Black Carbon Mixing-State in the Sacramento Urban-Biogenic Environment: CARES

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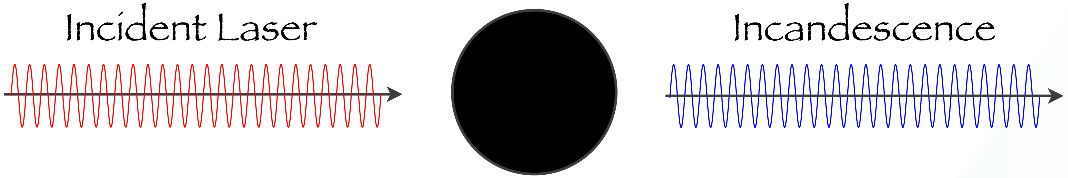
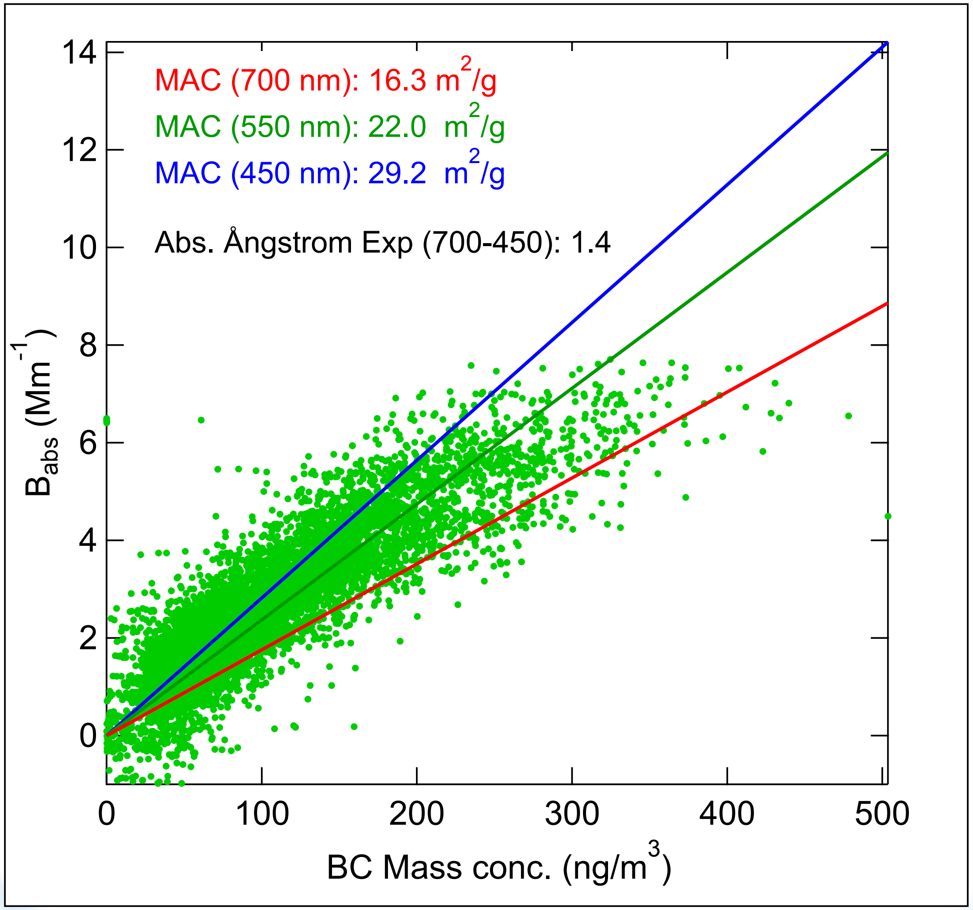
Measurement of Black Carbon: SP2

- SP2 (single particle soot photometer): particle-by-particle instruments
- High specificity towards soot (black carbon)



SP2 provides complimentary information to aerosol light absorption enabling calculation of mass light absorption cross-section (m^2/g).

