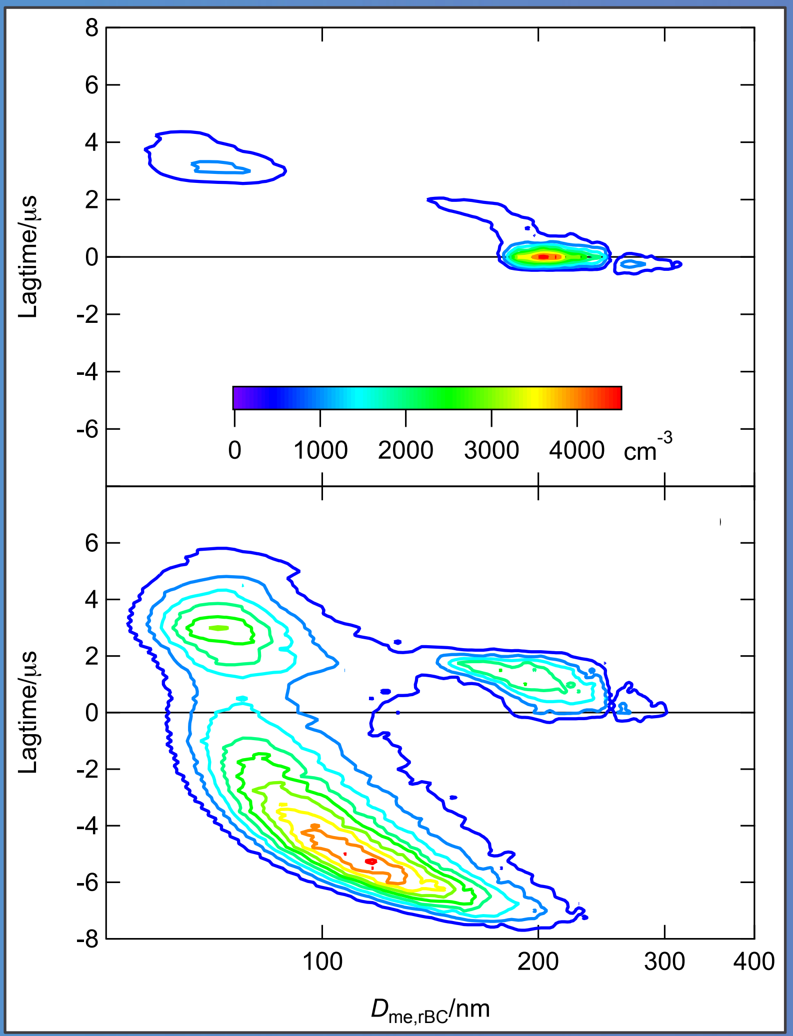
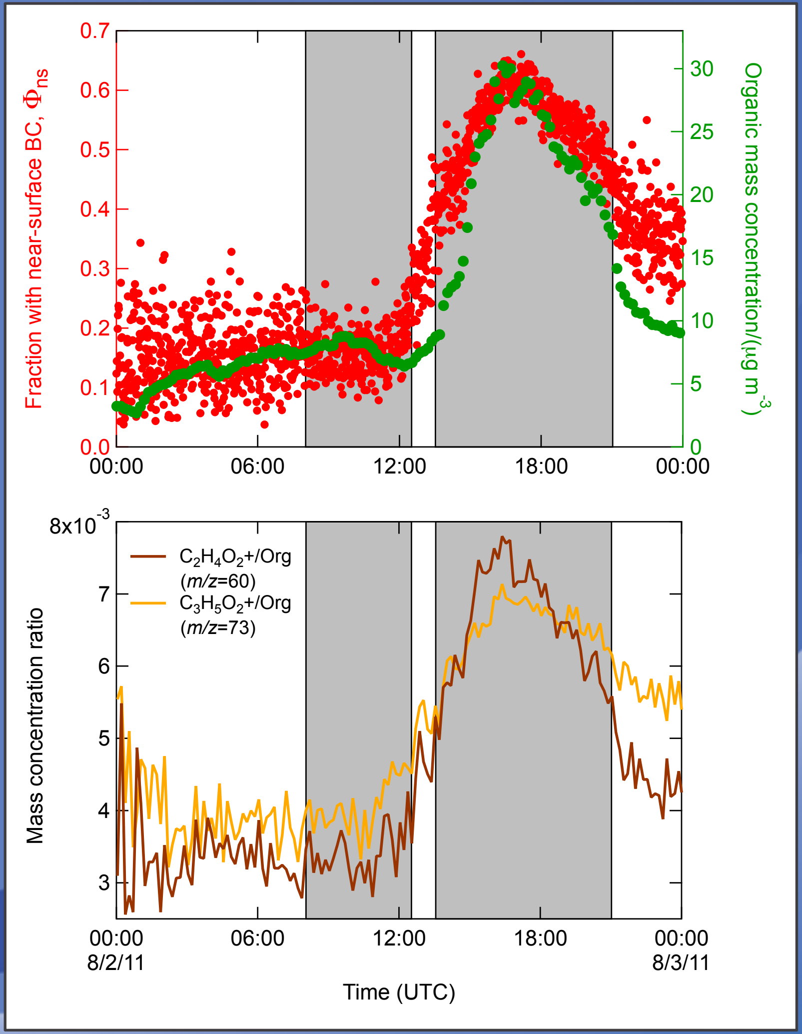
Determination of and Evidence for Non-core-shell Structure of BC-containing Particles using SP2

A. J. Sedlacek, E. R. Lewis, L. Kleinman (BNL)
J. Xu and Q. Zhang (UC/Davis)





ARM
CLIMATE RESEARCH FACILITY

BROOKHAVEN NATIONAL LABORATORY
a passion for discovery

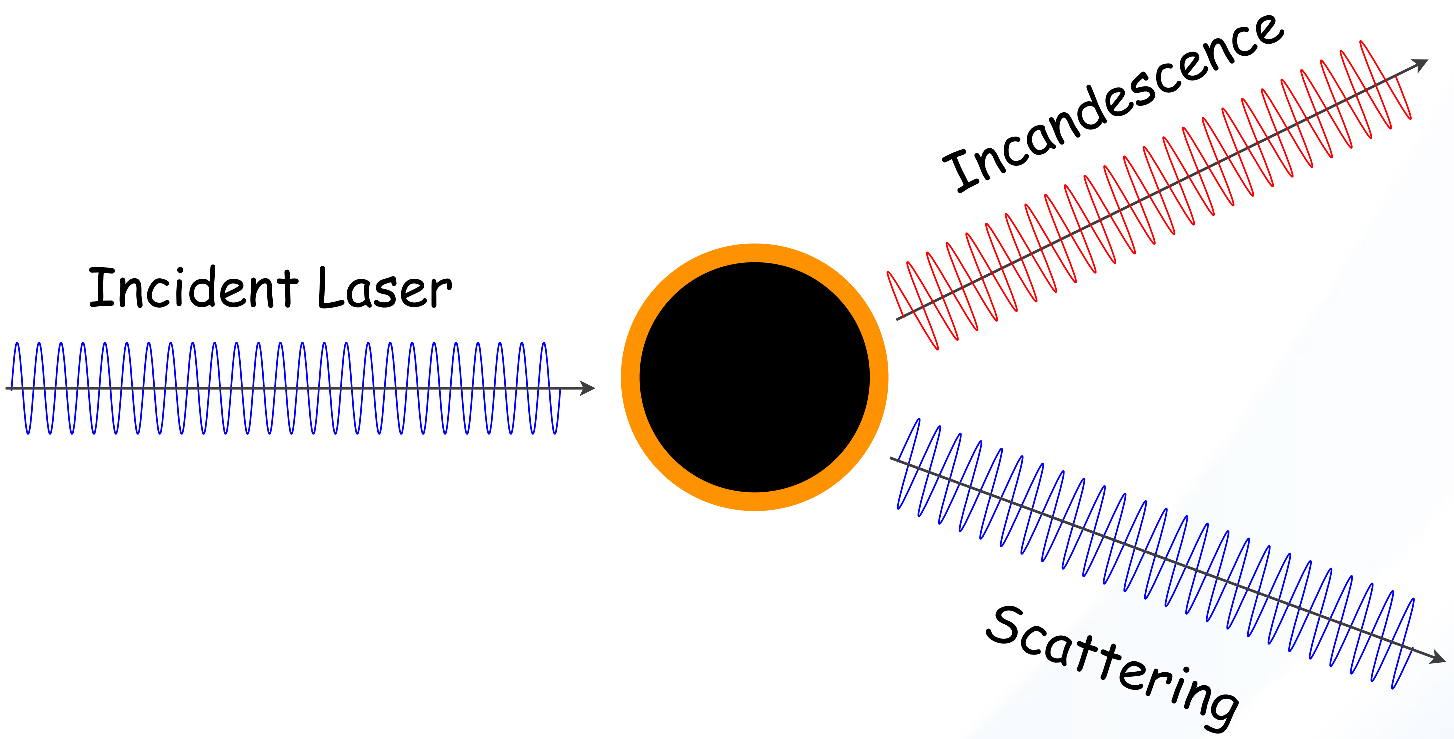
Office of Science
U.S. DEPARTMENT OF ENERGY

