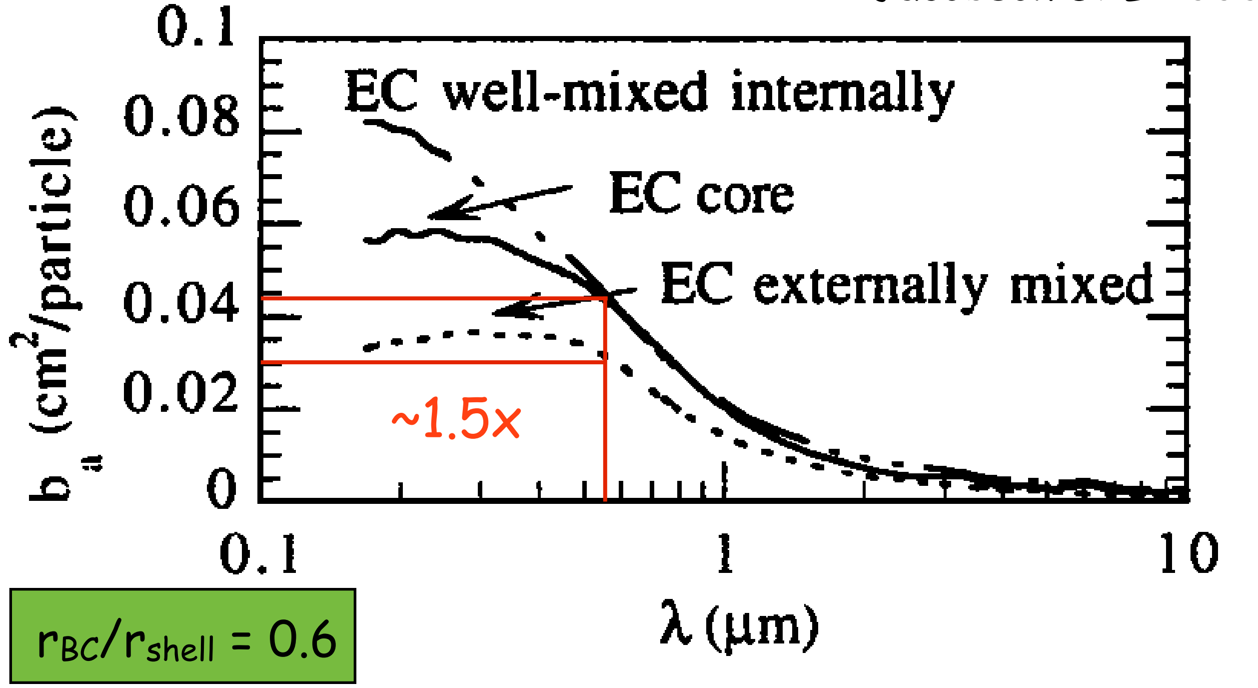
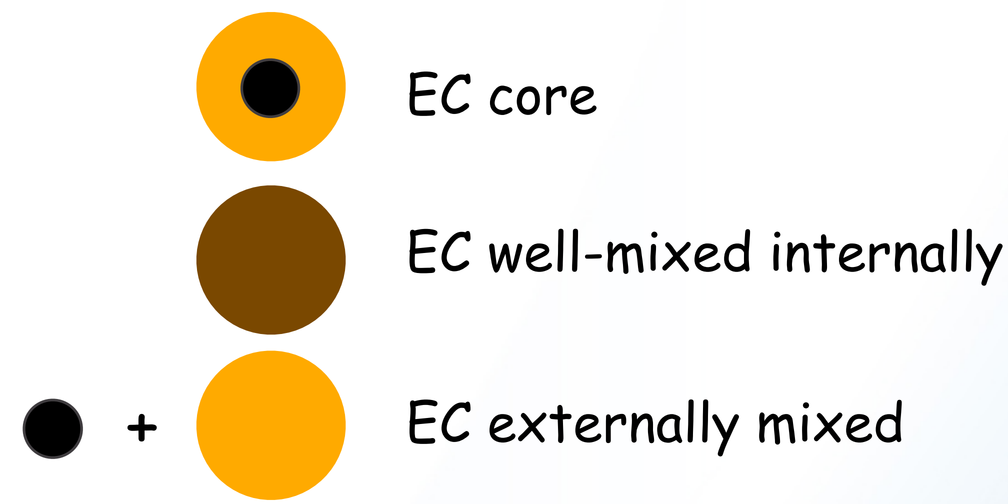
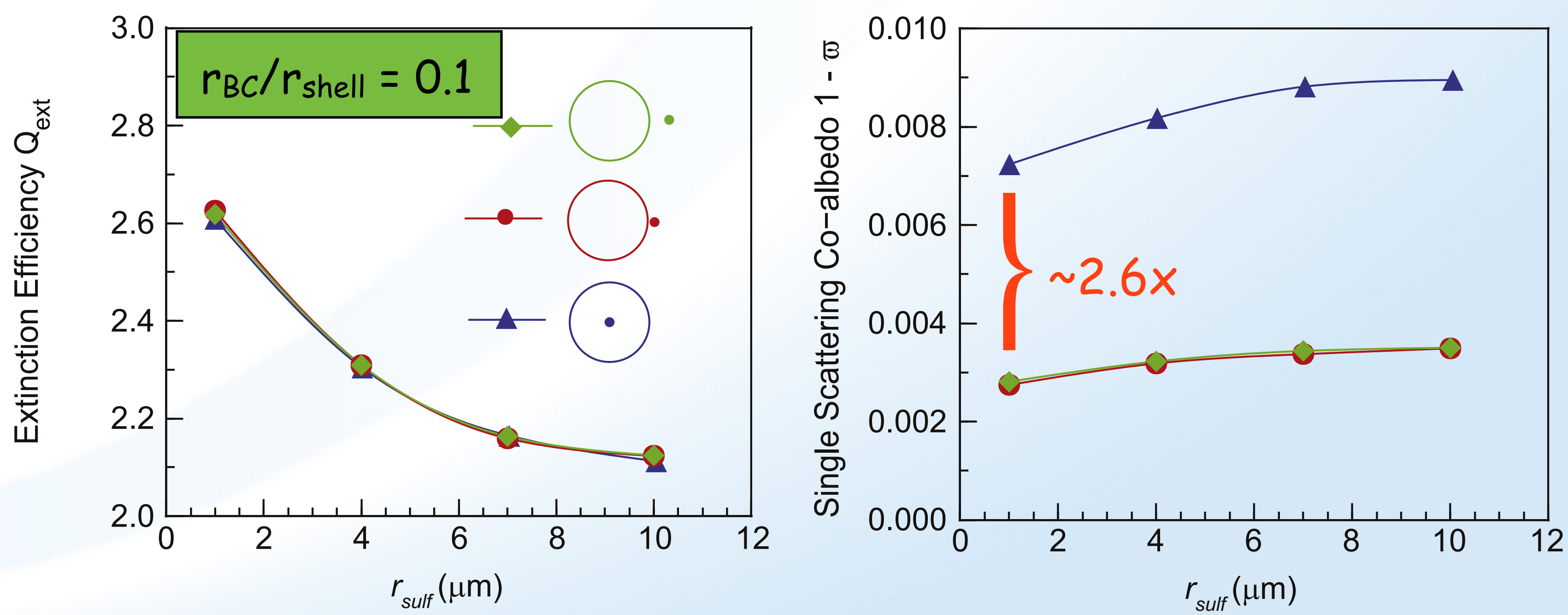
Influence of Black Carbon (BC) Mixing State on Light Absorption