June 28 -- Mass Absorption Cross-section



PSAP Corrections:

- Substrate
- Scattering

MAC (nascent soot) 7.5 m^2/g
(550 nm; Bond et al., 2006)

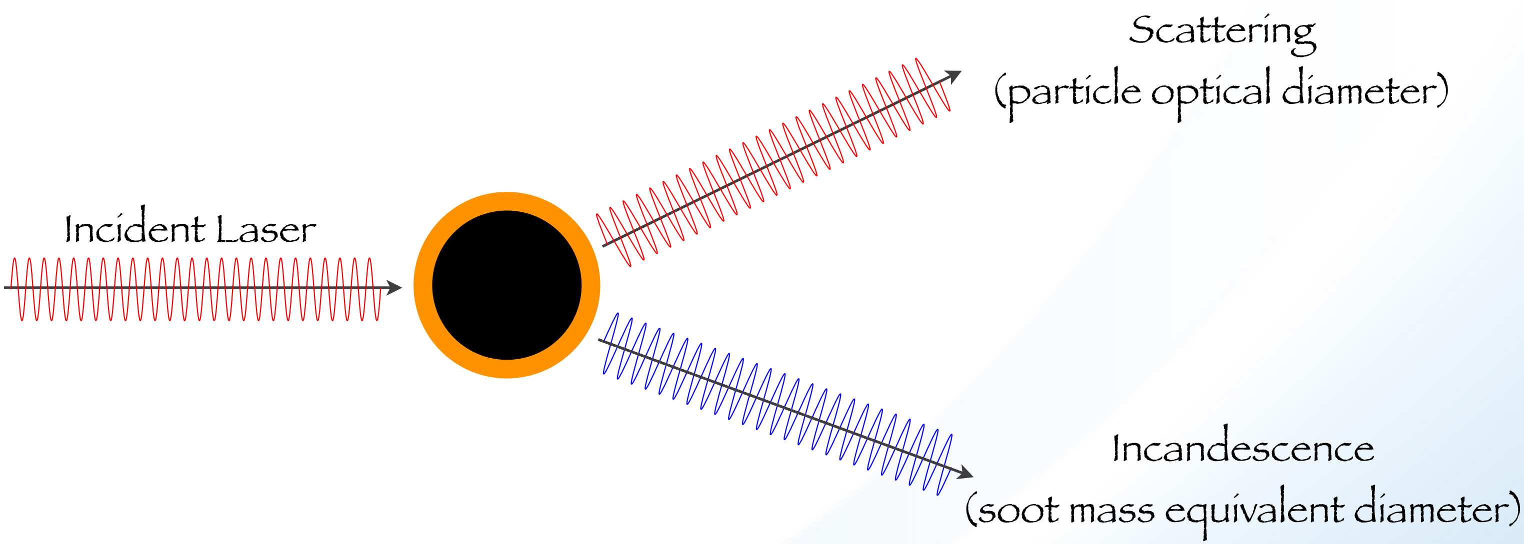
$22\ m^2g^{-1} / 7.5\ m^2g^{-1}$ =
Abs. Enhancement: 2.9

What does core-shell model predict?

Measurement of Black Carbon Mixing State

Approach to probing the mixing state of BC:

- Compare the temporal profiles of the scattering and incandescence signals
  - If both signals nominally peak at the same time, then coating ~ 0
  - If the incandescence signal 'lags' the scattering signal, then coating ≠ 0

