Probing rBC-containing Particle Morphology with a Single Particle Soot Photometer (SP2)

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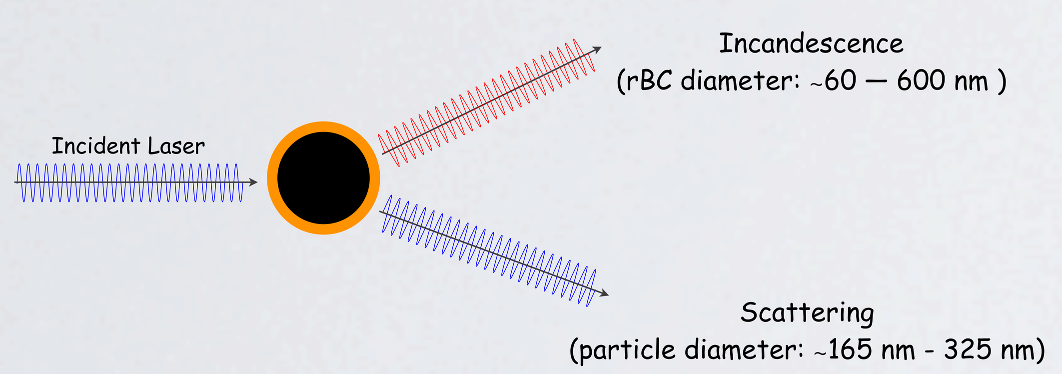
Abstract

A significant source of the uncertainty associated with refractory black carbon (rBC) radiative forcing is due to uncertainty in the rBC-containing particle chemical composition and morphology. While mass spectrometry is now routinely utilized to provide in situ, online analysis on the chemical composition of these particles, morphology is still limited to microscopy. One class of instrumentation that has recently shown promise towards addressing this gap is the Single Particle Soot photometer (SP2).

Using the SP2 lagtime method, Sedlacek et al., (2012) interpreted the existence of a particle scattering signal after rBC incandescence as evidence of rBC inclusions at or near the surface of their non-refractory hosts. In an effort to further evaluate this initial interpretation, elucidate the origins of these signal conditions, and to explore the utility of this methodology for studying rBC particle morphology, a series of laboratory-based experiments were carried out as part of the ASR-sponsored Boston College Black Carbon study (BC3).

Probing rBC Mixing State with SP2

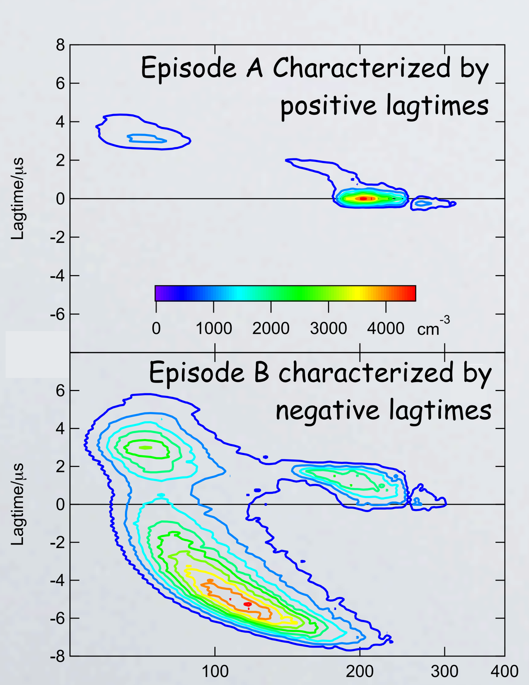
Schwartz et al., 2006; Moteki & Kondo, 2007, Subramanian et al., 2010