ASR
Atmospheric System Research

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:

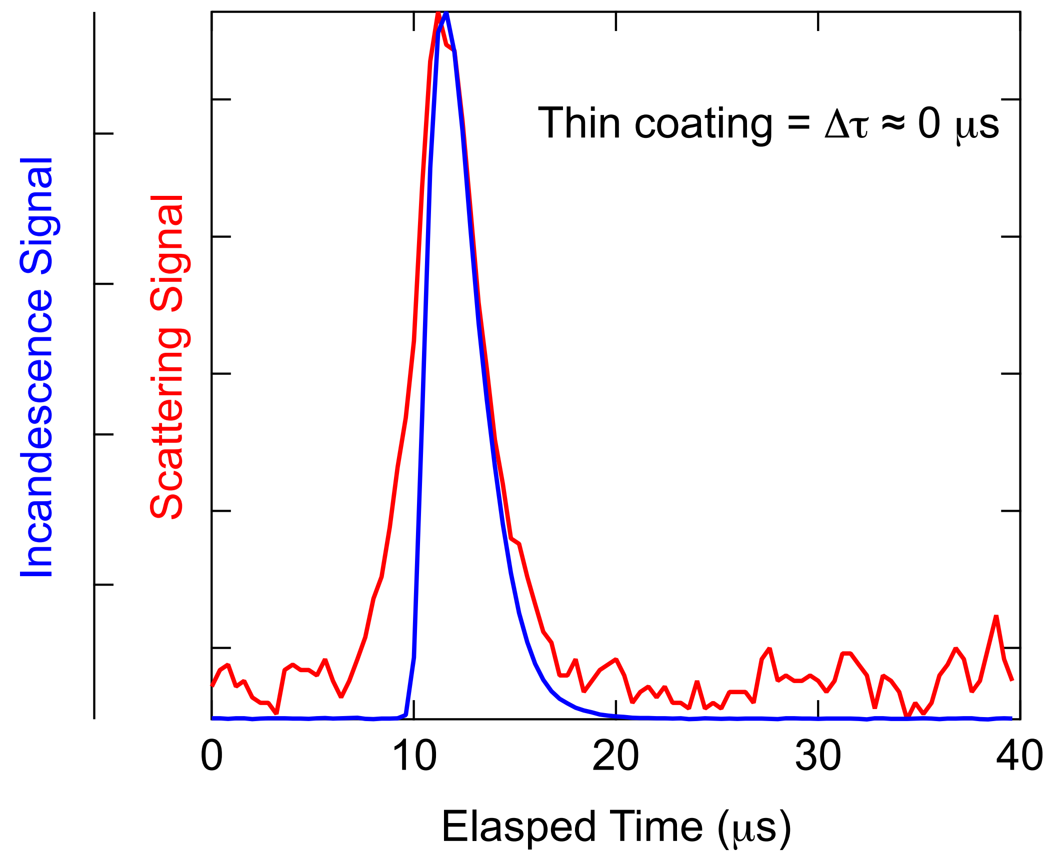
- Probe coating thickness: optical and BC mass equivalent diameters
- Examine temporal profiles of the scattering and incandescence signals

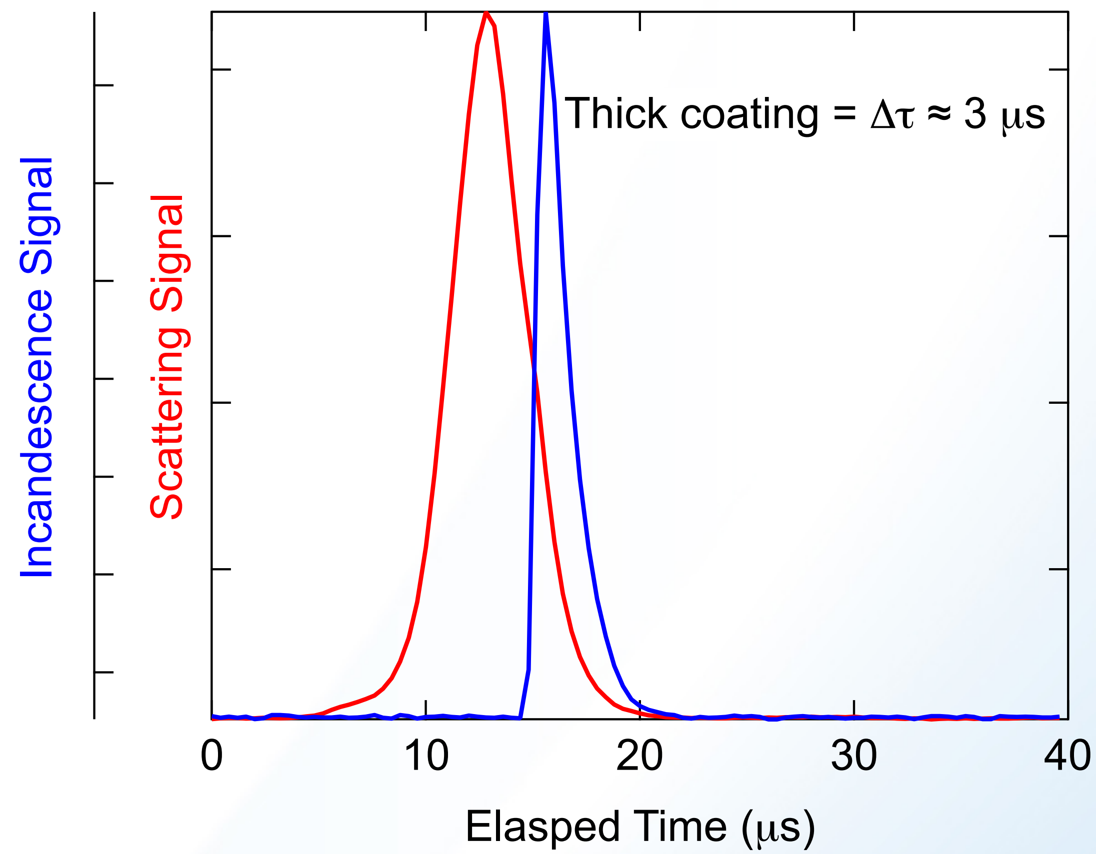
BROOKHAVEN NATIONAL LABORATORY

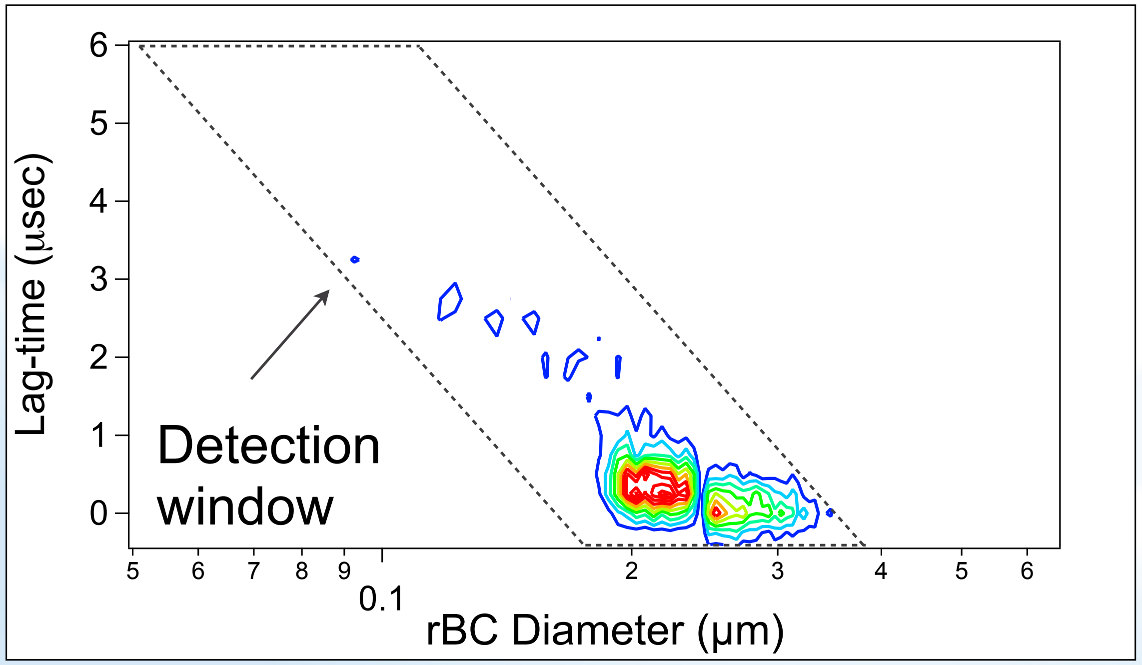
Proxy for Coating thickness: Incandescence Lagtime