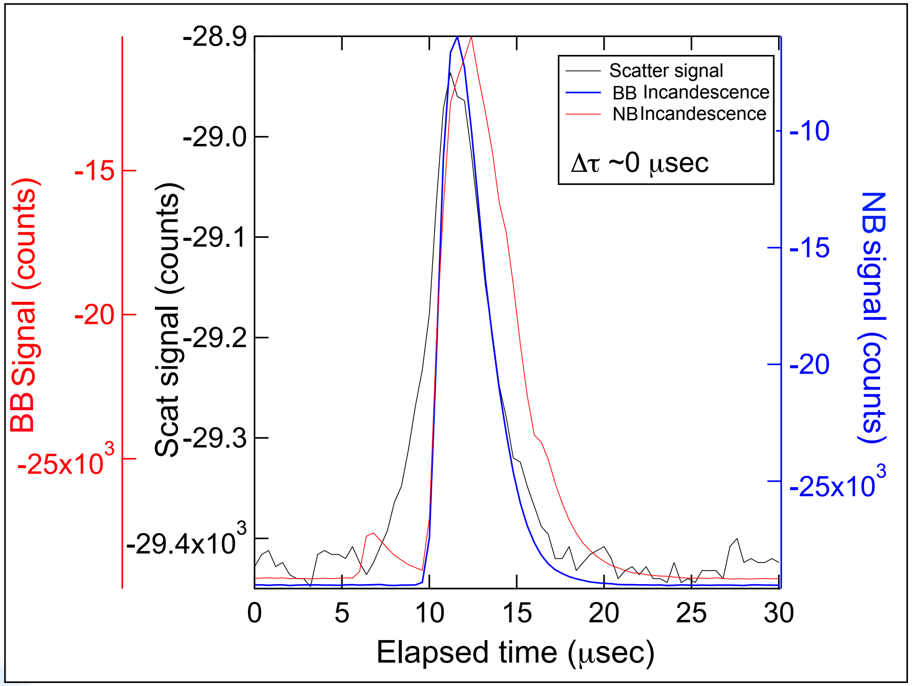


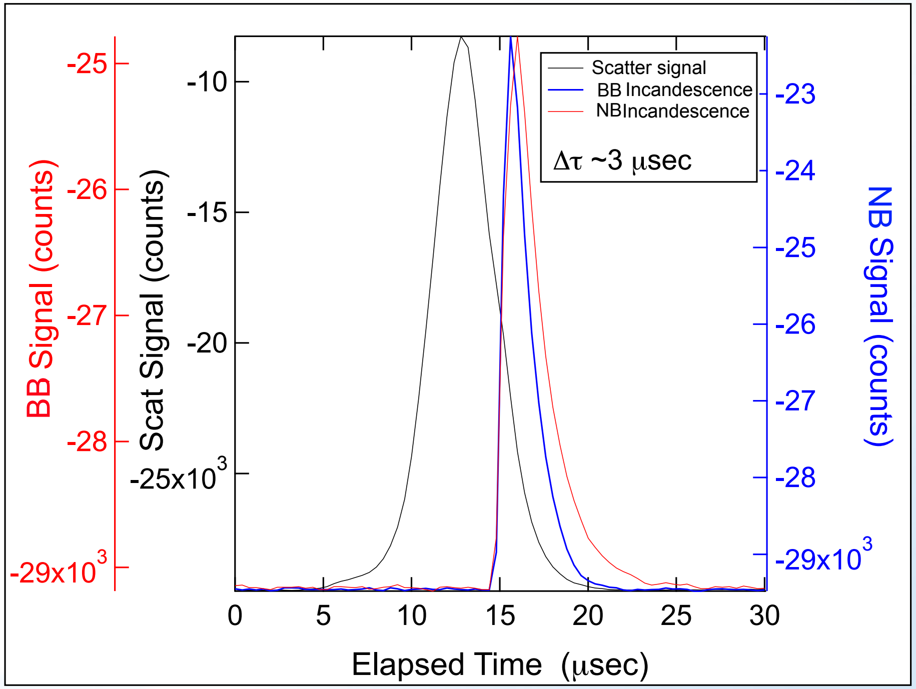
Aerosol Science: Black Carbon Mixing State