Jacobson GRL 2000





Position of BC within particle predicted to influence light absorption

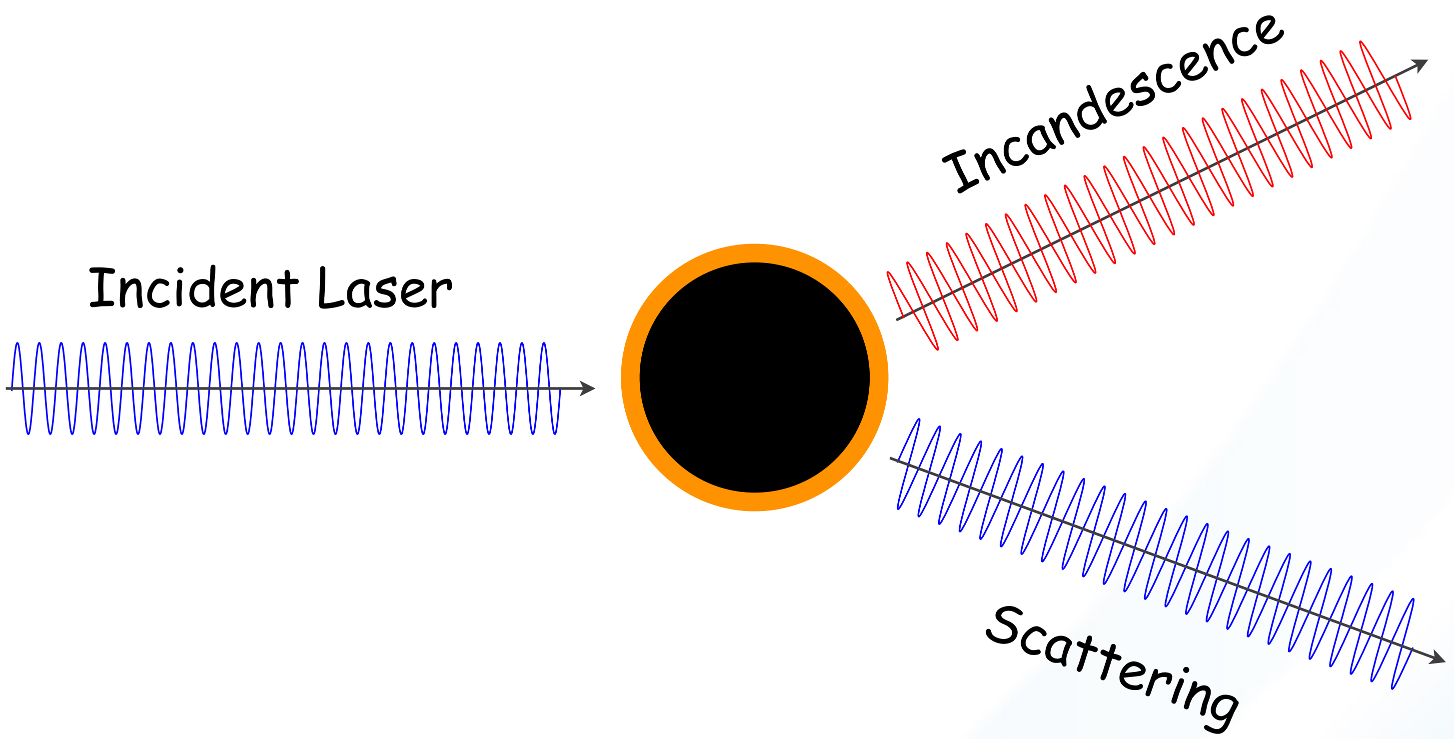


Liou et al., JQSRT 2011

Probing BC Particle Structure

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

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

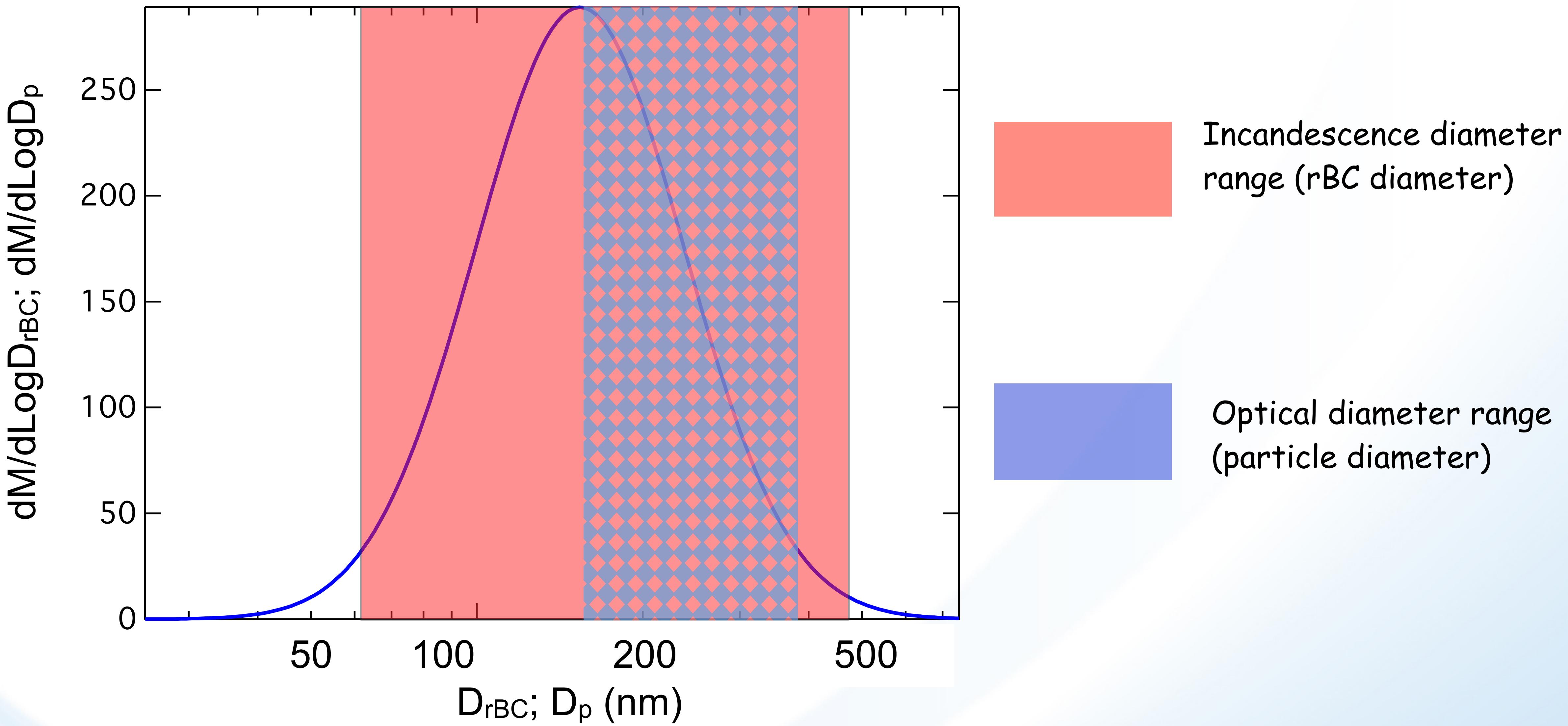


Probe BC structure:

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

Incandescence & Scattering Detection Ranges

Mixing state analysis requires data from both the incandescence and scattering channels