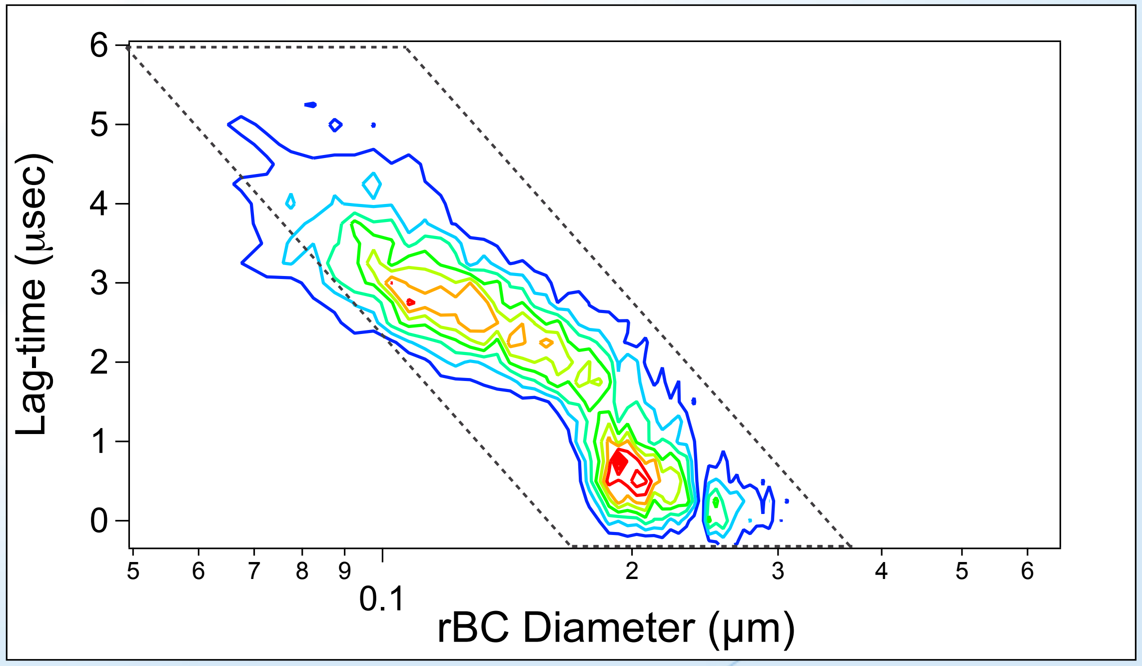
Moteki and Kondo, 2007; Subramanian et al., 2010