Probing BC mixing-State Using Lag-time

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

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

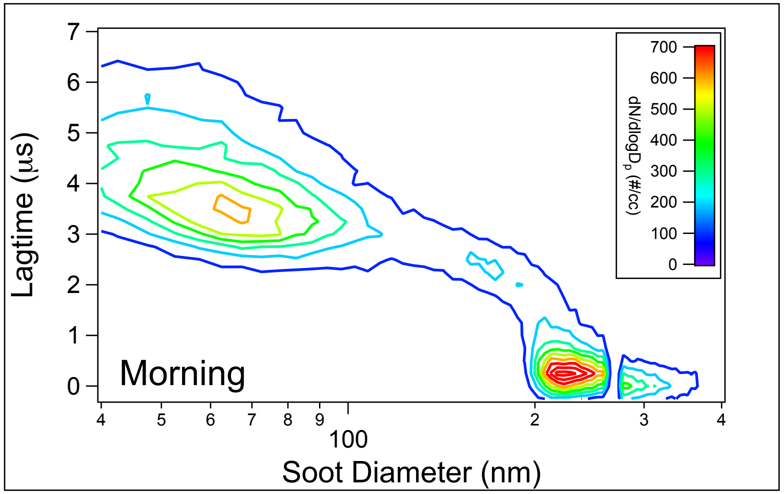


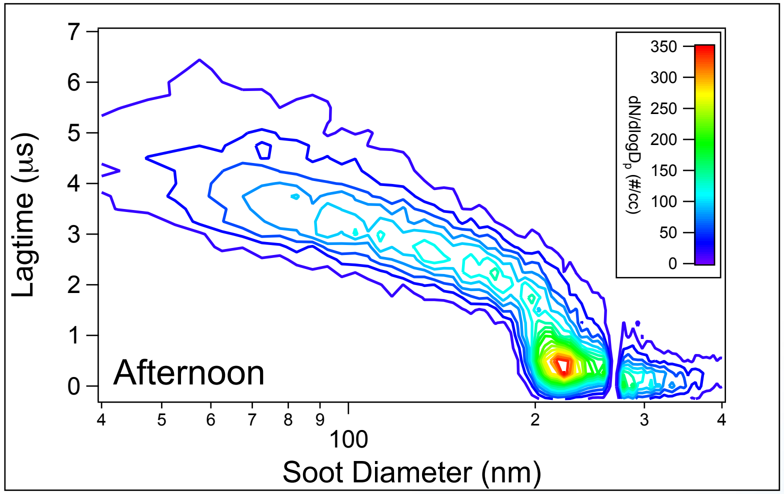


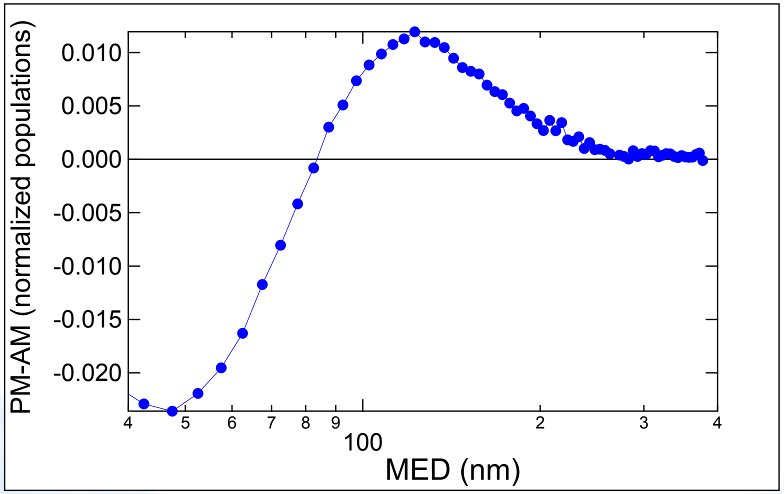
Thin coating ≡ $\Delta\tau \approx 0$

Thick coating ≡ $\Delta\tau \neq 0$

Daily Variation in Coating Thickness: June 28







Morning to Afternoon Observations:

Decrease in number of thickly coated particles

Increase in number of coated particles with larger cores

Less BC (mass) in afternoon

Bimodal distribution in the morning (aged and nascent soot)

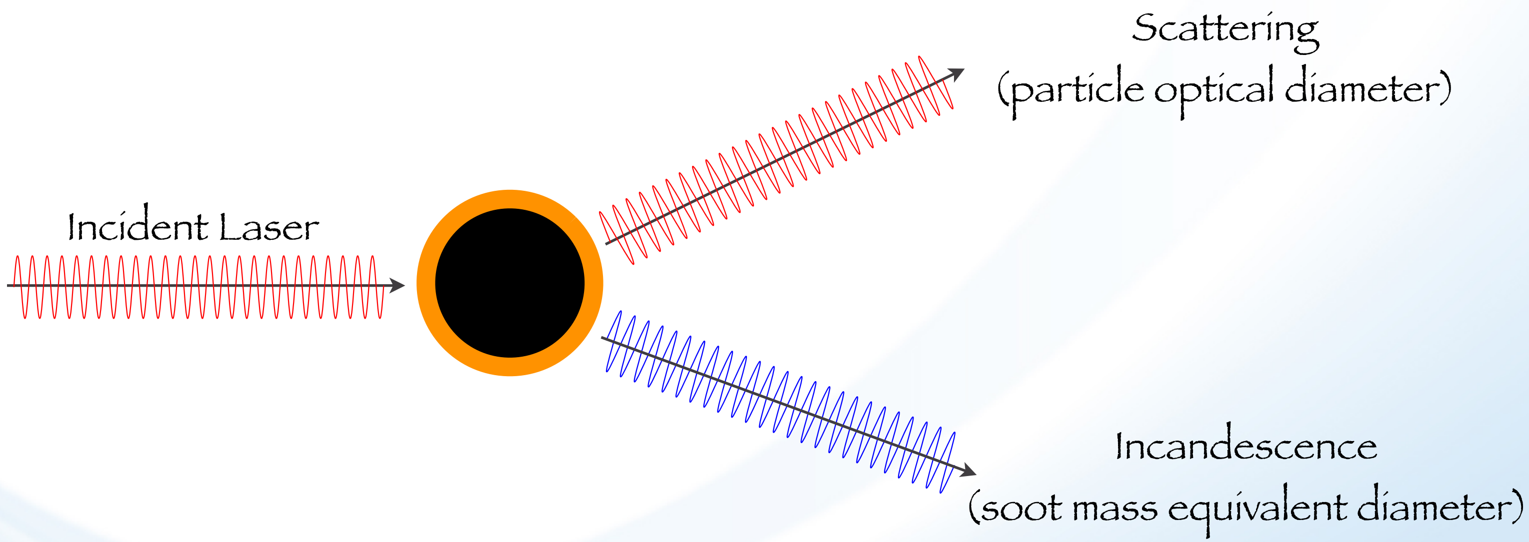
Growth of coating in the afternoon.