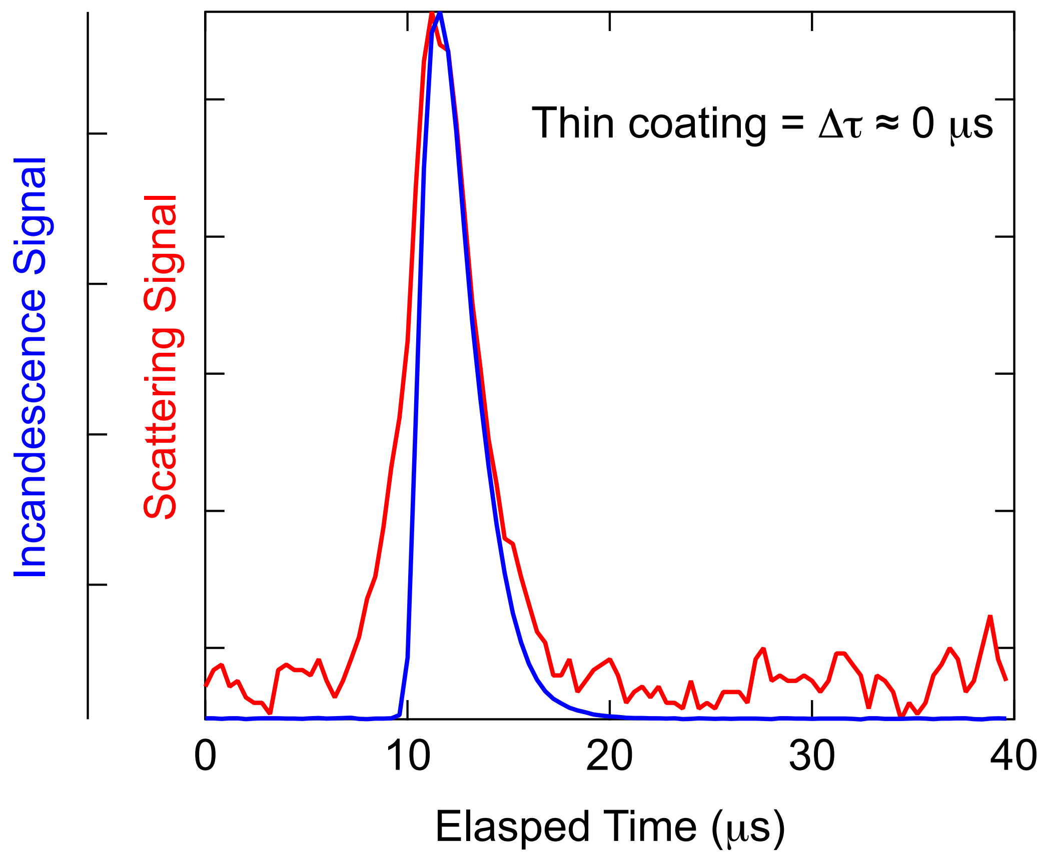


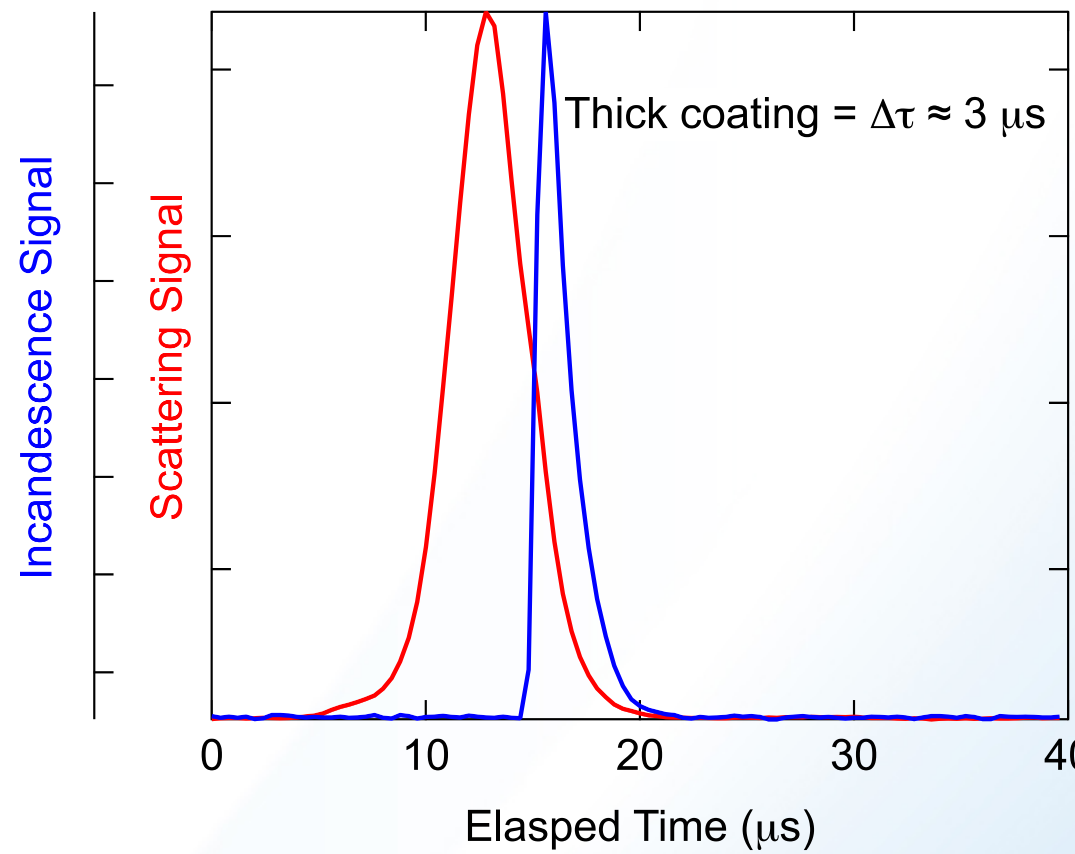
'Useful' diameter range for SP2-based BC mixing state analysis: ~160 - 375 nm

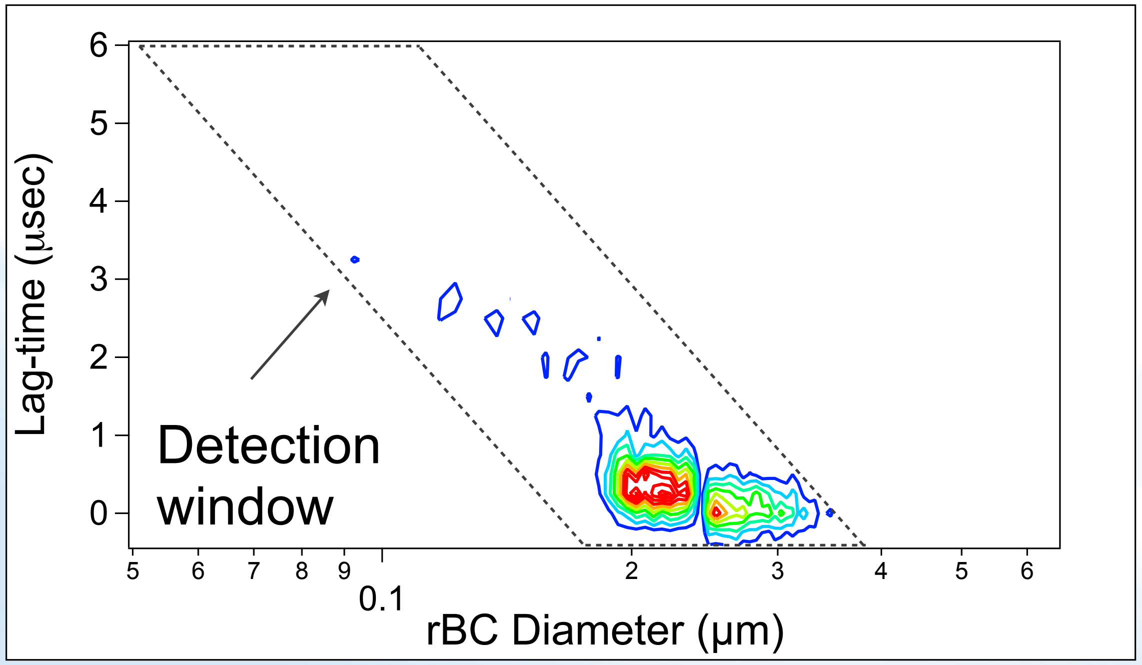
Proxy for Coating thickness: Incandescence Lagtime

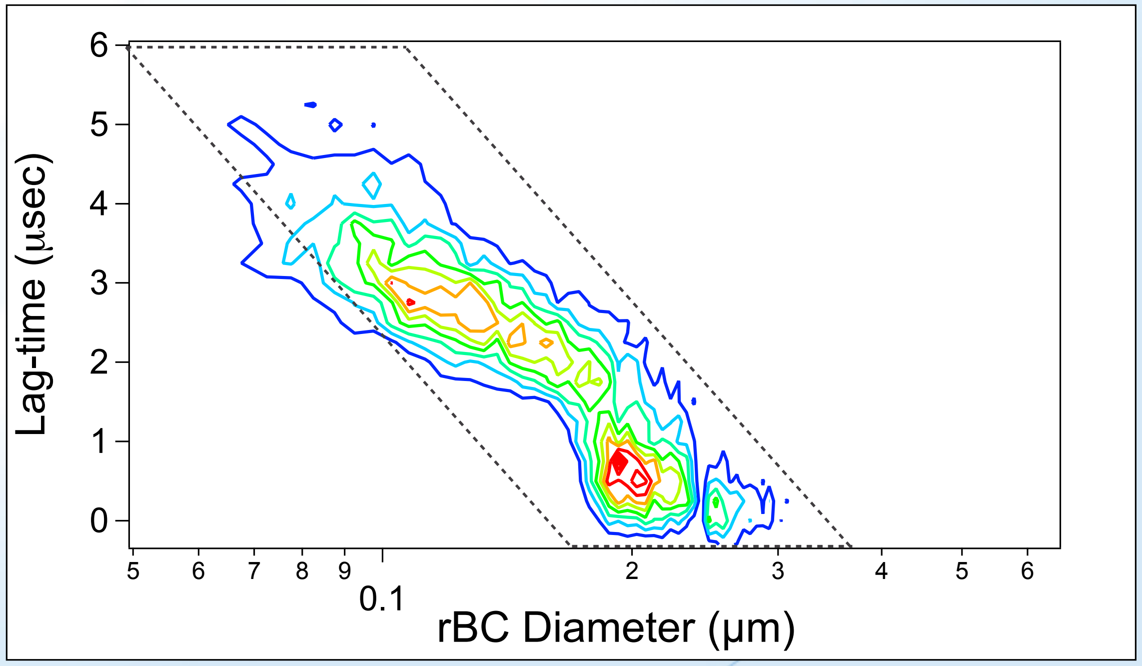
Moteki and Kondo, 2007; Subramanian et al., 2010