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

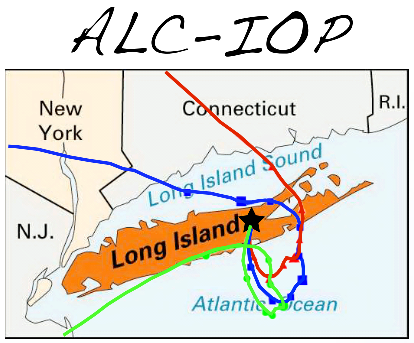




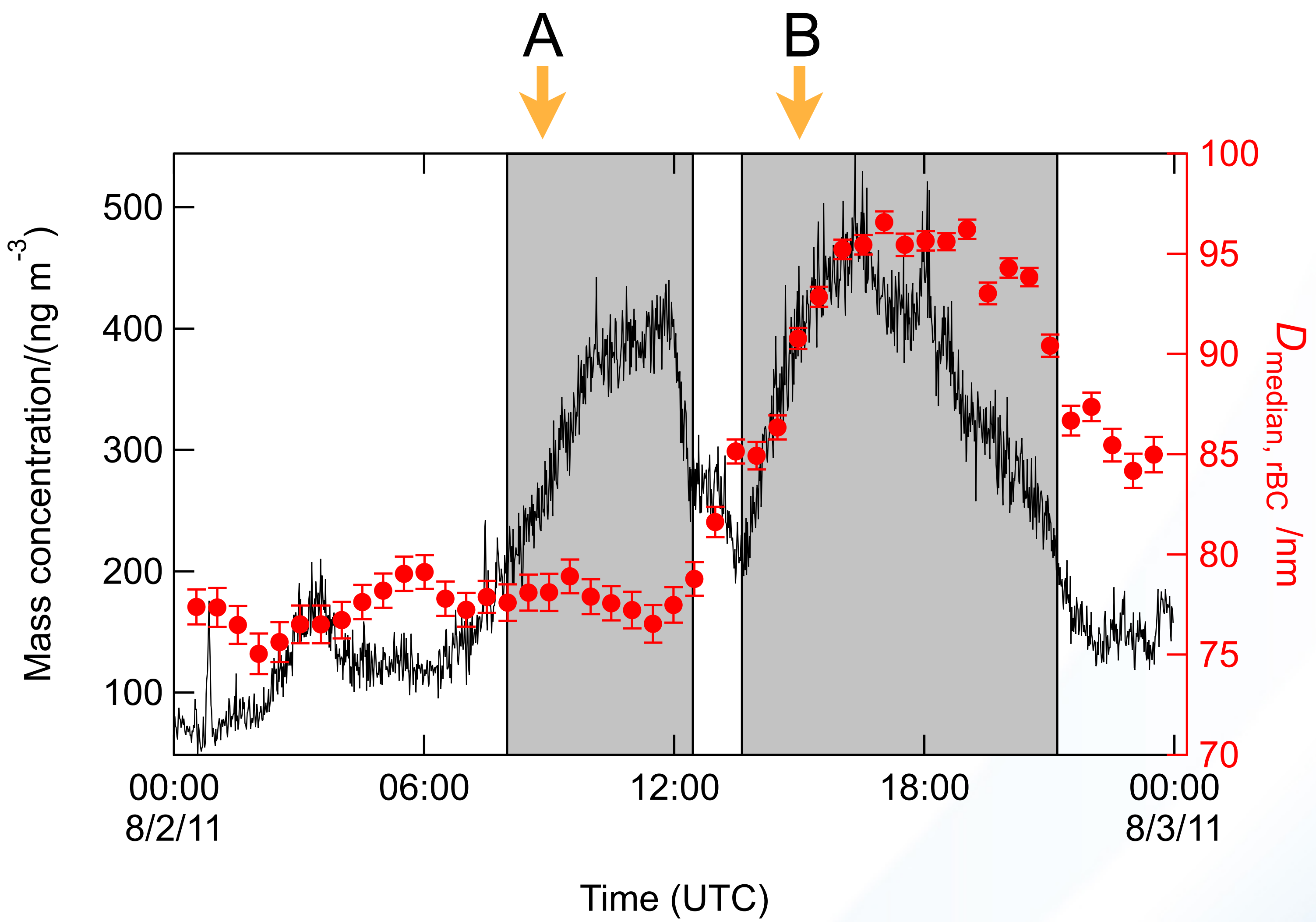




BNL-IOP: August 2, 2012



Characterized by two episodes: A and B

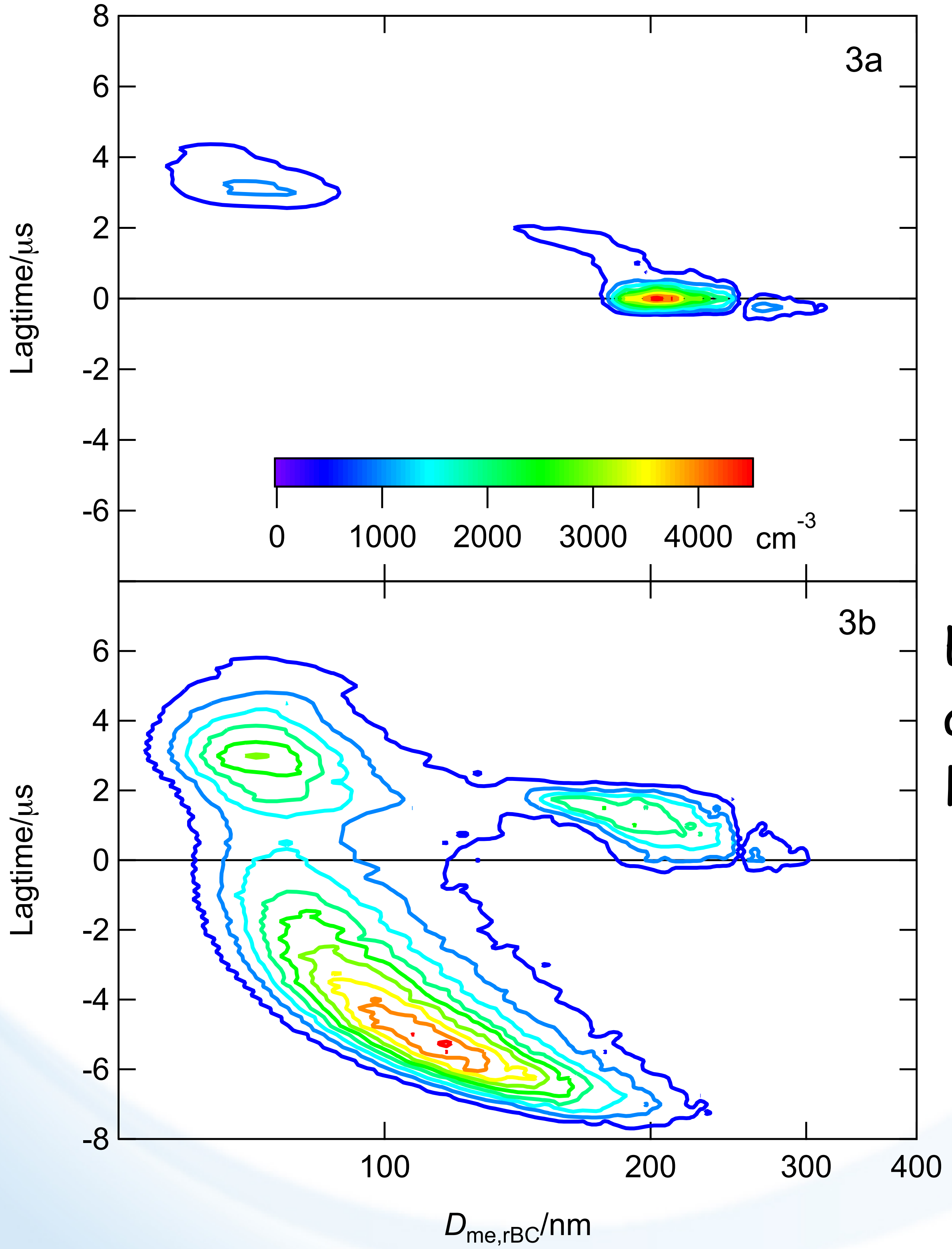


- Large increase in the mass conc of BC were observed
- 246±53° for episode A and 272±24° for episode B
- Pronounced increase in $D_{median,\ BC}$ for < 80 nm to near 100 nm in episode B

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).

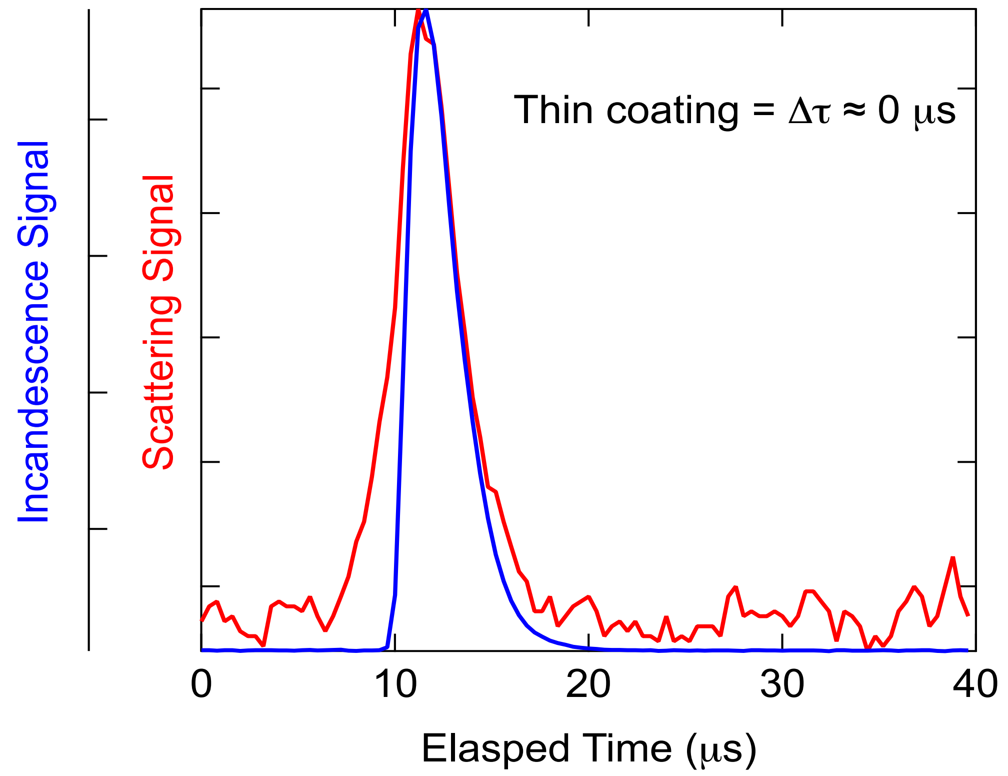
Episode B is characterized by negative lagtimes along with a bimodal distribution of positive lagtimes (thin/thick-coated rBC)

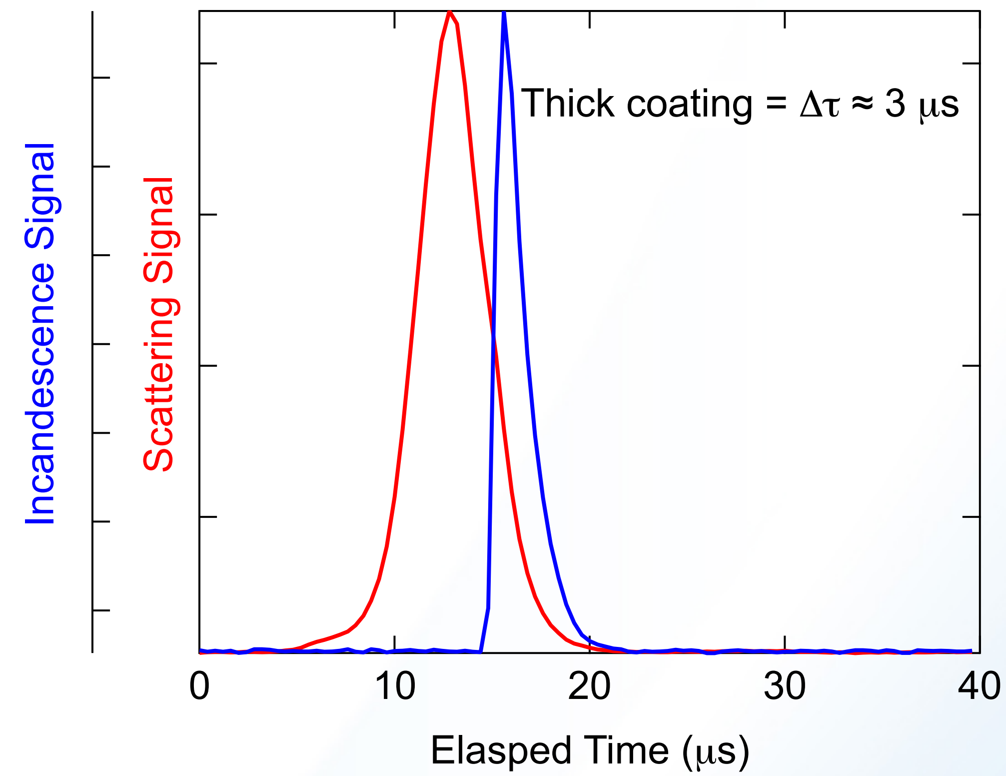
BROOKHAVEN NATIONAL LABORATORY

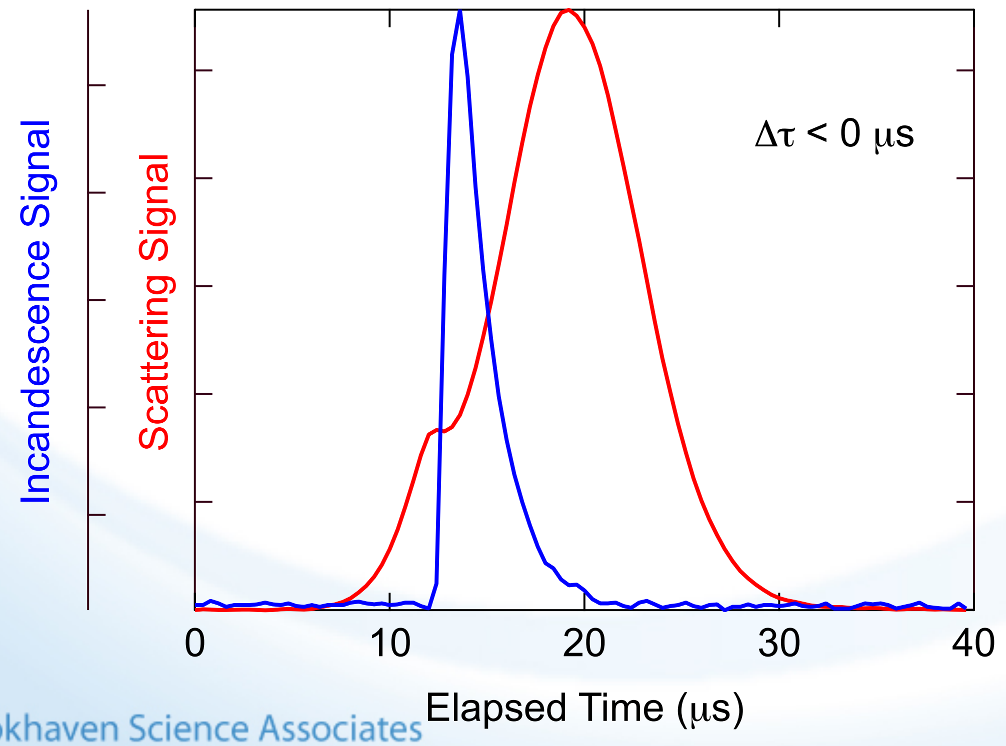
Proxy for Coating thickness: Incandescence Lagtime

