS)
- ρ_{rBC} =1.8 g/cc (BC)

BROOKHAVEN NATIONAL LABORATORY

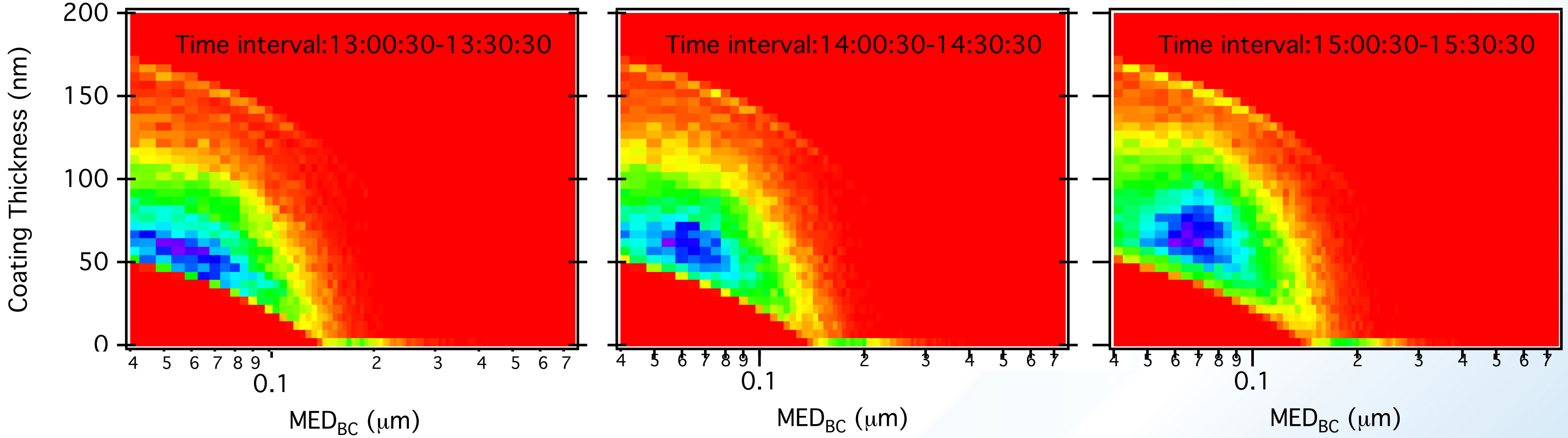
Measuring Growth of Coating

Optical diameter - mass equivalent diameter = coating thickness

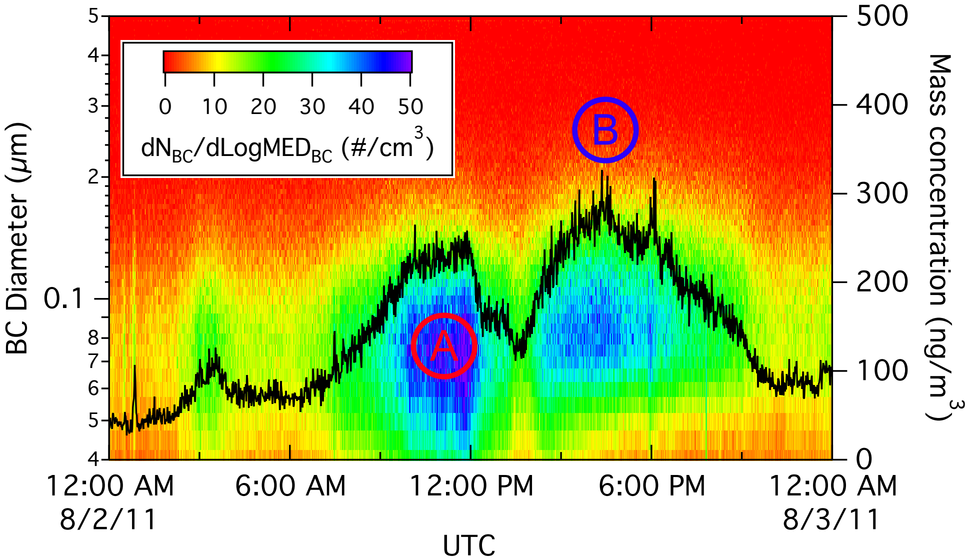


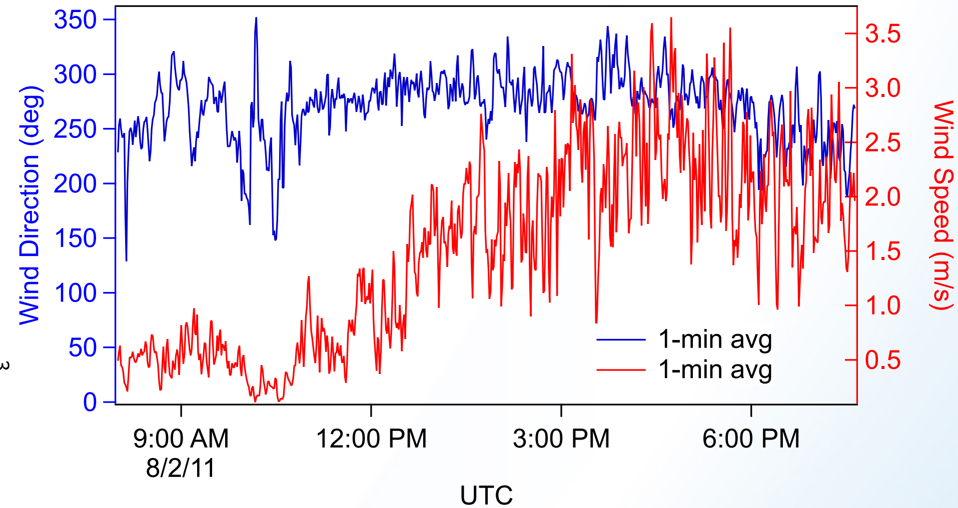
BC Coating Time Series

Aerosol Lifecycle-IOP: August 2, 2011



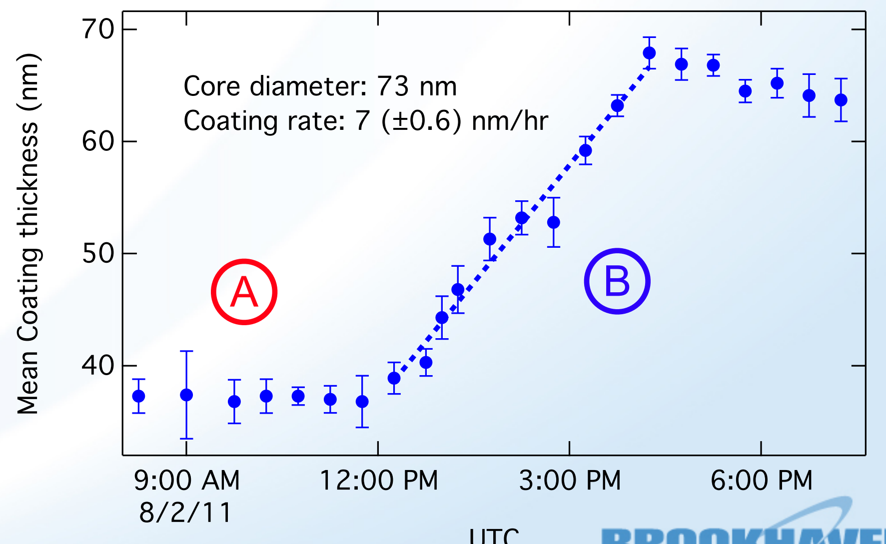
Evolution of Black Carbon Mixing State





In contrast to morning plume, coating thickness for the afternoon plume is observed to increase.

- Local growth or advection?



BROOKHAVEN NATIONAL LABORATORY

Time-Dependent Scattering Cross-Sections

Tactful assumption: the amplitude of the scattering signal reflected the nascent 'diameter' of the particle.

However, as a BC particle travels through the laser beam, light absorption results in particle heating, which, in turn, causes loss of coating material

Issue: How can we get a better estimate the nascent particle diameter?