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



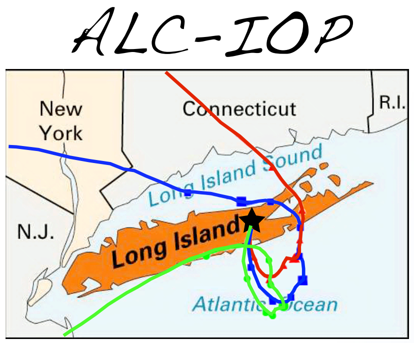




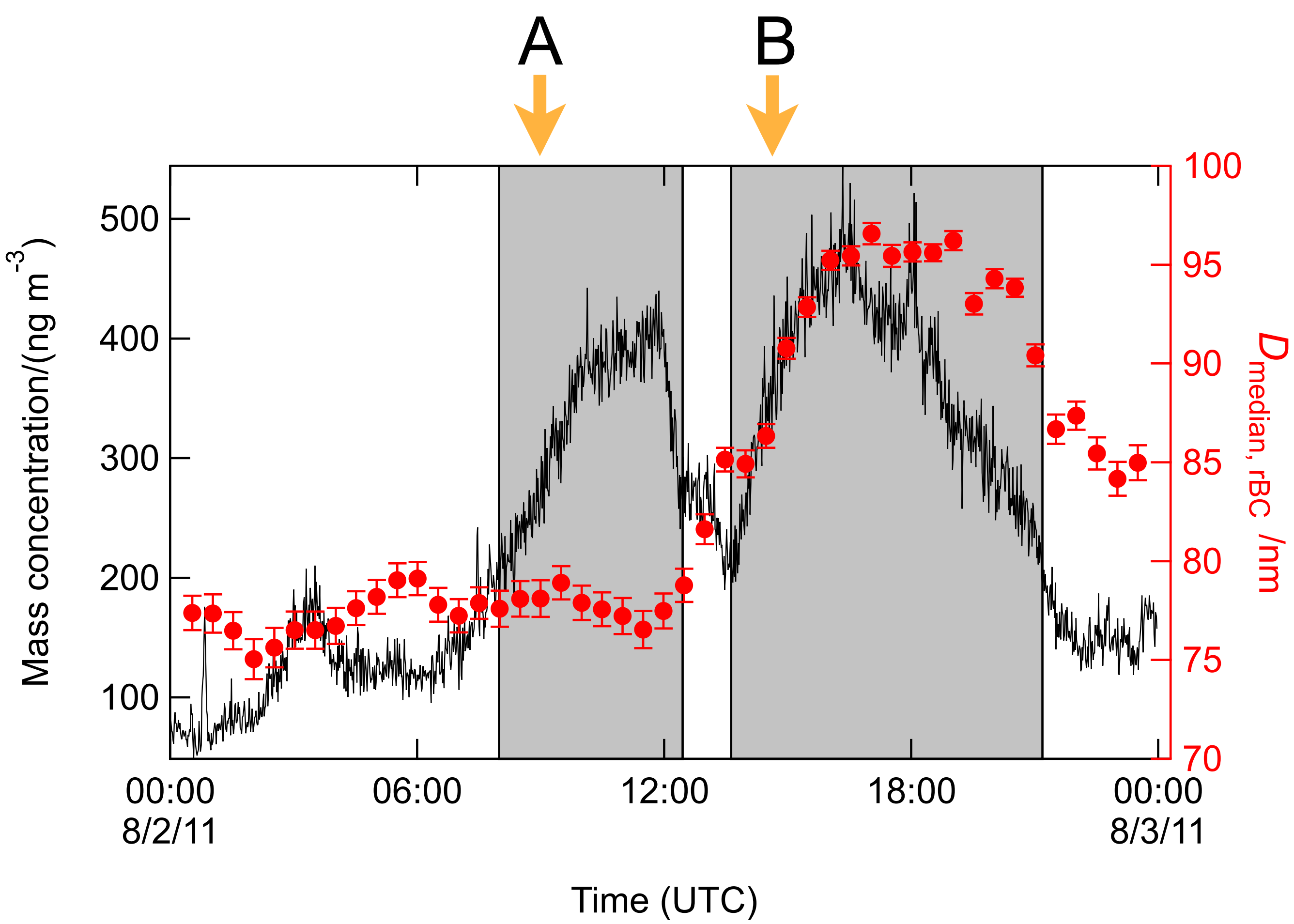


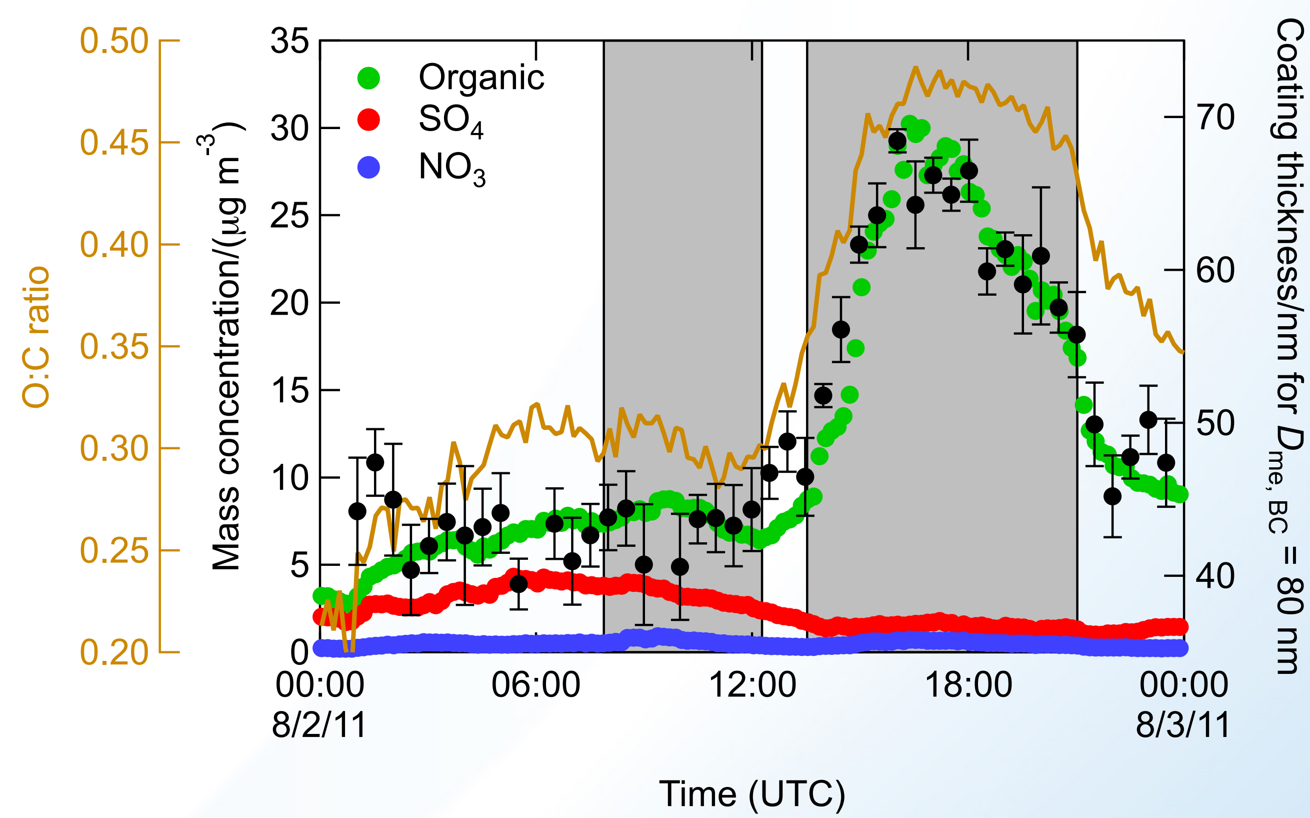
Change in BC Mixing State

Sedlacek et al., (GRL, 2012)