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

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





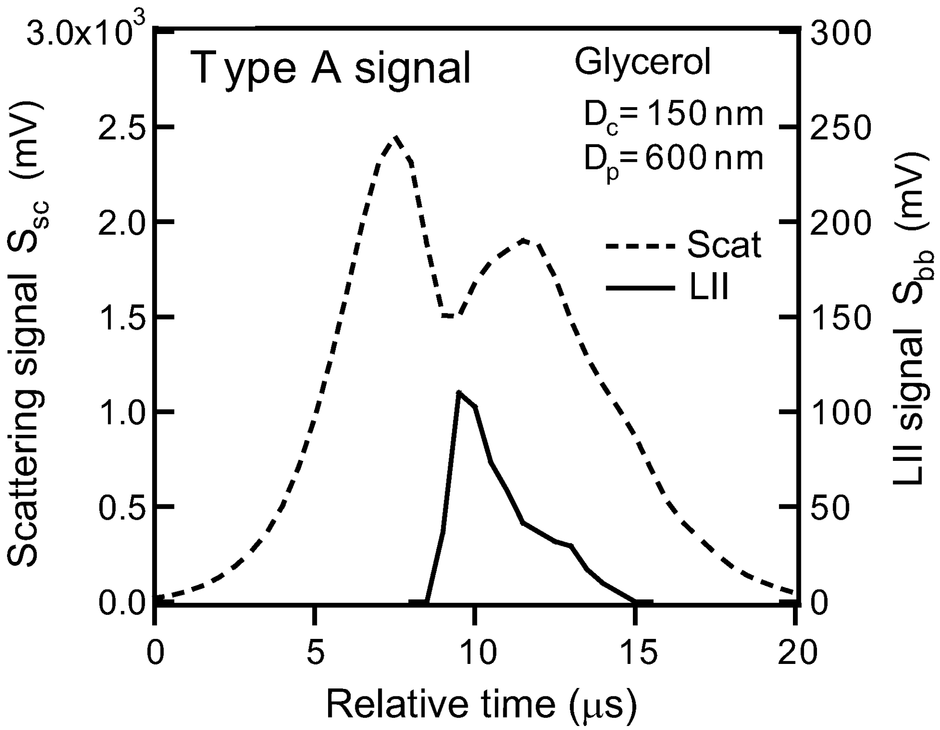


Negative lagtime (Sedlacek et al., GRL 2012)

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

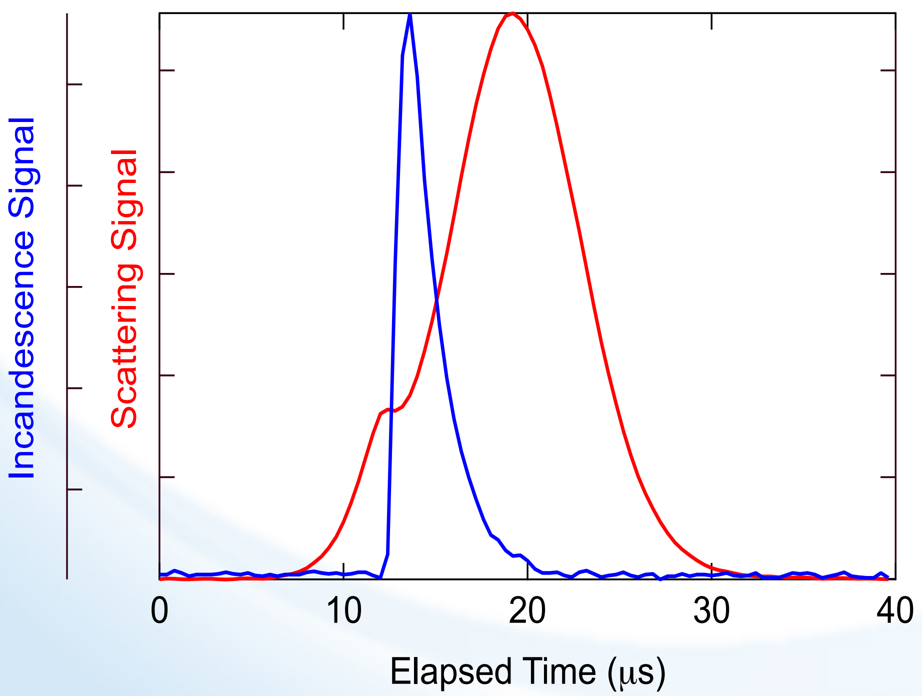
BROOKHAVEN NATIONAL LABORATORY

Negative Lagtimes and Fracturing of Coated BC Particle



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

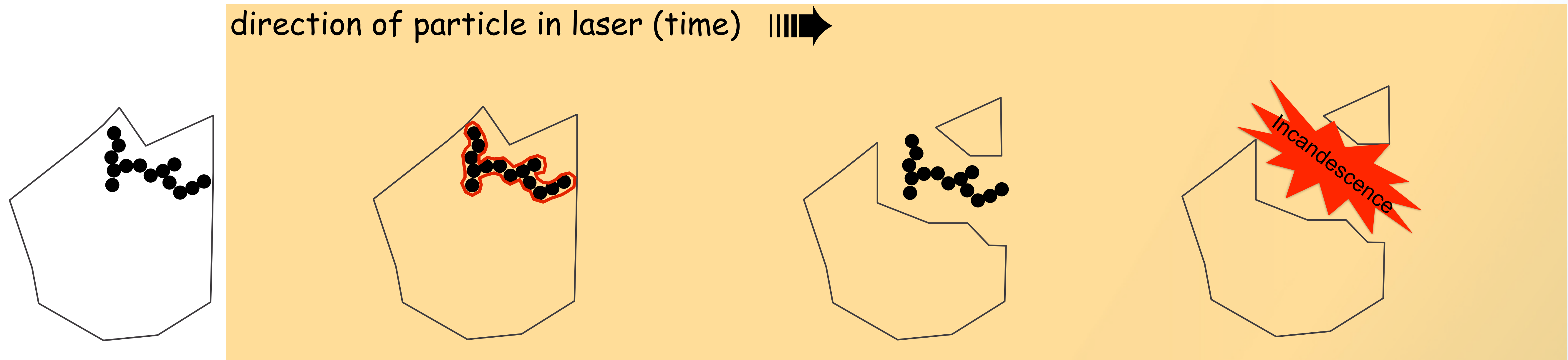
1. Evaporation rate of coating at the core-coating interface is faster than at the outer surface.
2. Coated particle breaks into BC core and coating material.
3. 'Denuded' core immediately undergoes incandescence while the rBC-free coating continues through laser.