Measurement of Black Carbon Mixing State

More quantitative approach to probing the mixing state of BC:

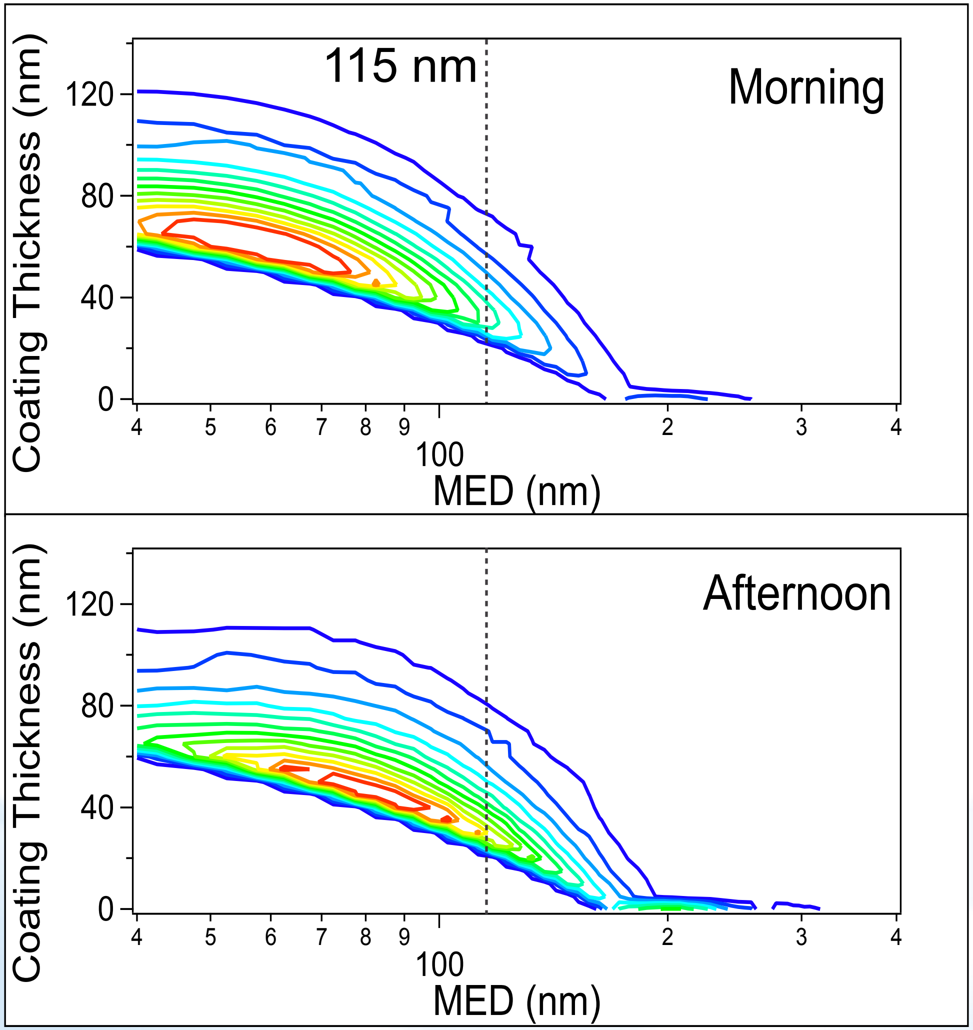
Compare the optical diameter (scattering channel) with the mass equivalent diameter (incandescence channel).

- If both diameter estimates are the same, coating thickness ~ 0
- If optical diameter > MED, difference equal to the coating thickness



100628b -- Core-Shell estimate of MAC

Estimation of coating thickness.