Aerosol Lifecycle field campaign held at BNL: August 2, 2012



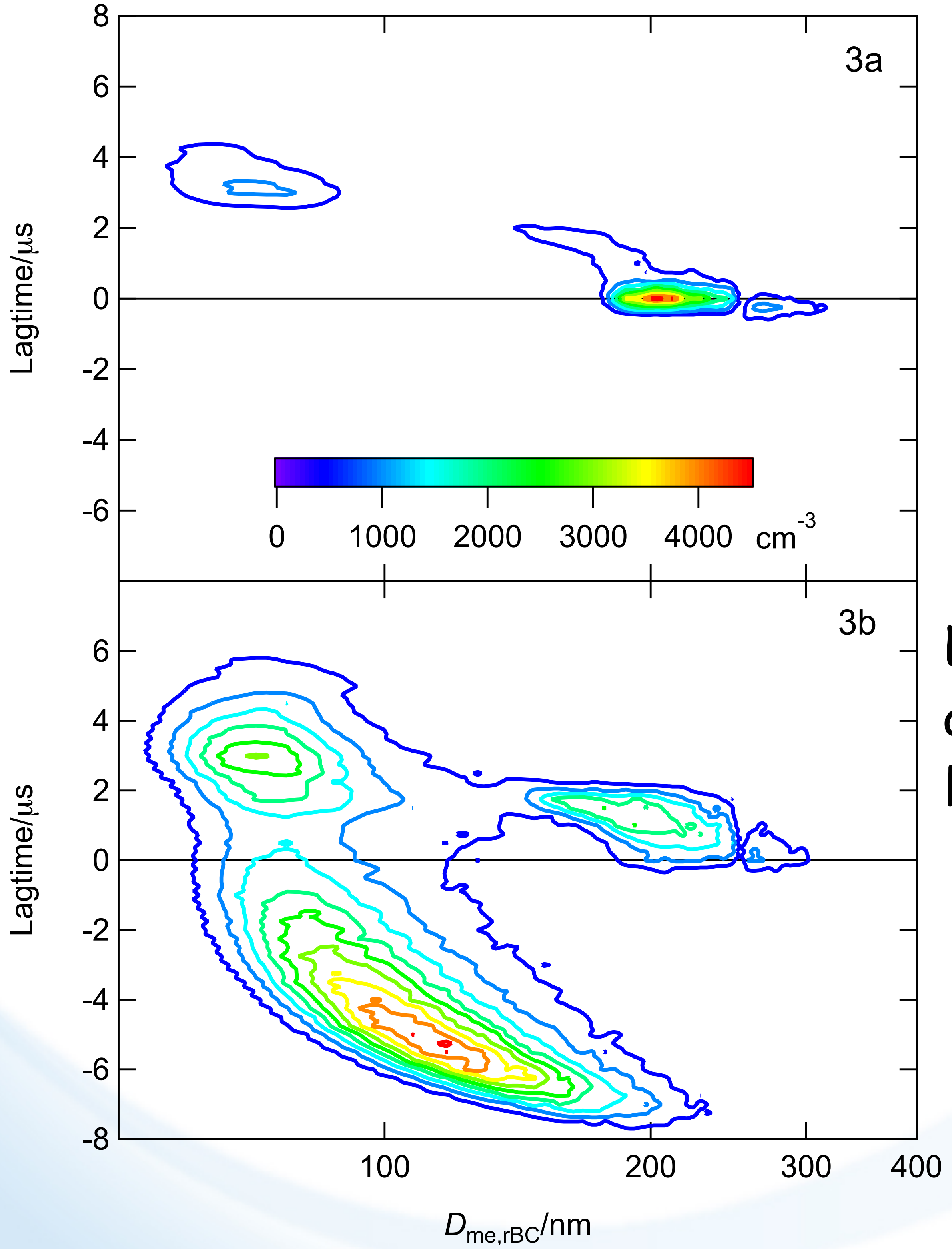


- Large increase in the mass conc of BC were observed
- 246±53° for episode A and 272±24° for episode B
- Pronounced increase in $D_{median, BC}$ for < 80 nm to near 100 nm in episode B
- High correlation with organic aerosol concentration
- O:C ratio indicates organic aerosol is aged

BROOKHAVEN NATIONAL LABORATORY

Lagtime behavior for this Plume Reveals Unique Insights

Sedlacek et al., (2012)



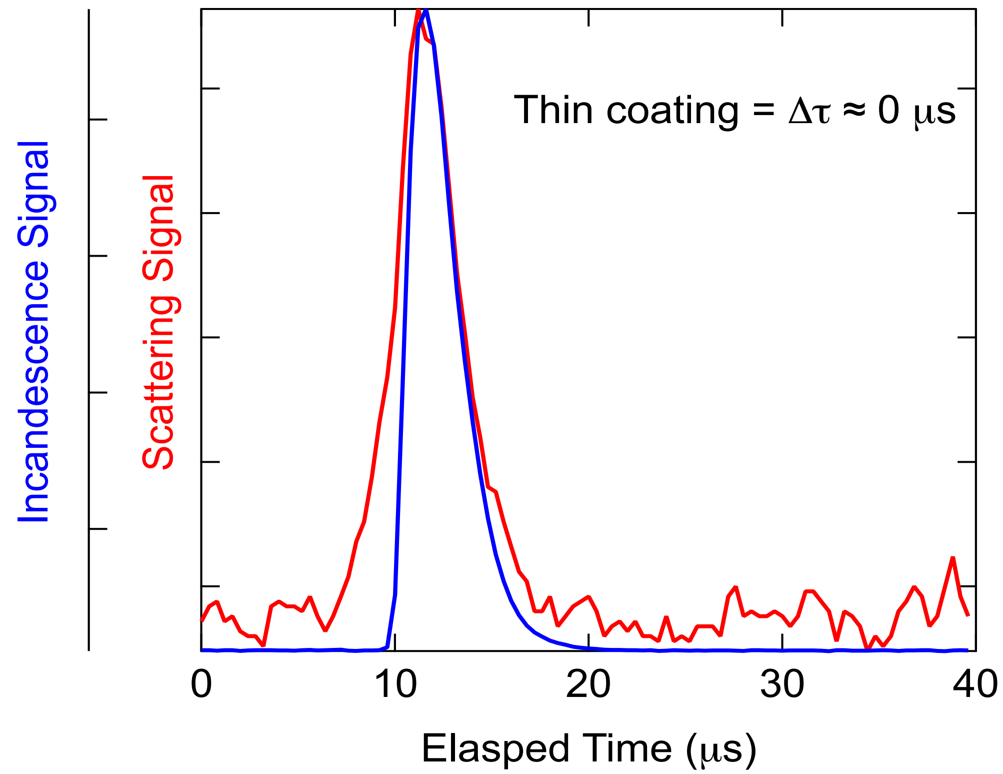
Episode A is dominated by short lagtimes for larger diameter rBC (thinly-coated) with some longer lagtimes associated with smaller diameter rBC (thickly-coated).

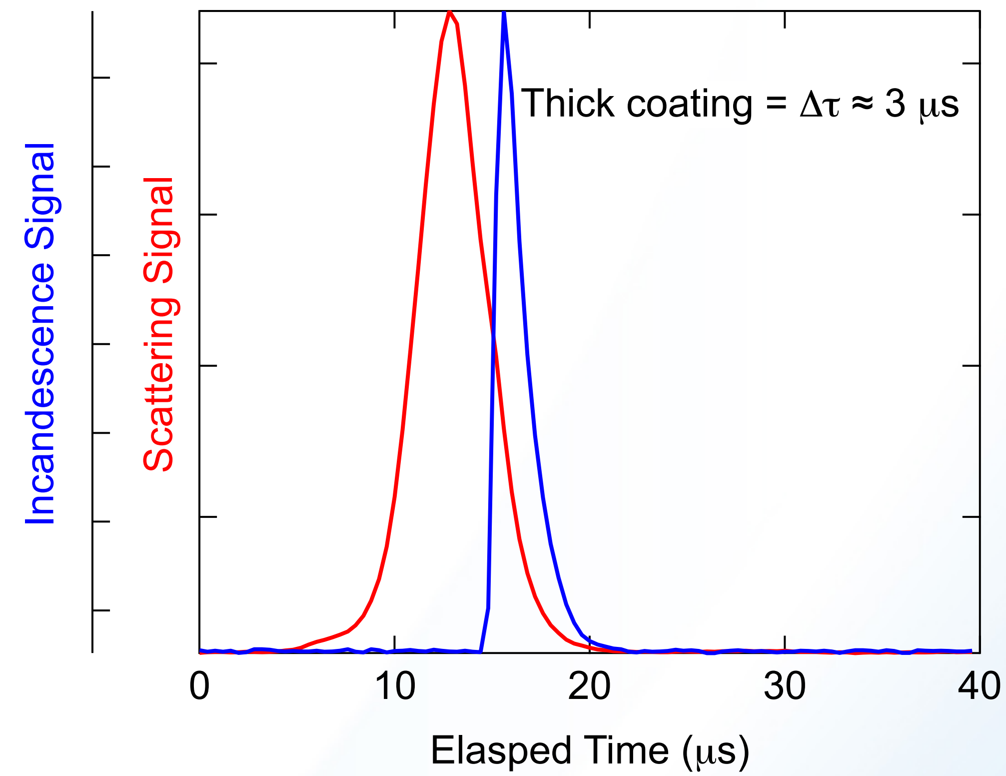
Episode B is characterized by negative lagtimes along with a bimodal distribution of positive lagtimes (thin/thick-coated rBC)