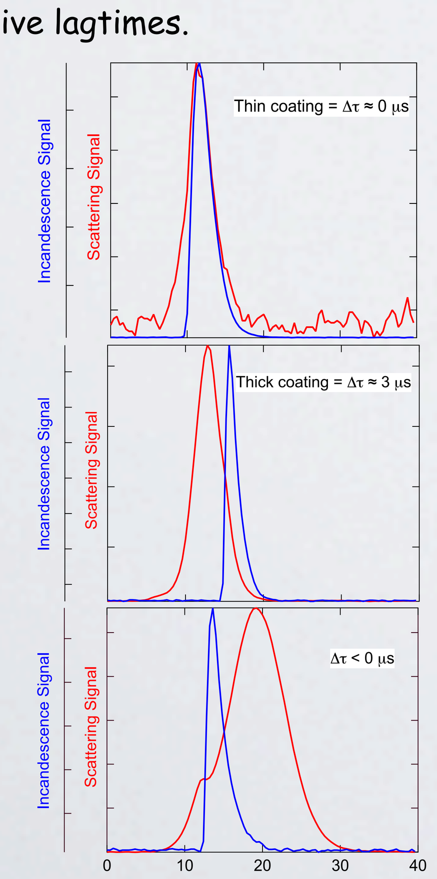


- Probe coating thickness: optical and BC mass equivalent diameters
- Examine temporal profiles of the scattering and incandescence signals

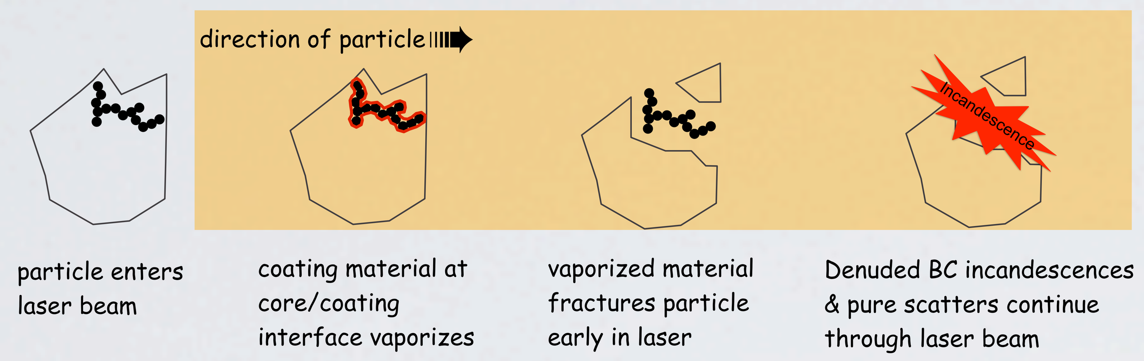
Motivation (Sedlacek et al., 2012 GRL)

During the DOE-sponsored Aerosol Lifecycle (ALC) field campaign episodes were encountered where large fractions of rBC-containing particles were characterized with negative lagtimes.