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

Source of Negative Lagtimes: rBC near surface of particle

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

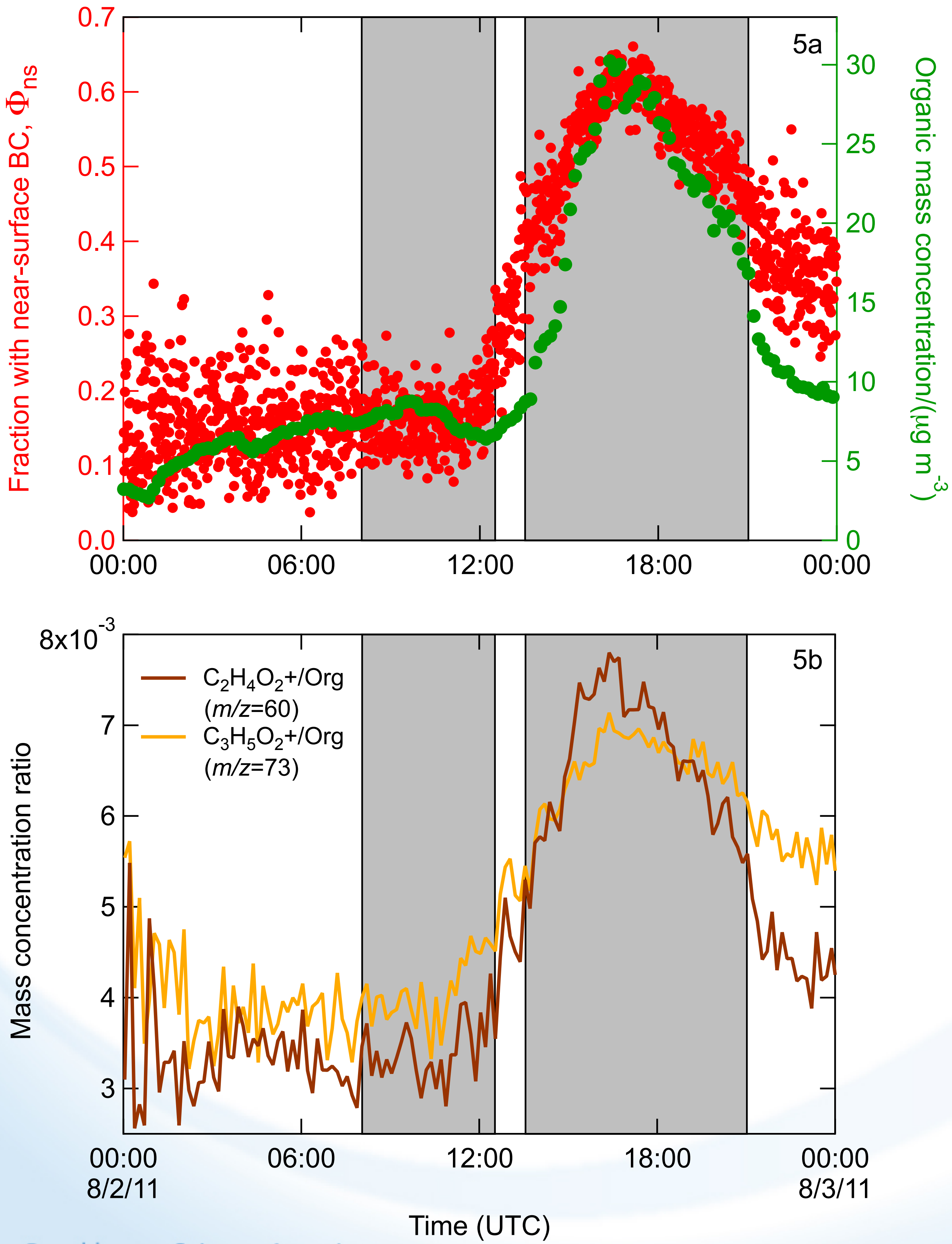
Denuded BC incandescences & pure scatters continue through laser beam

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



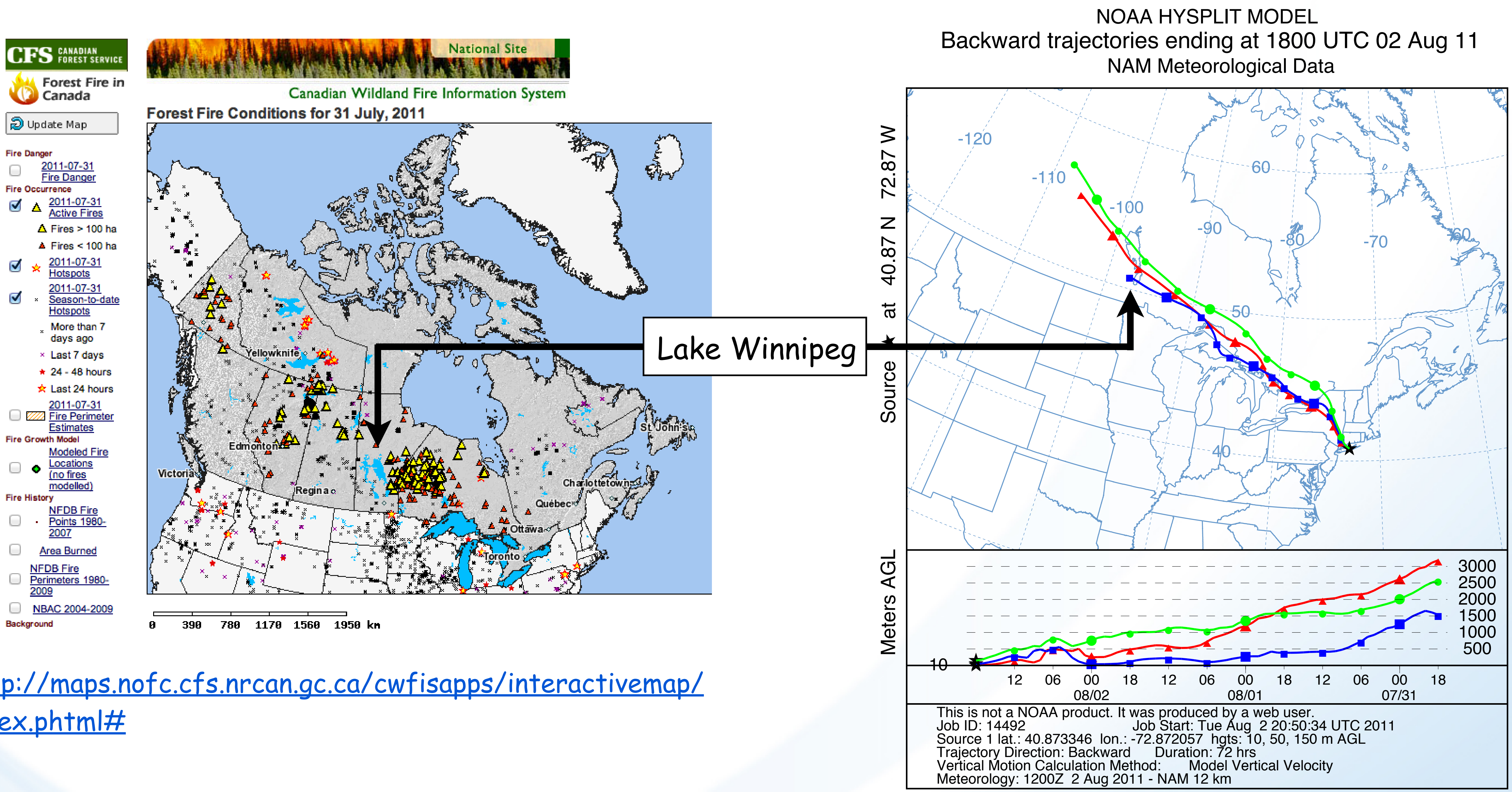
Likely formation mechanism is condensation of organic material onto rBC particles

What is the potential source for these biomass burn markers?

Sedlacek et al., (GRL, 2012)

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).



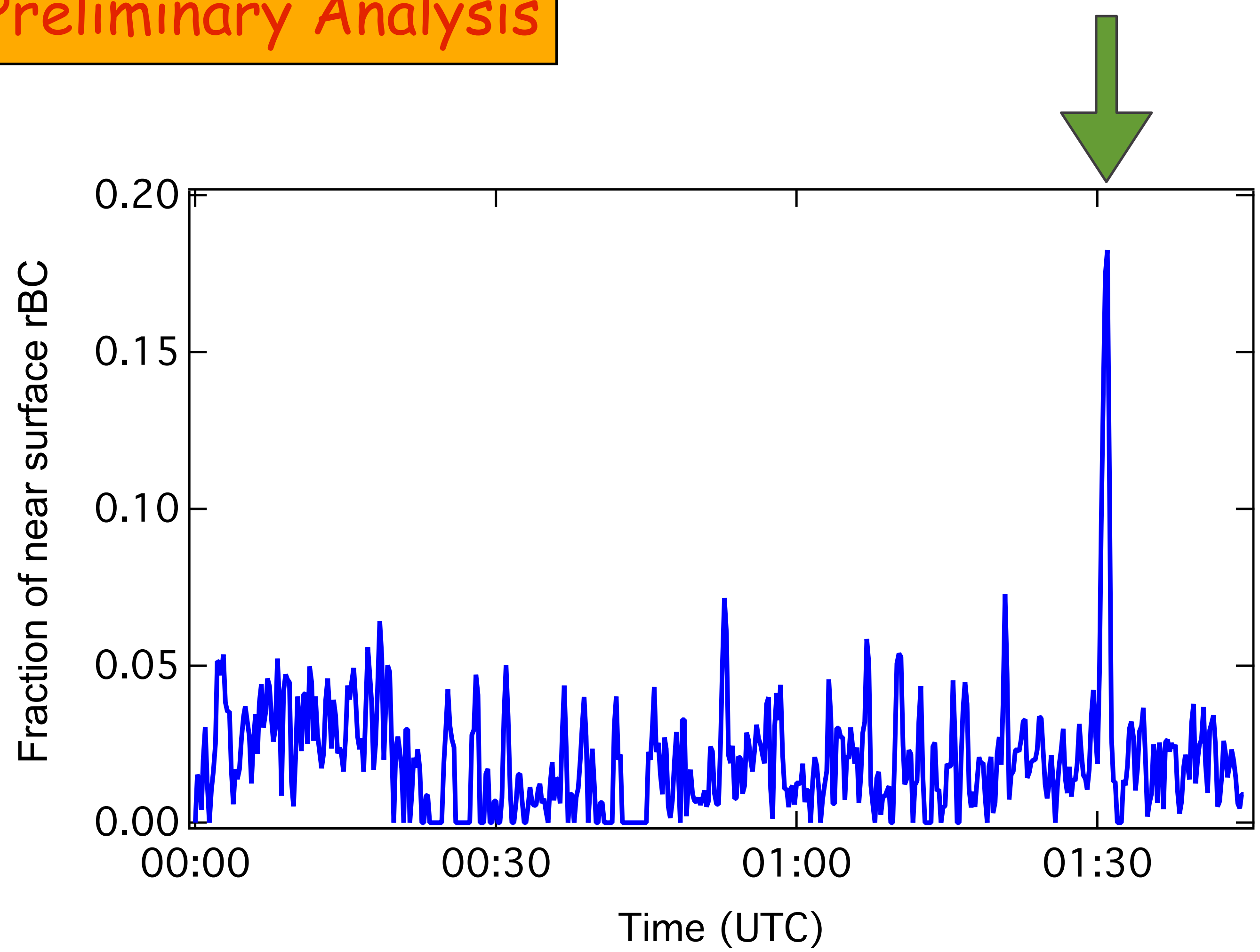
http://maps.nofc.cfs.nrcan.gc.ca/cwfisapps/interactivemap/index.phtml#