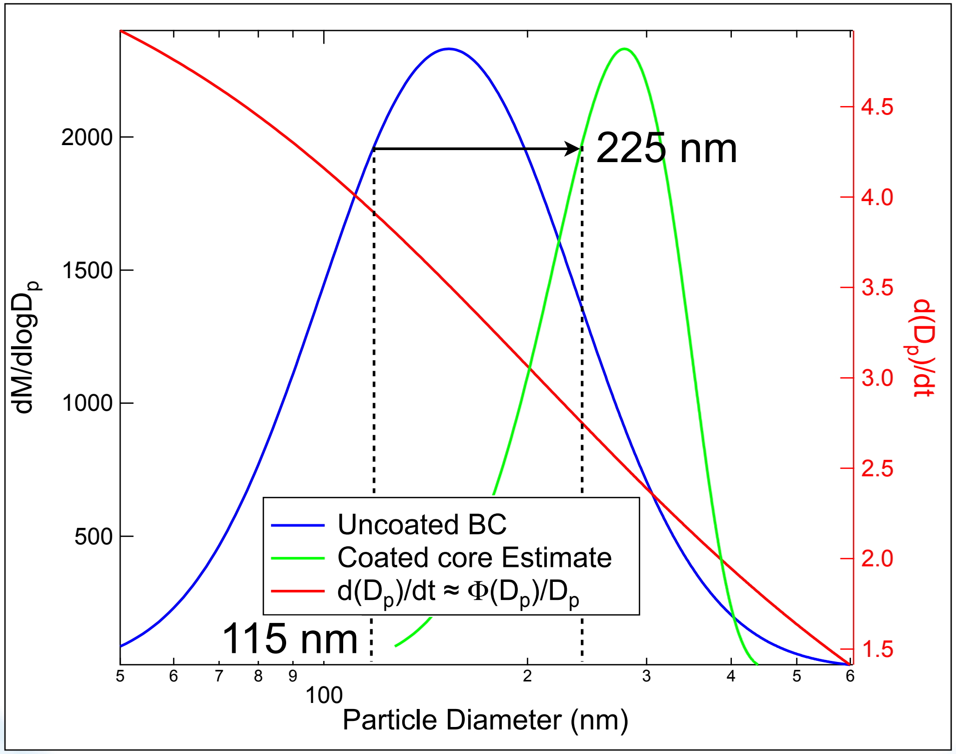
Optical detection window: 175 – 350 nm

Coating thickness (115 nm core): 55 nm

Coated 115 nm BC particles were negligible in the AM. During the day, these particles gained a coating to yield optical diameter D_p ~ 225 nm

Coating growth rate: ~10 nm/hr

Reconstructed $dM_{coated\text{-}core}/dlogD_p$ & Calc'd Absorption Enhancement



- Coating growth fixed by AM to PM SP2 observation
- Simple condensation model used to reconstruct coated-core distr. starting from uncoated BC observation

RI (core): 1.85 - 0.71i (Bond et al., 2007)