Two approaches:

- Leading Edge Optimization: LEO (Gao et al., 2007)
- Normalized Derivative Method: NDM (Moteki & Kondo, 2008)

Basis for both Approaches

- For particles < 1 μm, the scattering cross-section (for a given RI) is single-valued
- The scattering signal will reflect the Gaussian lineshape of the SP2 laser

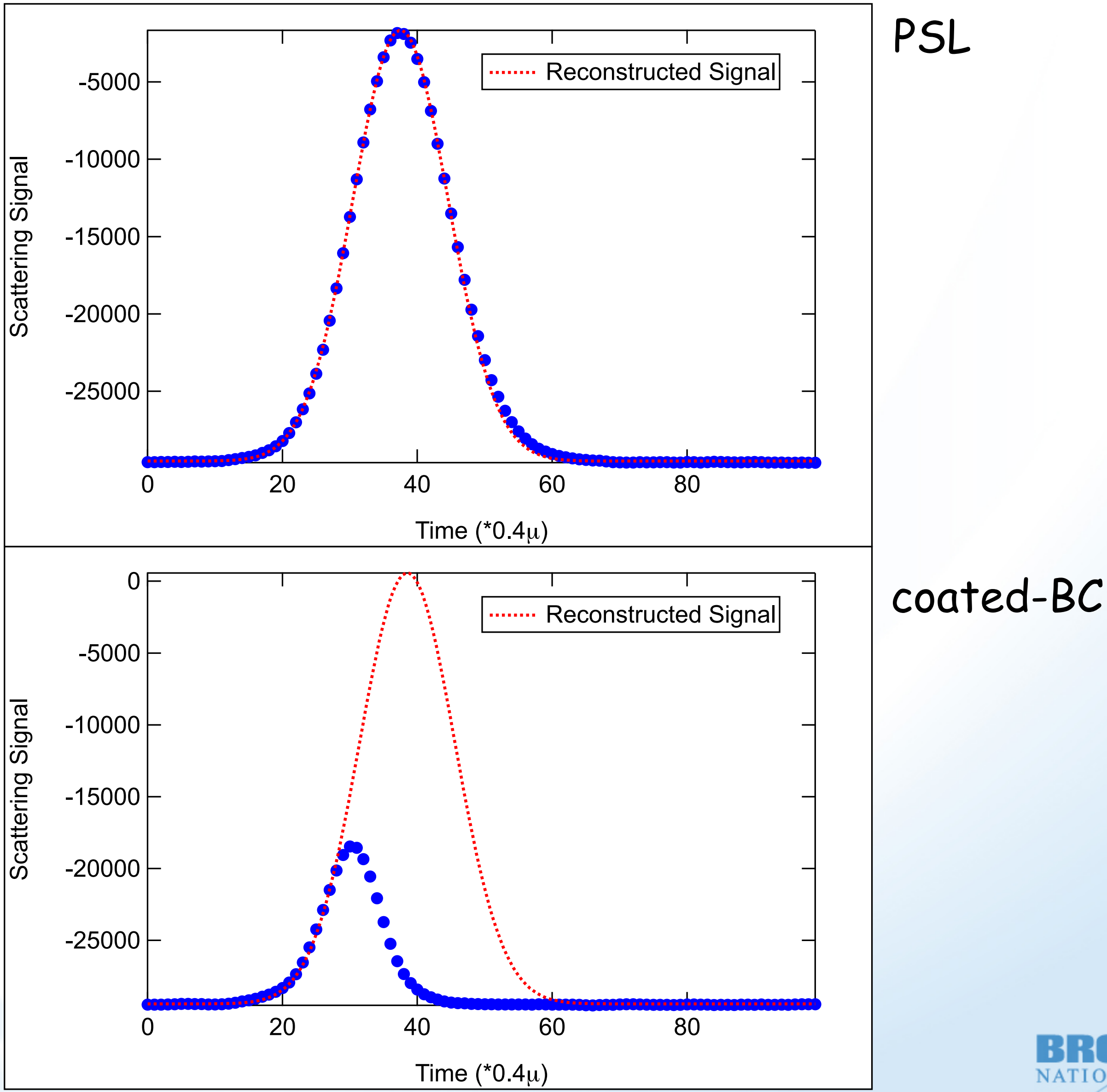
$$\beta + Amp \times e^{((t-\tau)^2/\sigma^2)}$$

- Gaussian amplitude can be related to a scattering x-section

Central proposition: with β , τ , and σ known, fit 'leading edge" of scattering signal to determine Gaussian amplitude.