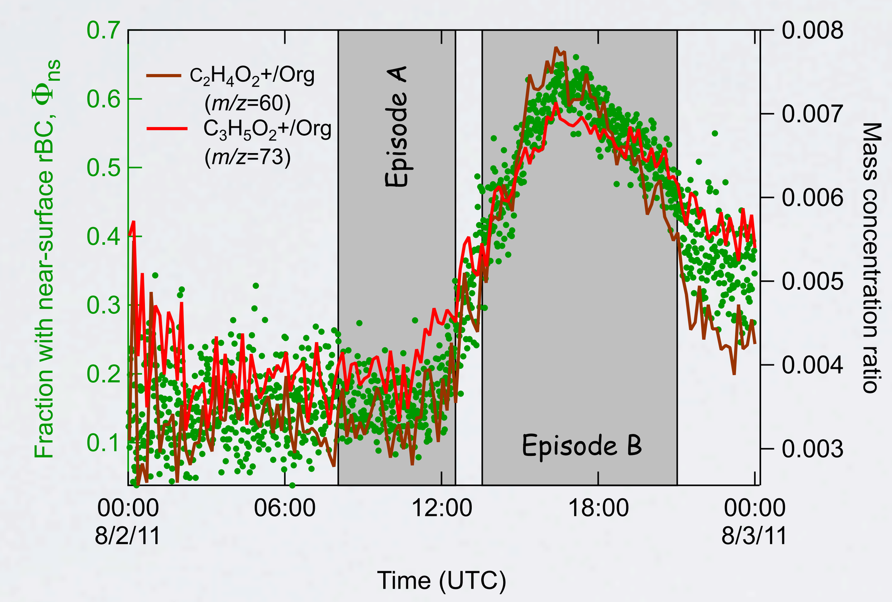




Asymmetric Amplitudes Interpreted as rBC Inclusions Near Surface



Near-Surface rBC-Containing Particles Correlated with Biomass Burn tracers



Conclusions from Sedlacek et al.:

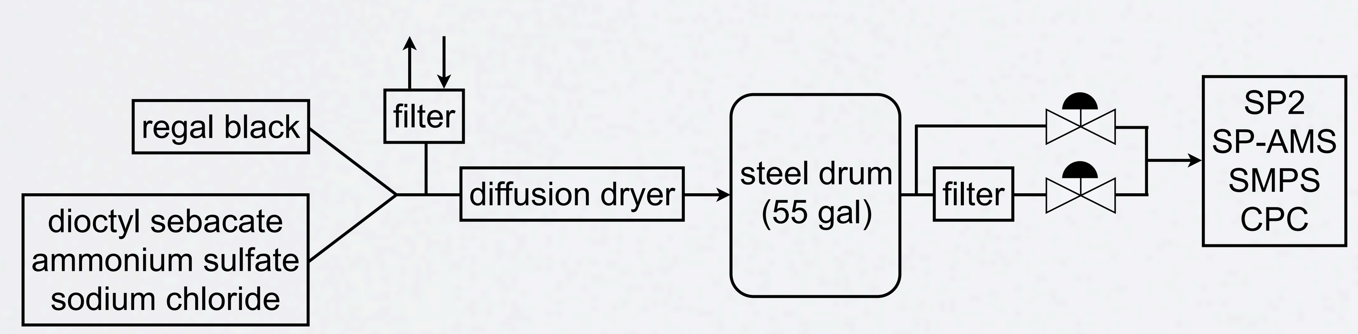
- Negative lagtimes interpreted as rBC inclusion near the surface of particle.
- Correlation of near surface BC with biomass burning.
  Observed in the present study, CARES, and StormVEx (Wallops wildfire).

Research questions:

- How common are near-surface rBC-containing particles?
- Are near surface rBC inclusions more likely from BB?
- Can this technique uniquely probe rBC-containing particle morphology?
- What is the radiative forcing impact of these particles?

Boston College Experiments - 2012

To answer some of the questions posed by the original work, a series of laboratory experiments were carried out as part of the ASR-sponsored Boston College Black Carbon Study-3 (BC3).



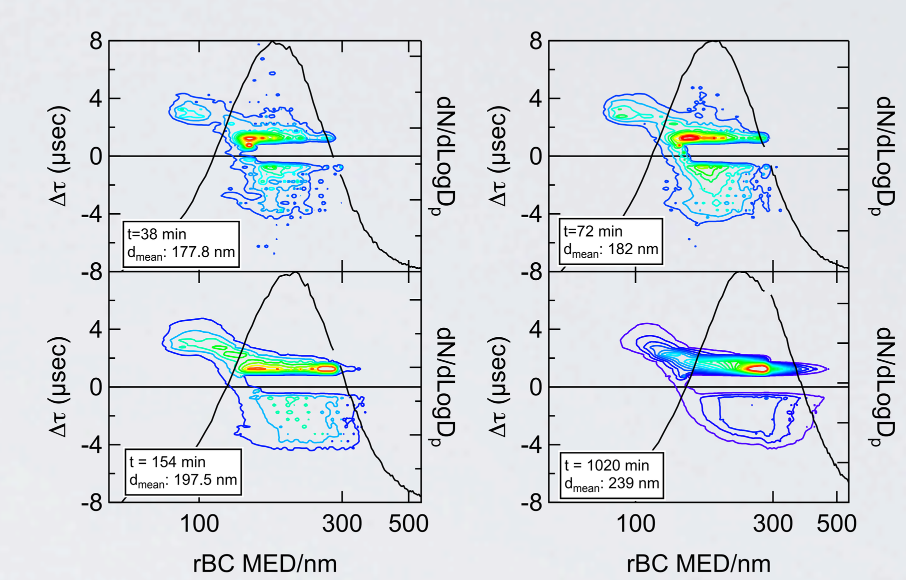
Particle formation through Coagulation and Condensation