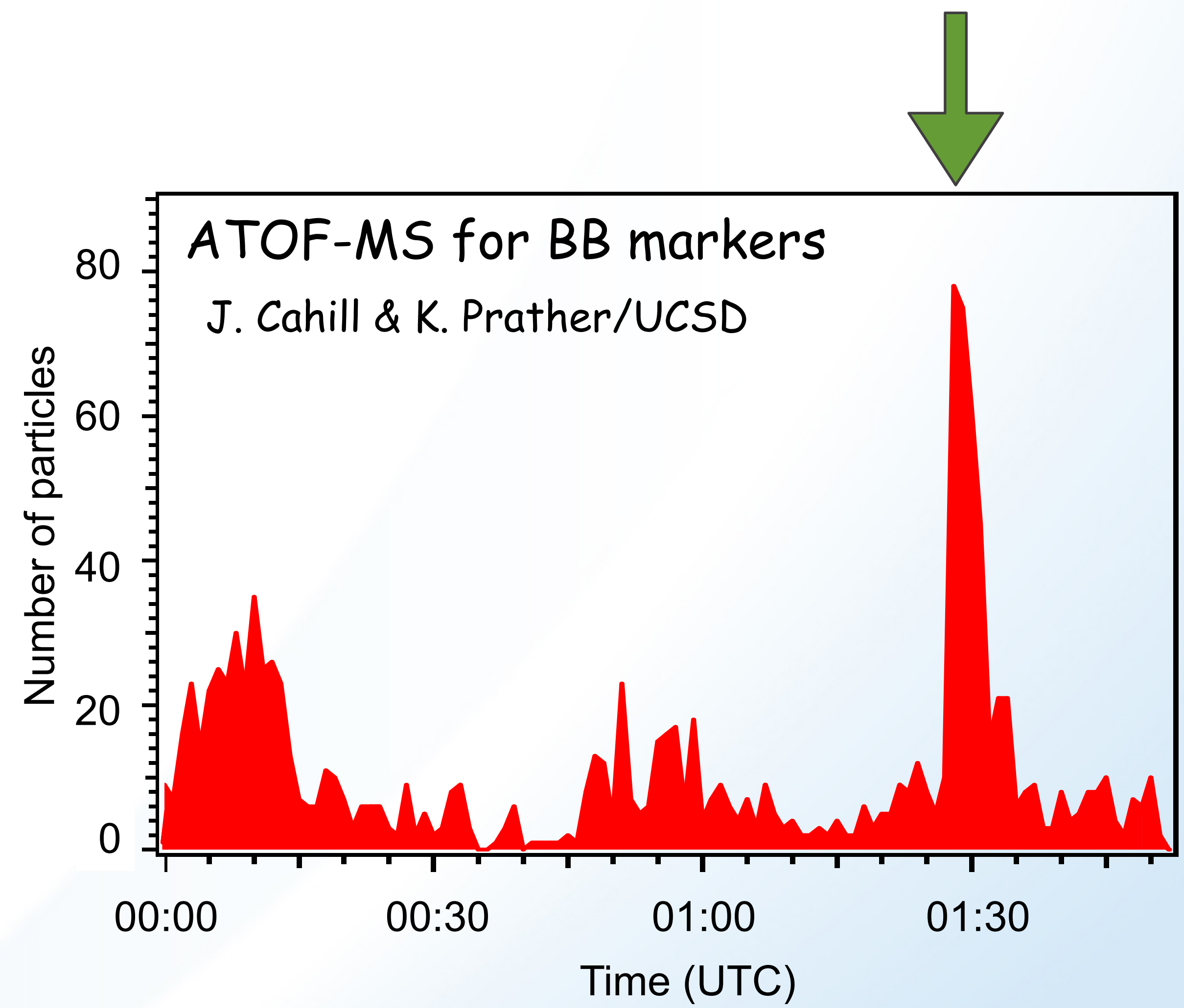
Are near surface rBC-containing particles unique to biomass burns?