Leading Edge Optimization (LEO)

Examples

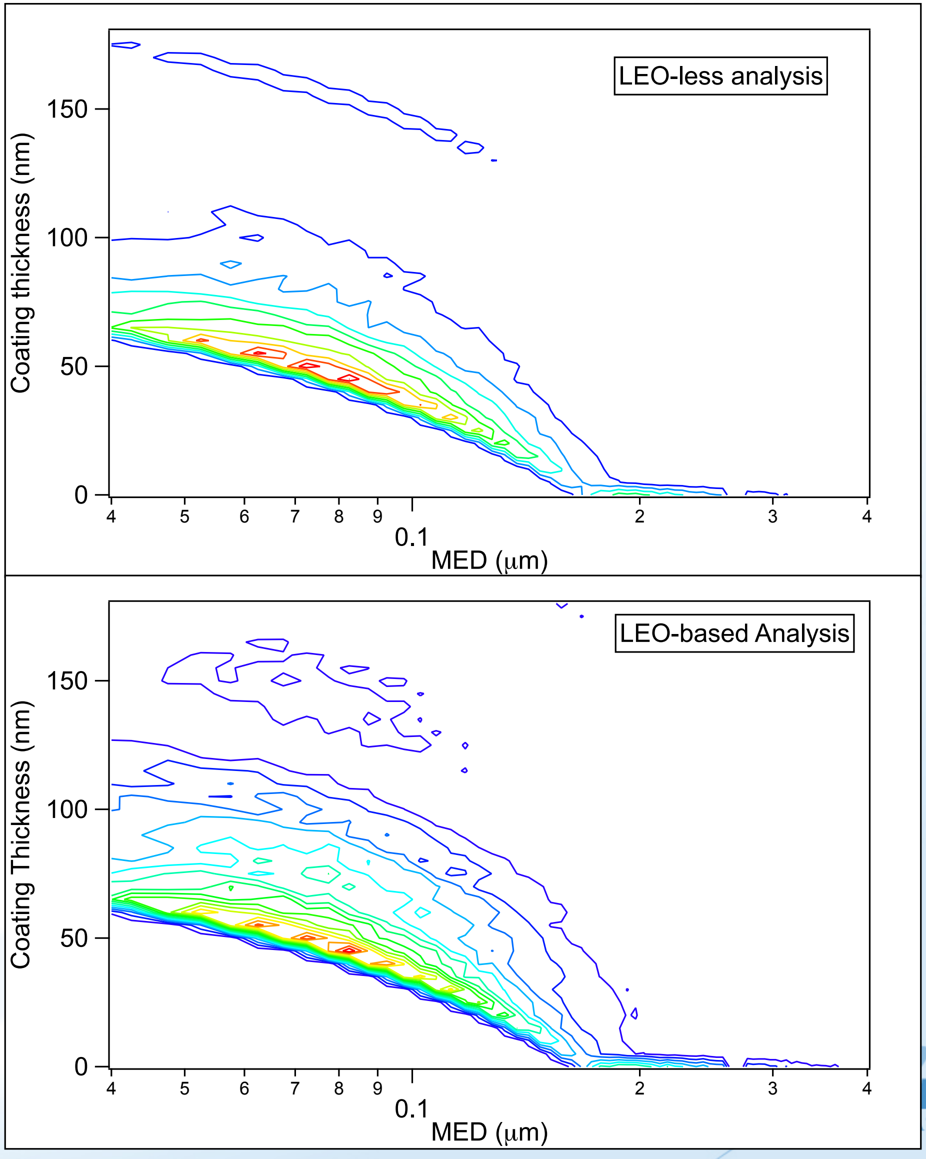


Leading Edge Optimization (LEO)