For the BC3 experiments, four systems were examined:

- Coagulation of regal black (RB) with Dioctyl Sebacate (DOS)
  solid + liquid
- Coagulation of RB with Sodium Chloride (NaCl)
  solid + solid
- Coagulation of RB with dry Ammonium Sulfate (AS)
  solid-solid
- Condensation of RB with DOS.

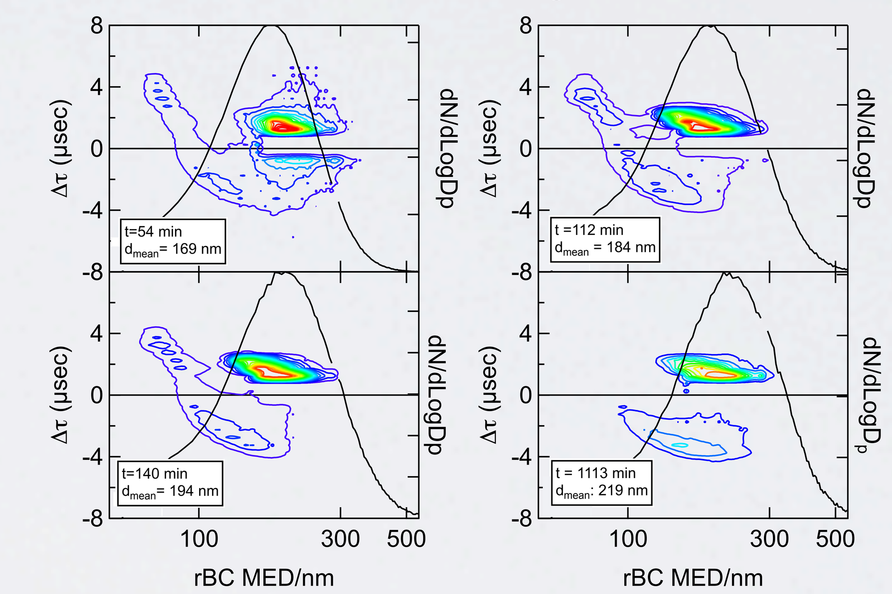
Particle Formation Through Coagulation

RB + NaCl coagulation



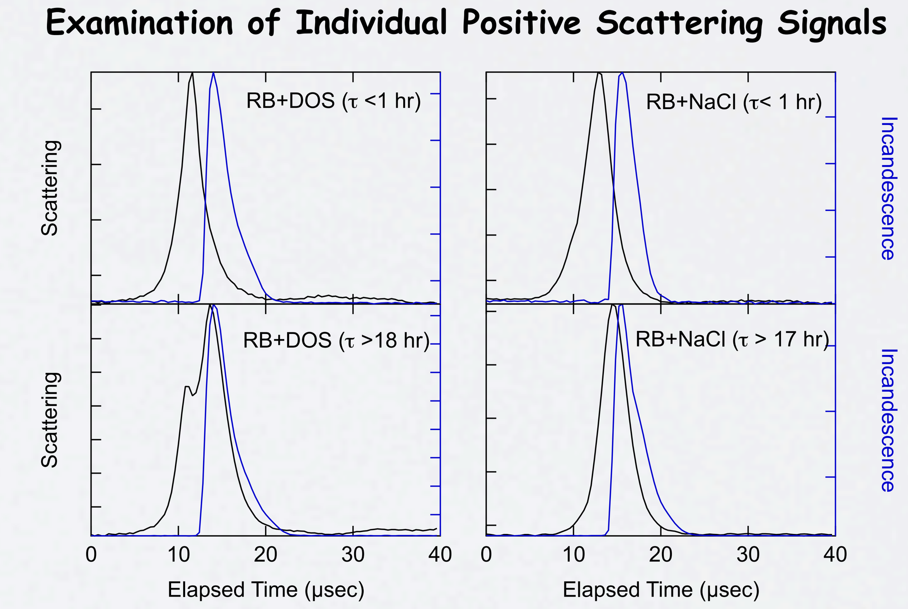
Lagtime distributions contain both positive and negative modes.