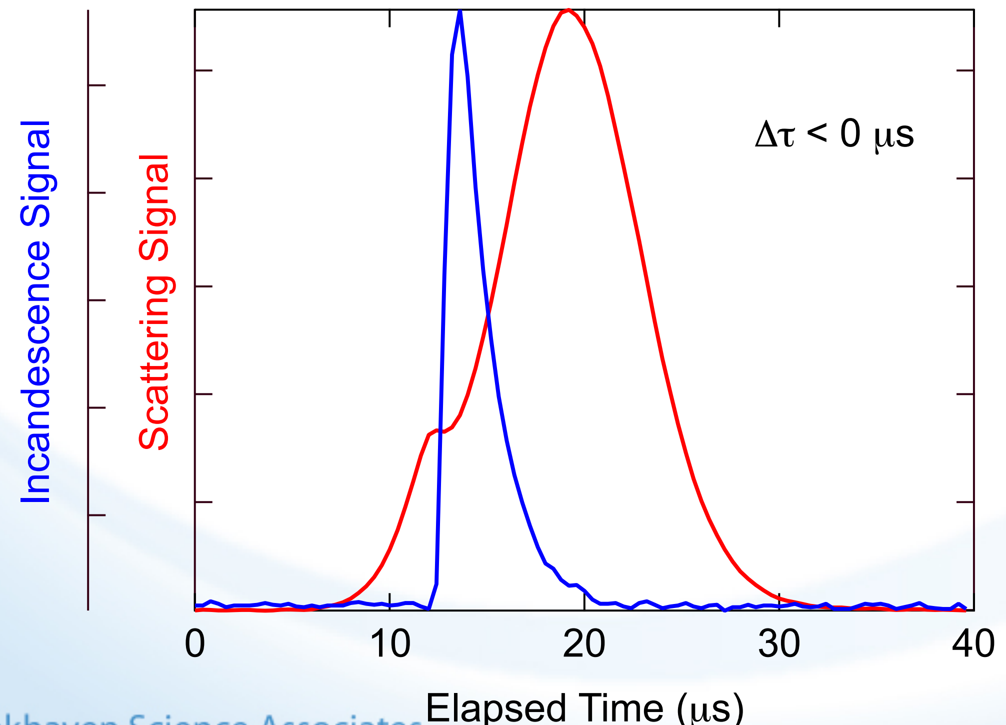
Proxy for Coating thickness: Incandescence Lagtime

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

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





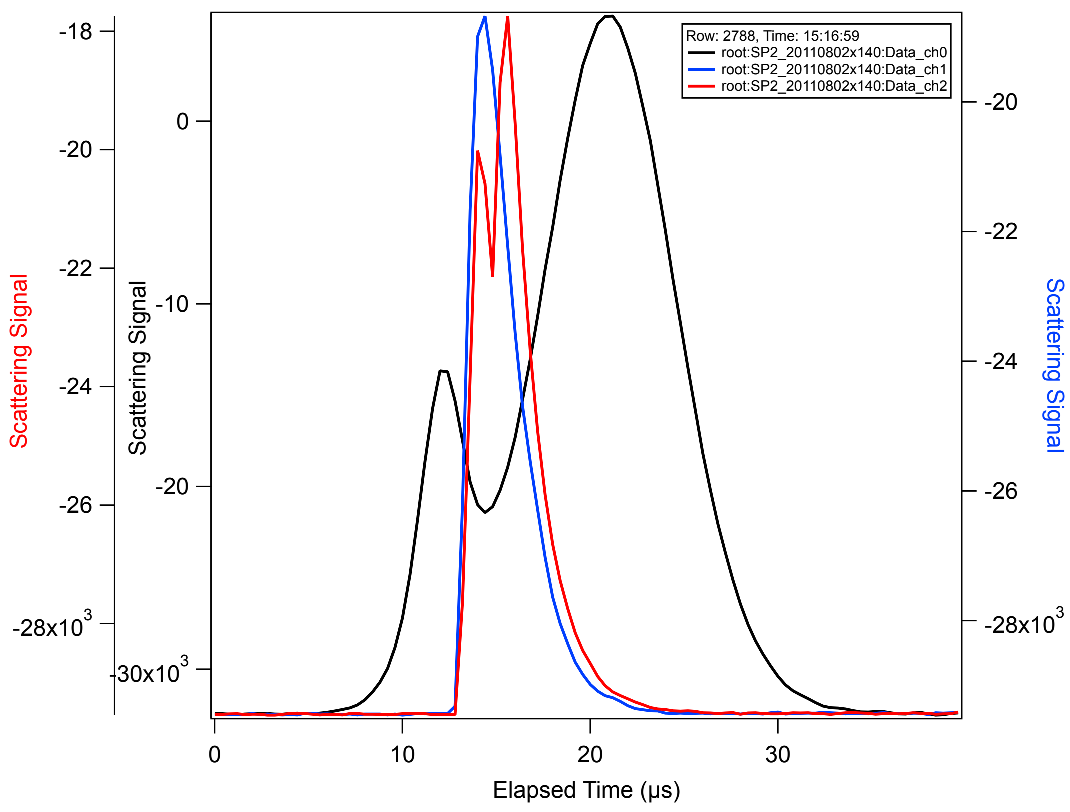


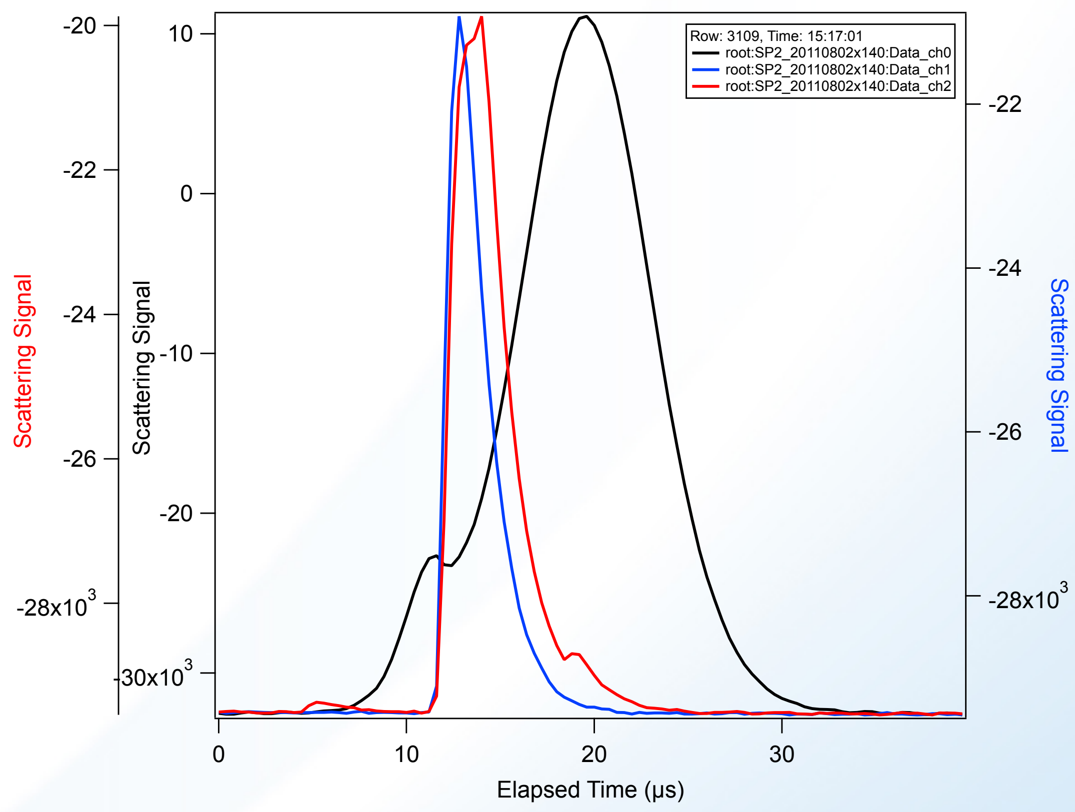
Negative lagtime (Sedlacek et al., GRL 2012)

- Most of scattering occurs after incandescence.
- Complex scattering signal contains information on particle break up.