RI (coating): 1.43-0.0i

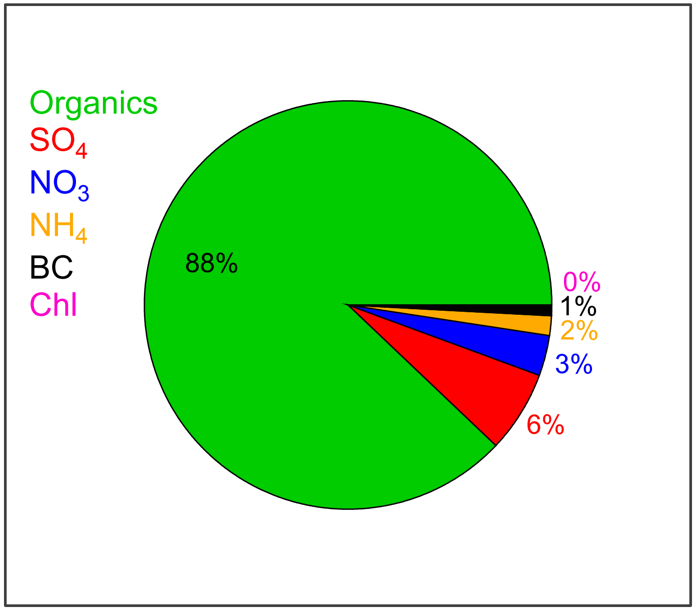
Absorption Enhancement

- Model prediction : 1.6
- Field measurement: 2.9

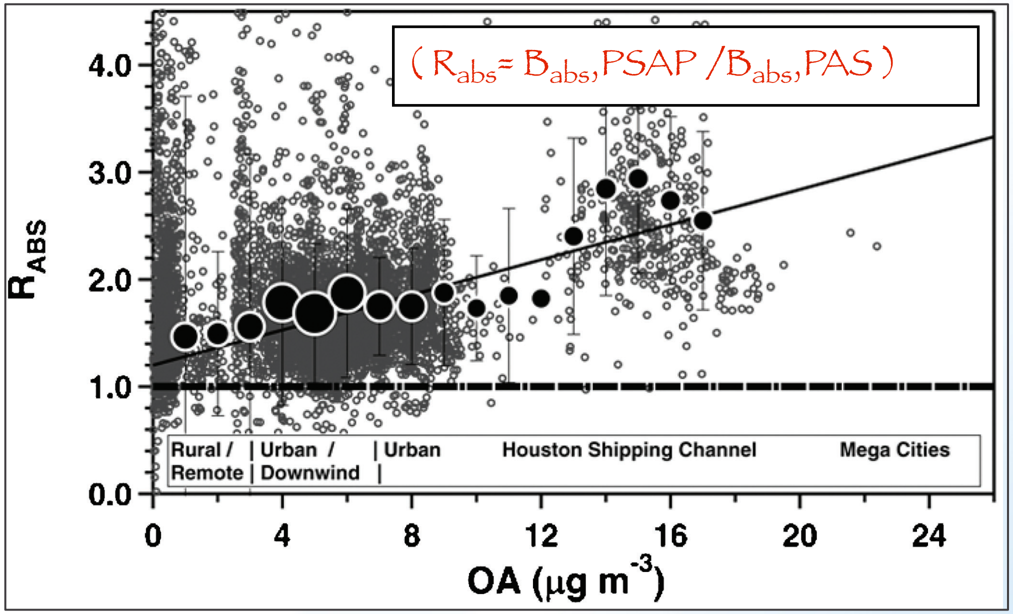
Nominally 2x difference between Model and Experiment.

Potential Explanation for Model-Experiment Discrepancy: OA bias in PSAP

June 28, 2010 - afternoon



Lack et al. (2008)



Average Org loading: ~12 µg/m³

OA loading of 12 µg/m³ (R_{abs} ~ 2)

Factor of 2 artifact estimated in PSAP measurement

Estimating of Coating Thickness: time-dependent scattering cross-sections

As the particle travels through the laser beam, light absorption results in particle heating which in turn causes loss of coating material (Δ in σ_{scat})

Issue: How do we estimate the unperturbed particle diameter?

Examine the lineshape of scattering signal (Moteki and Kondo 2008)

Assumptions:

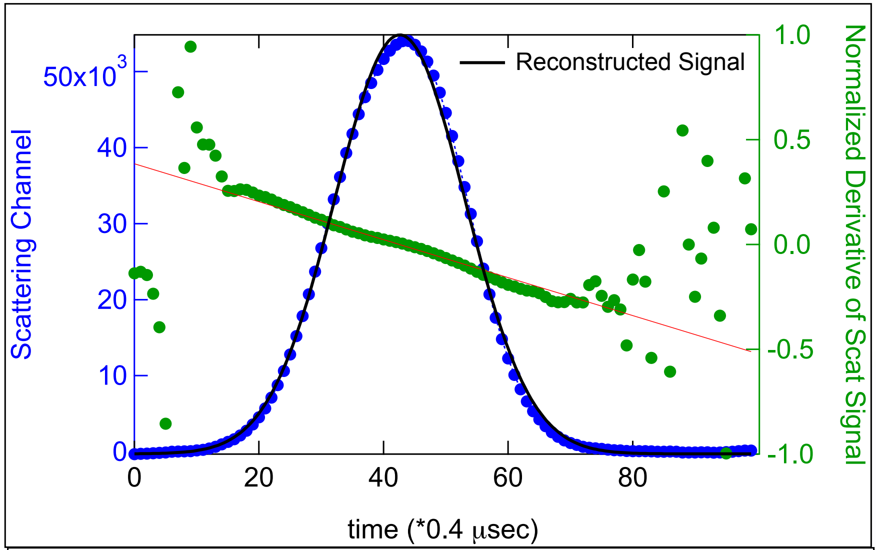
$$\frac{d\sigma_{scat}}{dt} = 0$$: Scattering lineshape will reflect Gaussian shaped laser beam

$$\frac{d\sigma_{scat}}{dt} \neq 0$$: Marked deviation from the Gaussian lineshape due to material loss