RB + DOS coagulation



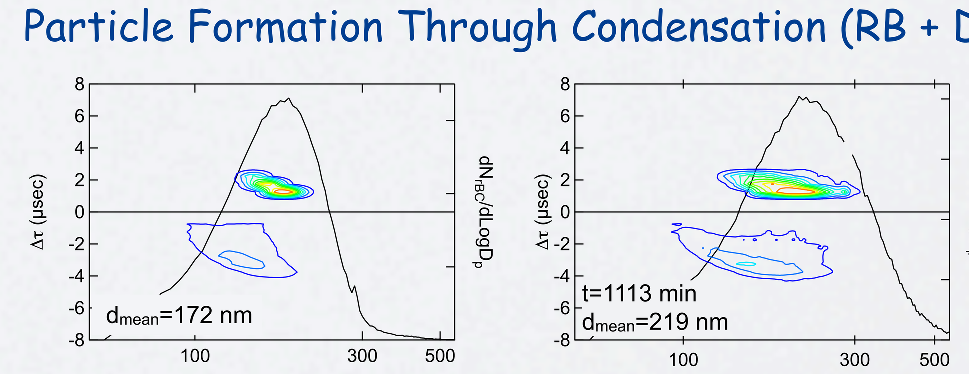
Negative mode observed to evolve. Note similarity in negative mode structure immediately following mixing with that observed for RB+NaCl system following overnight mixing.

Examination of Individual Positive Scattering Signals



Positive scattering signal structure is also found to evolve for RB+DOS system. This evolution hints at possible morphological changes with the RB+DOS system: RB diffusion in DOS?

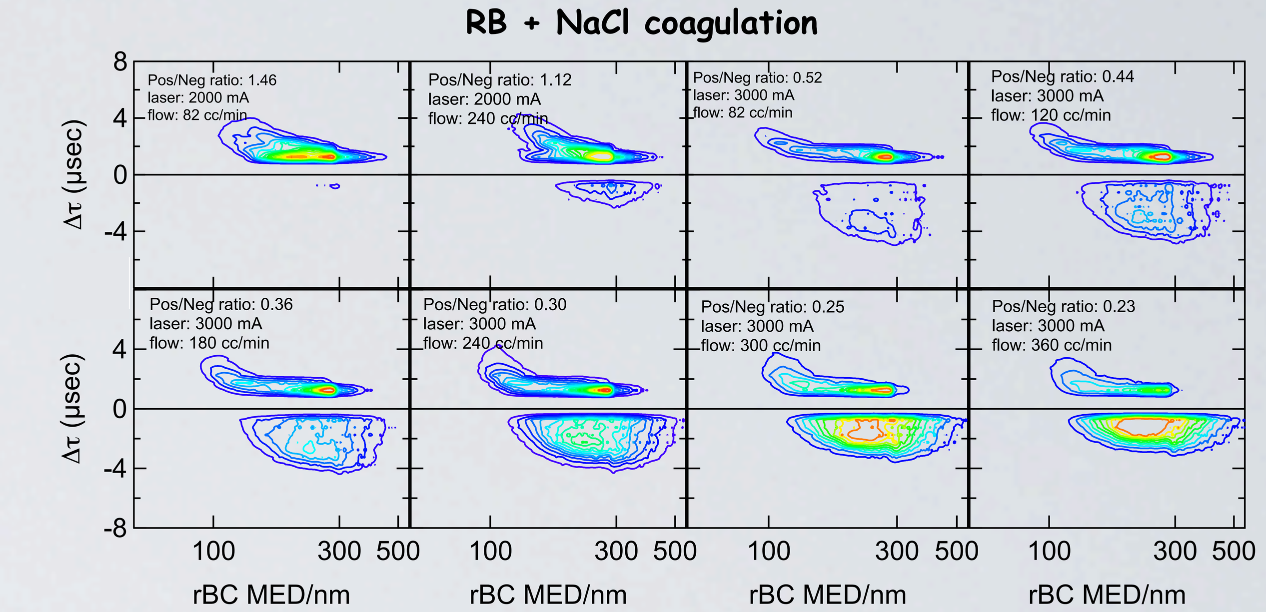
Particle Formation Through Condensation (RB + DOS)