Near Surface rBC: Supporting Material - CARES

June 19, 2010, DOE G-1 crossed a grass fire plume at 01:30

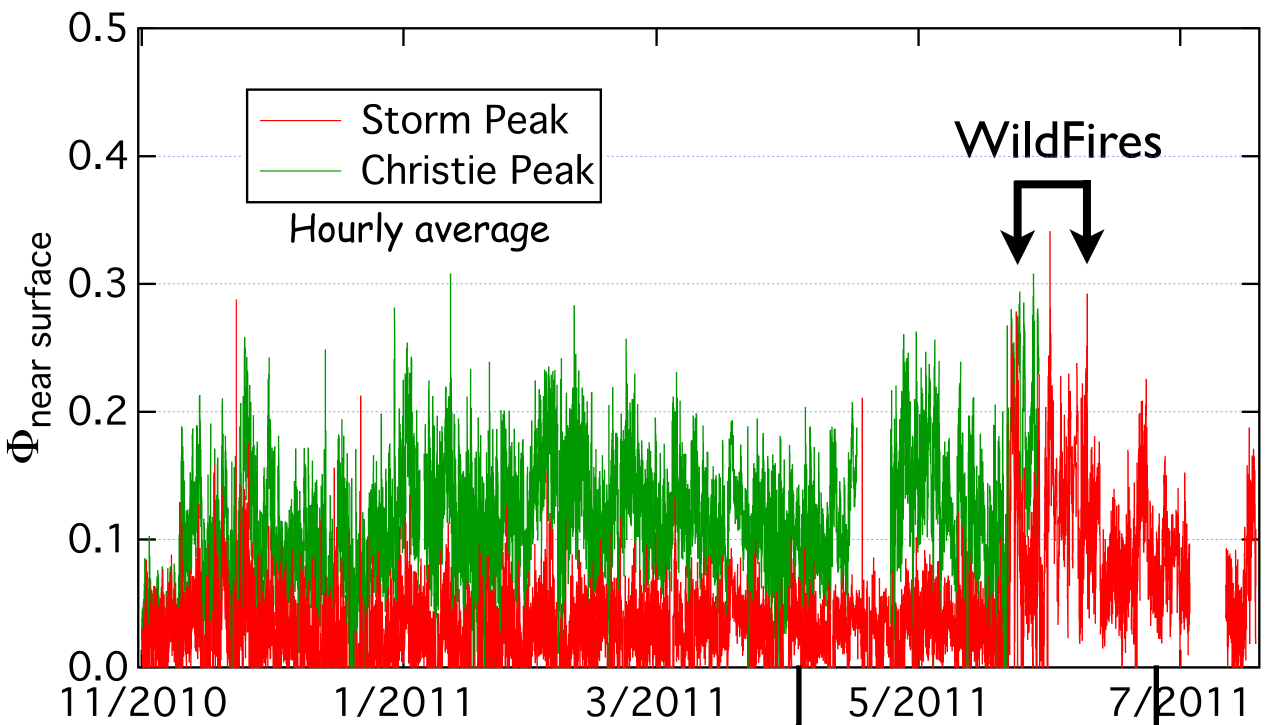
Preliminary Analysis





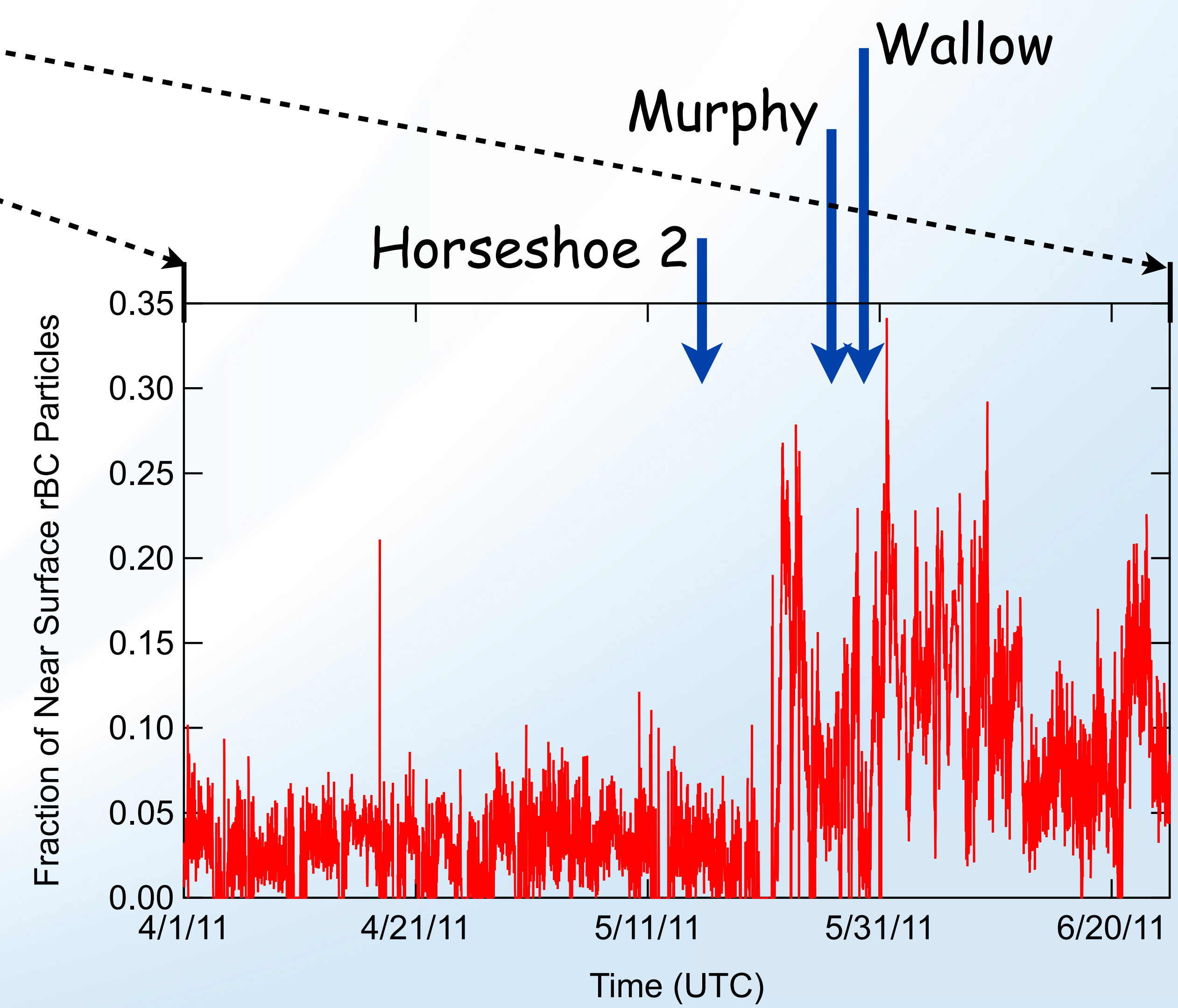
Arizona/New Mexico wildfires provided a target of opportunity during StormVEx

Near Surface rBC: Supporting Material - StormVEx



STORMVEx

- Consistently higher $\Phi_{near\ surface}$ at Christie Peak is tentatively ascribed to the influence of residential wood burning.

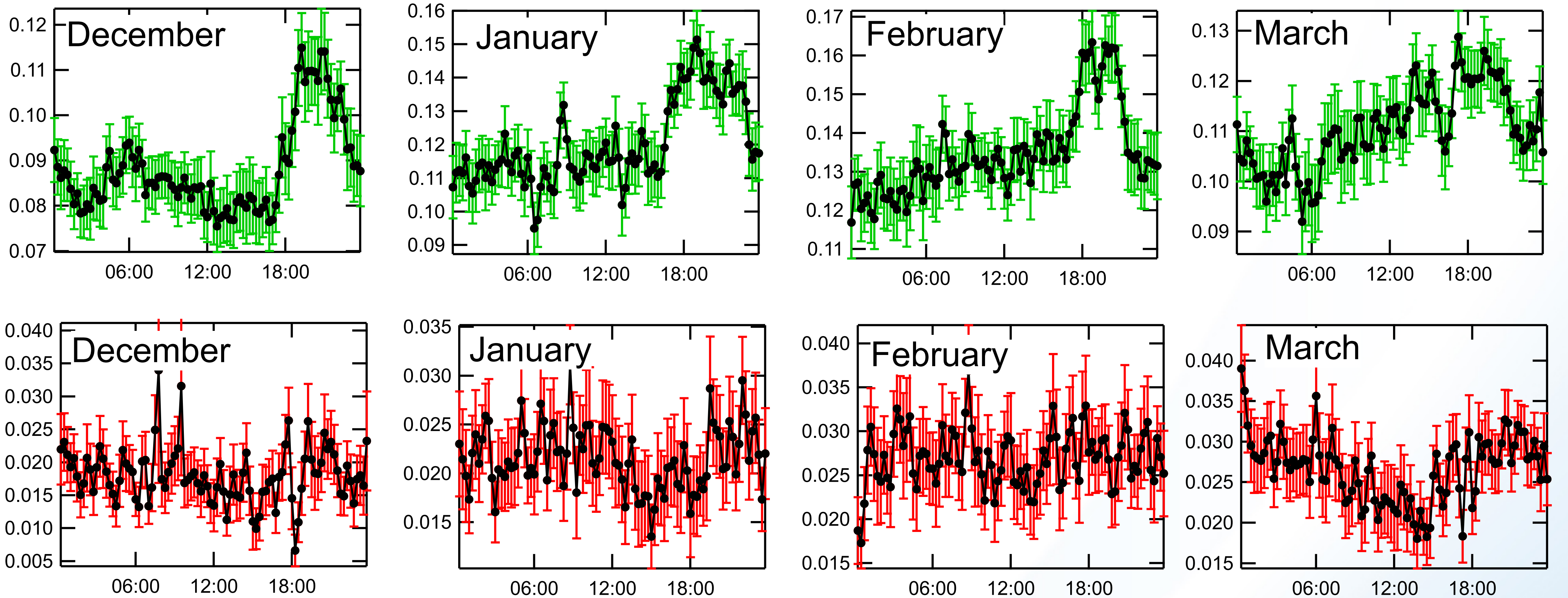


- Increase in $\Phi_{near\ surface}$ of >2x at Storm Peak lab in June due to wildfires. Observation consistent with earlier finding (Sedlacek et al., 2012). Christie Peak shutdown in May.

Preliminary Analysis

Diurnal Variation of Near-Surface Fraction of rBC

Christie Peak
Storm Peak



- Due to growth of boundary layer, valley emissions extend to Christie Peak.
- Increase in near-surface fraction of rBC tentatively ascribed to residential wood burning emissions.
- Free troposphere contains < 5% near-surface rBC-containing particles.

Preliminary Analysis

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)

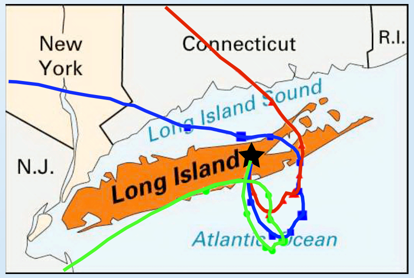
Acknowledgements

R. Subramanian (DMT) - Discussions regarding SP2
Shan Zhou (UCD) for assistance in AMS data analysis
Danielle Weech (UIUC) - HYSPLIT calculations

DOE-ARM for SP2

This research was performed under sponsorship of the
U.S. DOE under contracts DE-AC02-98CH10886 and DE-FG02-11ER65293

ALC-IOP



ARM CLIMATE RESEARCH FACILITY
Brookhaven Science Associates

CARES

STORMVEx

ASR Atmospheric System Research

BROOKHAVEN NATIONAL LABORATORY