Storm Peak Lab SP2

2010-12-07

Optical detection window: 175 – 350 nm



LEO method detects the presence of smaller, more thickly coated particles that would otherwise be missed

Desert Research Institute

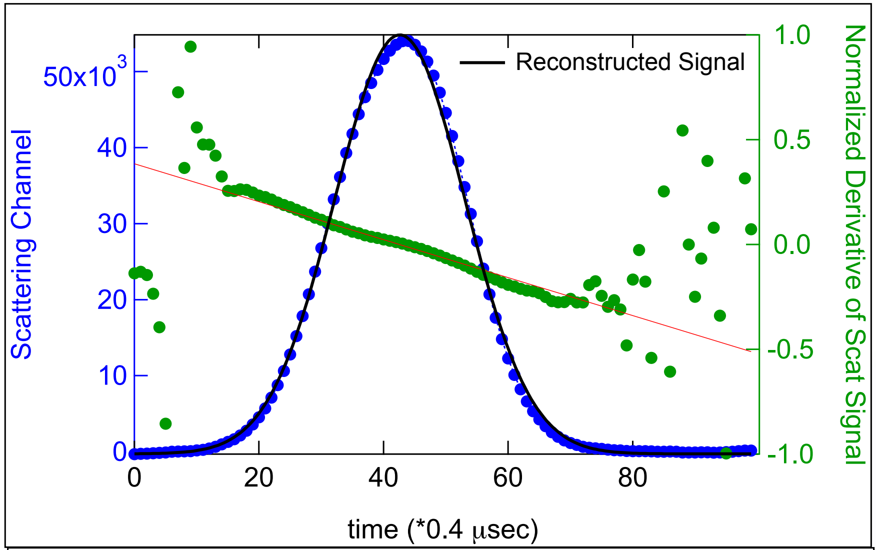
Brookhaven Science Associates

STORMVEx

Time-Dependent Scattering Cross-Sections

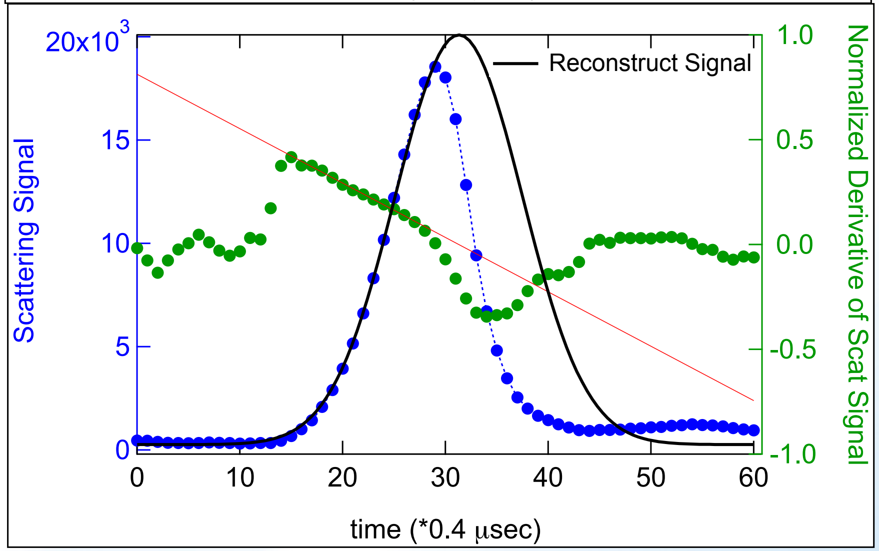
Examples

$$\frac{d\sigma_{scat}}{dt} = 0$$



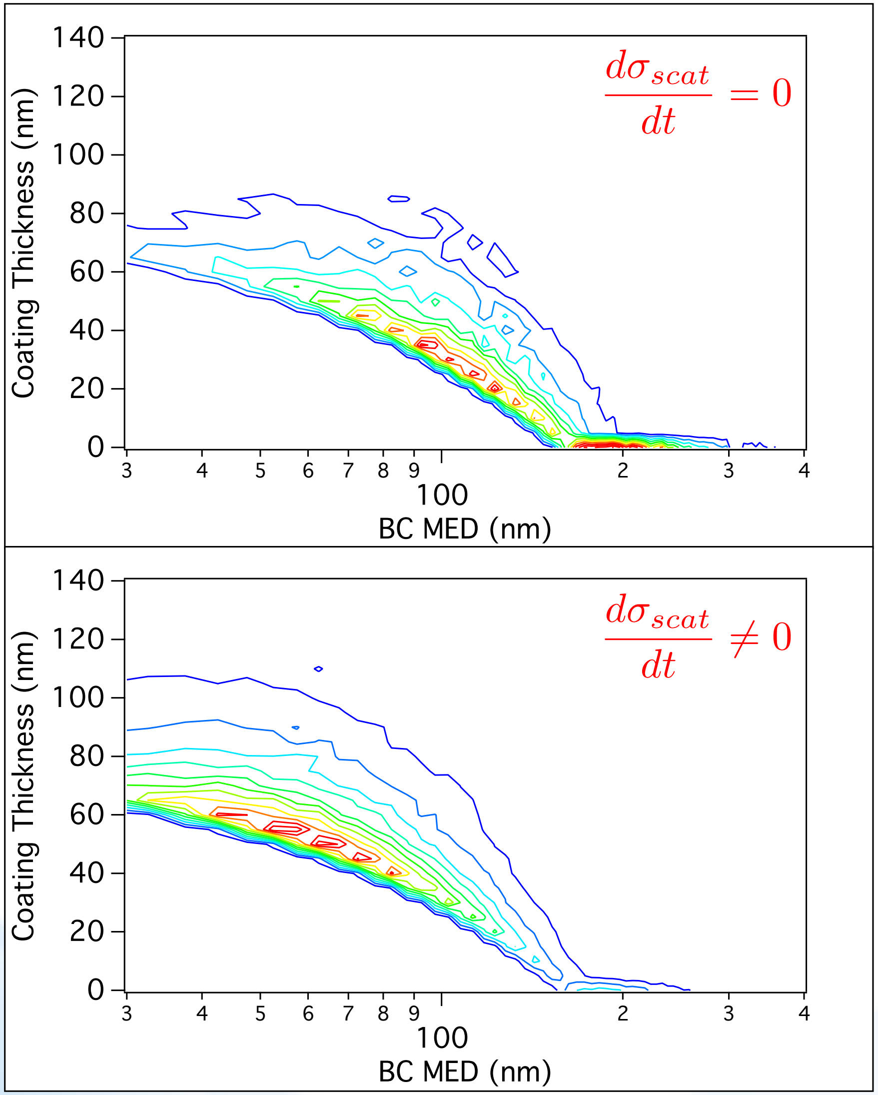
pure scatter

$$\frac{d\sigma_{scat}}{dt} \neq 0$$



coated BC

Normalized Derivative Method



Storm Peak Lab SP2

2011-01-29

Optical detection window: 175 – 350 nm

ND method detects the presence of smaller, more thickly coated particles that would otherwise be missed

Advantages/Disadvantages

Over the next few months a more quantitative assessment will be made. But even now, a comparison of the advantages/disadvantages is still instructive

Item of interest	LEO	NDM
CPU requirements, robustness of approach*	+	--
Optics (e.g., alignment)	-/+	+
Fixed σ	+ (-)	+
Science (loss of SD channel would enable incorporation of add'l scat detector OR the addition of a dust channel)	-	+

*to be evaluated more completely in coming months

Summary

SP2 provides useful information on BC mixing state & the evolution of that mixing state

- Simplified condensation model can be used to reconstruct coated-core distribution

  June 28, 2010 (CARES) estimate $M_{coating}/M_{rBC} \sim 3$

- Evolution of coating thickness

  August 2, 2011 (ALC-IOP) estimate coating growth rate: 7 nm/hr

- Work is currently underway to address time-dependent scattering cross-section