Lagtime behavior for this Plume - particle analysis

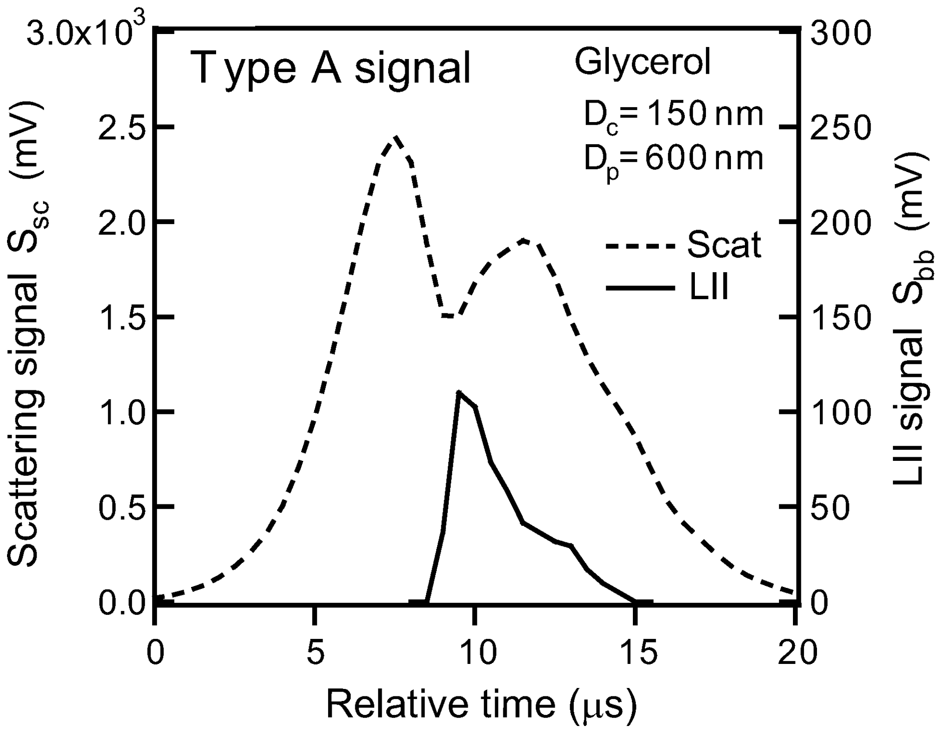
Examination of the temporal behavior of the scattering channel reveals the presence of complex behavior





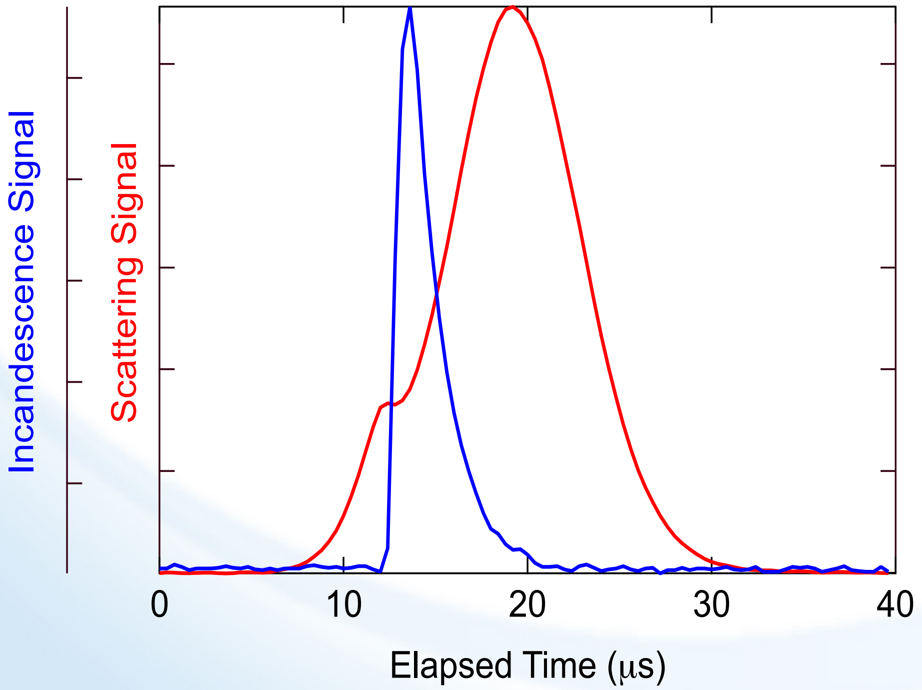
Over 60% of rBC particles in this plume exhibit this behavior

Negative Lagtimes and Fracturing of Coated BC Particle



Moteki and Kondo (AST, 2007) observed similarly complex scattering signals for thickly-coated graphite:

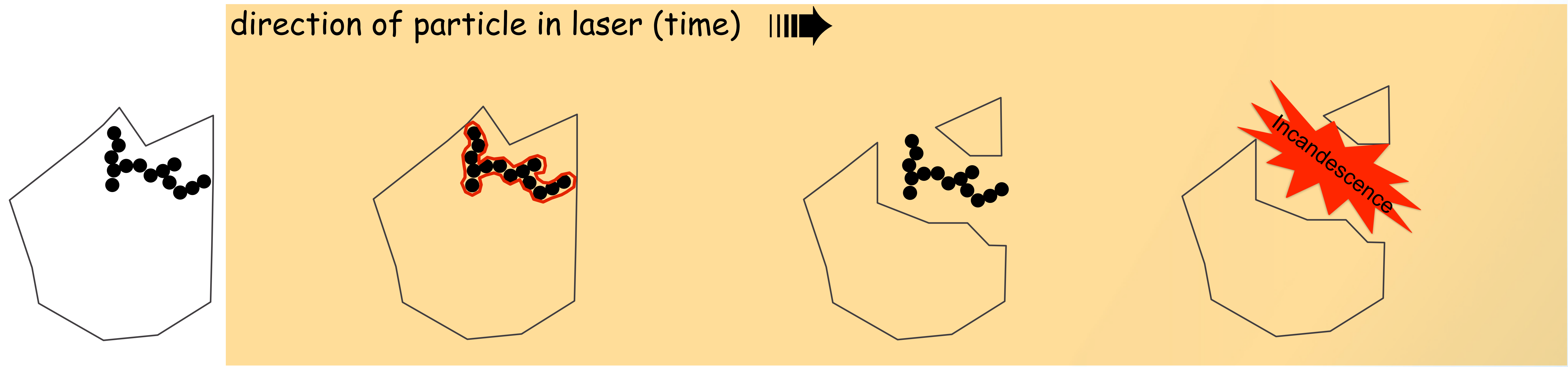
1. Evaporation rate of coating at the core-coating interface is faster than at the outer surface.
2. Coated particle breaks into BC core and coating material.
3. 'Denuded' core immediately undergoes incandescence while the rBC-free coating continues through laser.



Pronounced asymmetry in the scattering amplitudes is attributed to rBC located near or at the surface of the 'host' material.