Time-Dependent Scattering Cross-Sections

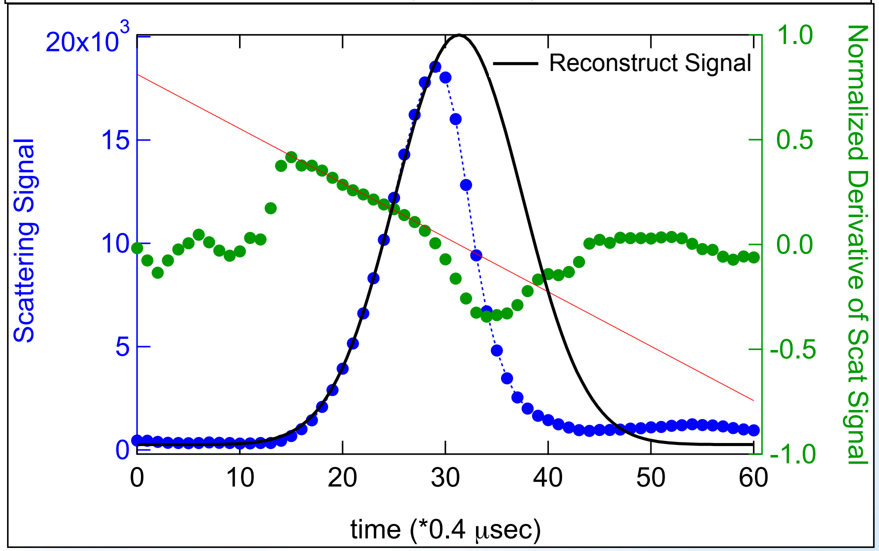
Examples

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



pure scatter

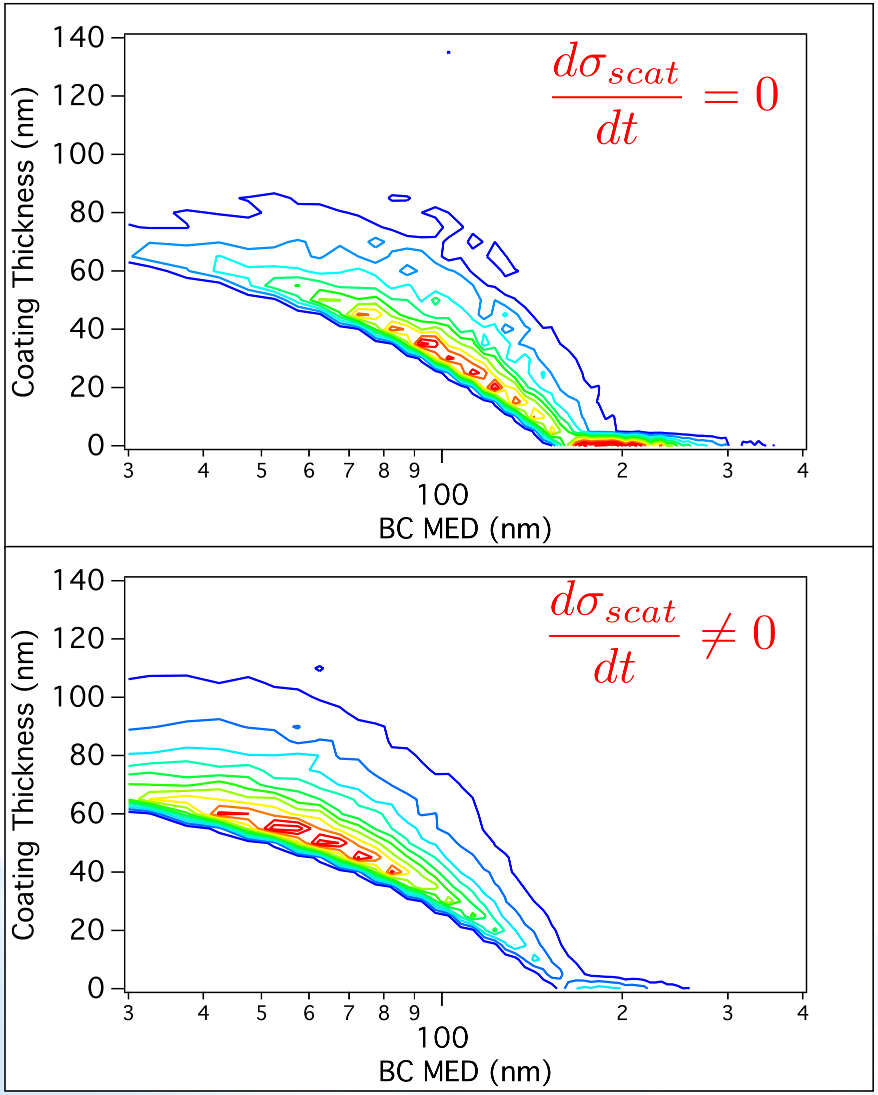
$$\frac{d\sigma_{scat}}{dt} \neq 0$$



coated BC

Time-Dependent Scattering Cross-Sections

Examples



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

Preliminary qualitative assessment indicates add'l increase in positive radiative forcing due to lensing effect (coating)

The normalized derivative method extends the dynamic range of the SP2 coating measurement.

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Summary

- June 28, 2010:

  Bimodal coated-core size distribution observed in AM not present in PM

  Relatively fast coating growth observed

  2x discrepancy observed between predicted light absorption enhancement and measurement -- OA bias in PSAP??

  Observation of BC without accompanying CO

- Normalized Derivative method for Estimating BC coating thickness

  Implementation very recently completed. Application on CARES datasets (Flight days 100628; 100615) will begin in the very near future

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