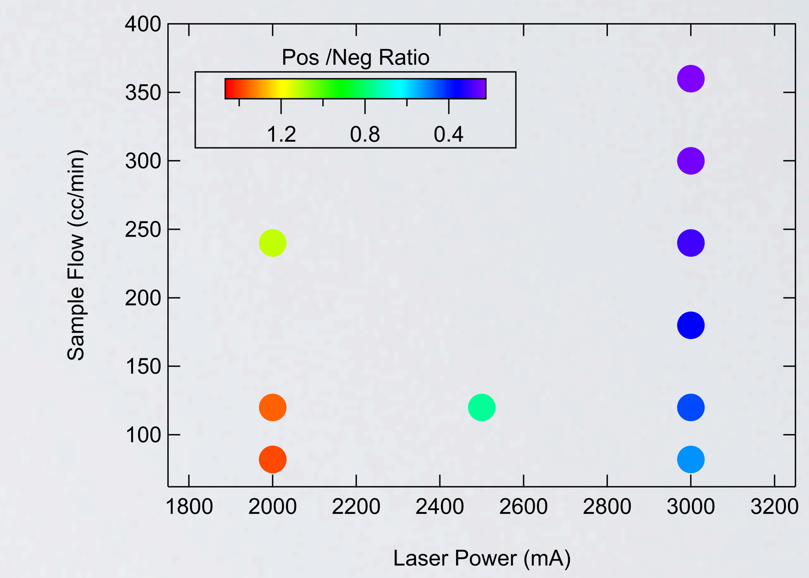
Striking similarity between lagtime distributions for RB+DOS coagulation and condensation suggests similar particle morphology. Signals also similar to that observed by Sedlacek et al., (2012).

Dependence of Scattering Signal on Acquisition Parameters

RB + NaCl coagulation



By varying the sample flow and laser power (diode injection current) positive lagtime scattering signals can be shifted to negative lagtime signals



Ratio of positive-to-negative lagtimes as a function of sample flow and laser power reveals systematic dependence on these parameters.

Implications of Present Study

- Positive lagtimes for coagulated particles could be misinterpreted as a core-shell particle resulting in an overestimate of light absorption.
- Dependence of lagtime distribution on laser power and sample flow warrant caution when interpreting rBC mixing state.

Use of Negative Lagtime Signals to Quantify rBC Mixing State

- Negative lagtime scattering signal unambiguously defines center of laser beam, greatly simplifying reconstruction of unperturbed particle diameter via normalized derivative method. (Moteki and Kondo, 2008)
- Analysis is currently underway on the refinement of the interpretation of negative lagtime signals

Conclusions

- Negative lagtime signals observed by Sedlacek et al. (2012) reproduced in laboratory.
- Lagtime distributions suggest differing particle morphology for RB+NaCl and RB+DOS systems.
- Striking similarity in negative lagtime signal structure for RB+DOS coagulation and condensation suggest similar particle morphology.
- Incandescence lagtime and scattering signal structure is strongly dependent upon laser power and sample flow rate.
- SP2 lagtime analysis can provide useful information on rBC-containing particle morphology. Complement chemical composition measurements.

Acknowledgements

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