Asymmetric Signal Amplitudes

Asymmetric scattering amplitudes could be explained by a rBC core that is located near the surface



nascent particle enters laser beam

coating material at core/ coating interface vaporizes

vaporized material fractures particle early in laser

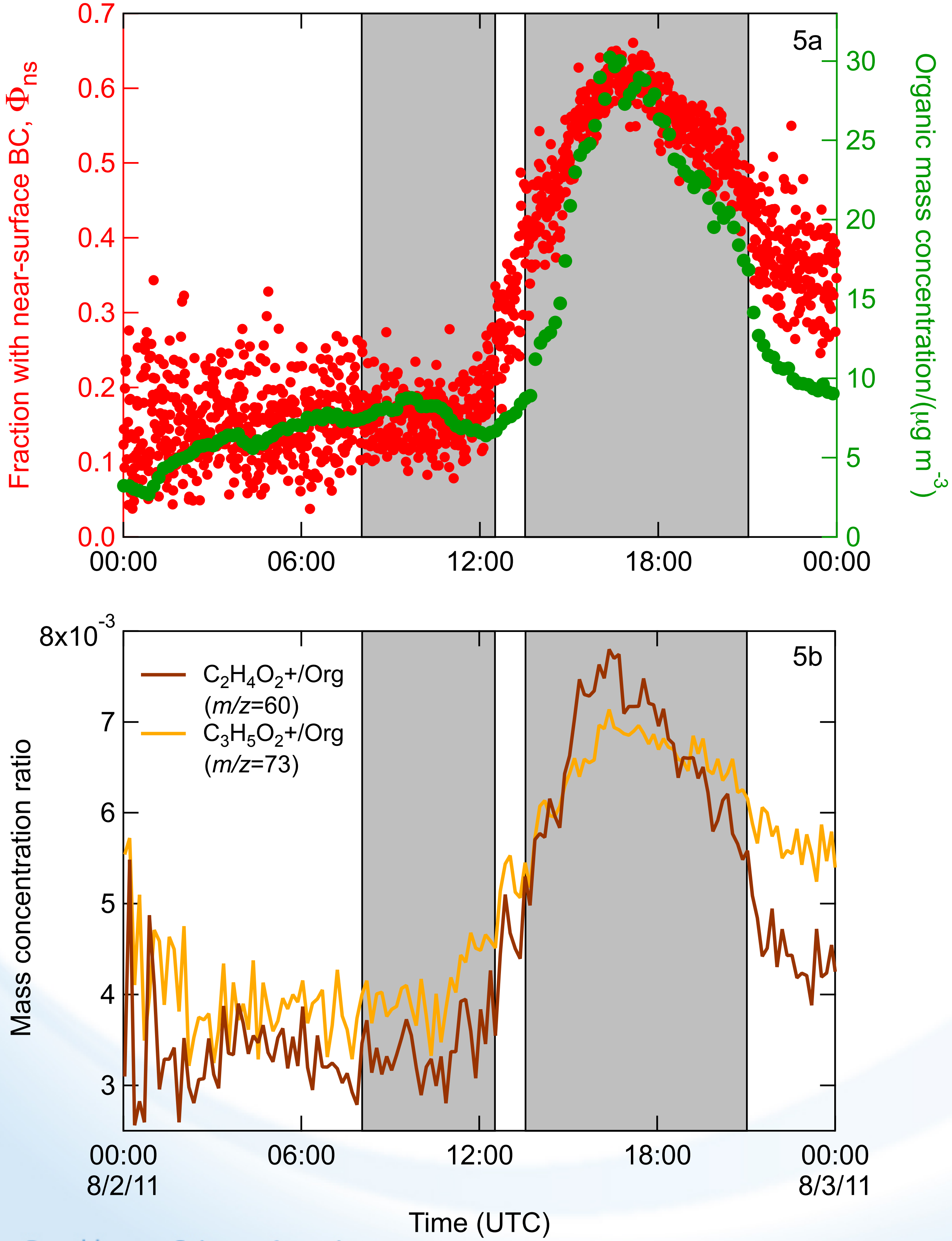
Denuded BC incandescences & pure scatters continue through laser beam

TEM analysis of Mexico City soot by Adachi et al., (2010) reported that:

> "long distances between the mass centers of the soot and those of the host particles suggest that most soot lies near the surface rather than center of the host particles."

What is the origin of near-surface rBC-containing particles?

Episode B Contains Markers for Biomass Burn



Likely formation mechanism is condensation of organic material onto rBC particles

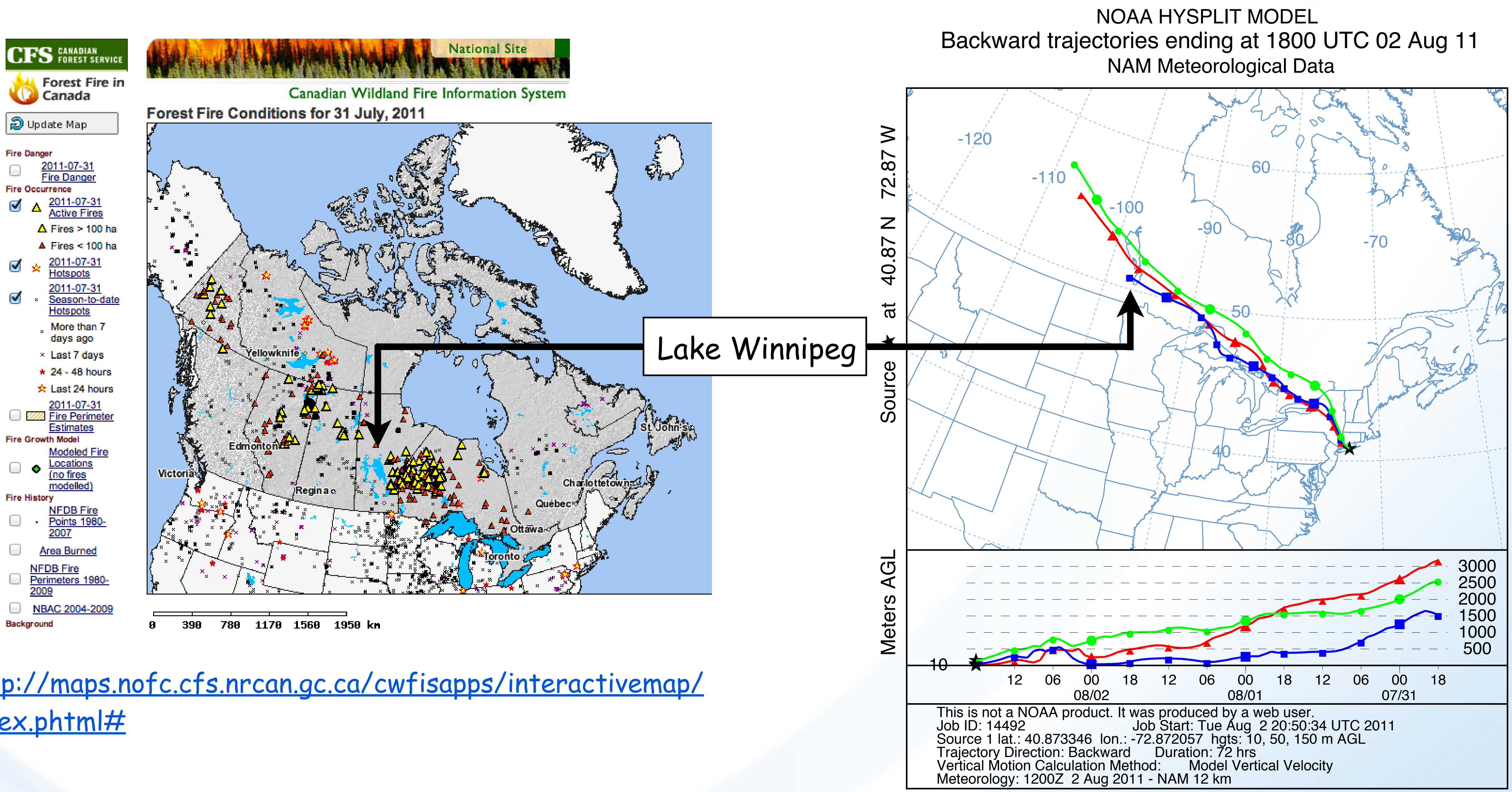
What is the potential source for these biomass burn markers?

Sedlacek et al., (GRL, 2012)

BROOKHAVEN NATIONAL LABORATORY

Potential Aerosol Source for Episode B

Three-day back trajectory calculations suggest that the advected air mass should contain telltale signs of biomass burning (BB).



http://maps.nofc.cfs.nrcan.gc.ca/cwfisapps/interactivemap/index.phtml#

Are near surface rBC-containing particles unique to biomass burns?

Conclusions

Nugget 1: SP2 can provide details into the structure of rBC at high-time resolution

- Optical diameter method can quantify coating thickness for the core-shell configuration
- Lagtime method can probe for the presence of near-surface rBC-containing particles

Nugget 2: Near-surface rBC-containing particles appear to be associated with biomass burning

Follow on research questions

- How common is this class of near-surface BC containing particles?
  40% of BC emissions are from biomass burns; 20% from open-pit cooking
- What is the impact of near-surface rBC containing particles on radiative forcing?
  Buseck and co-workers (Adachi etal.., 2010) have calculated a 20% reduction in soot direct forcing due to non-concentric core-shell configuration
- What kind of experiments are needed?
  Field campaign focused on biomass burns (proposal invited for submission)
  Laboratory-based studies (Boston College study starting in spring of 2012)