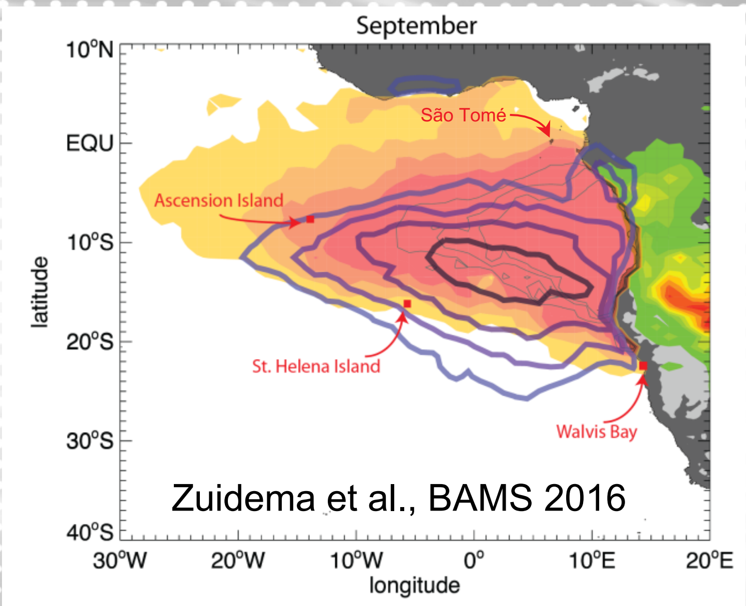
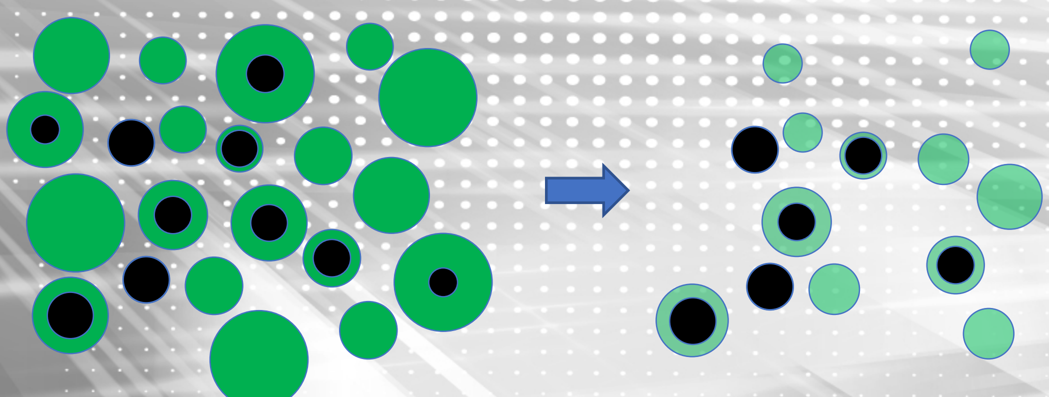
Cloud Processing of Biomass Burning Plumes Drives Black Carbon to Center Stage



A. J. Sedlacek III & E. R. Lewis
Brookhaven National Laboratory



BROOKHAVEN NATIONAL LABORATORY

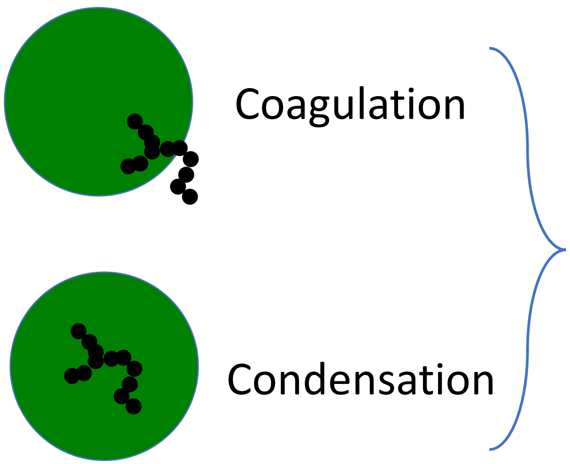
U.S. DEPARTMENT OF ENERGY

Probing rBC Mixing State

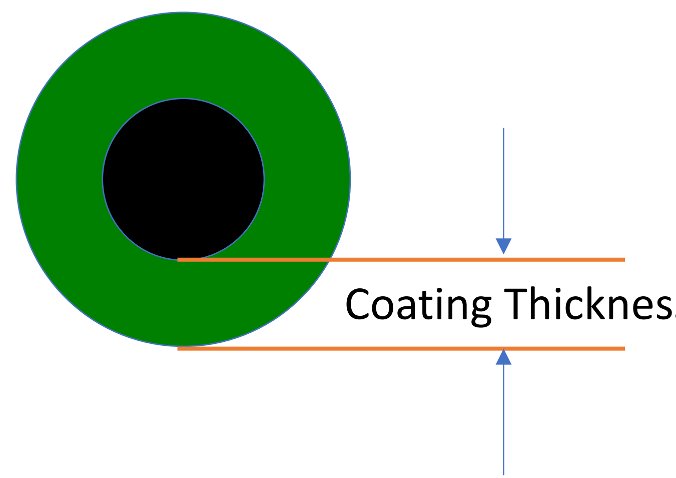
Wildfires offer a **unique** set of **conditions** that favor a variety of rBC-containing particle morphologies.

- rBC – POA coagulation
- condensation of organic material on rBC

Today's discussion: Assume an idealized "core-shell" morphology



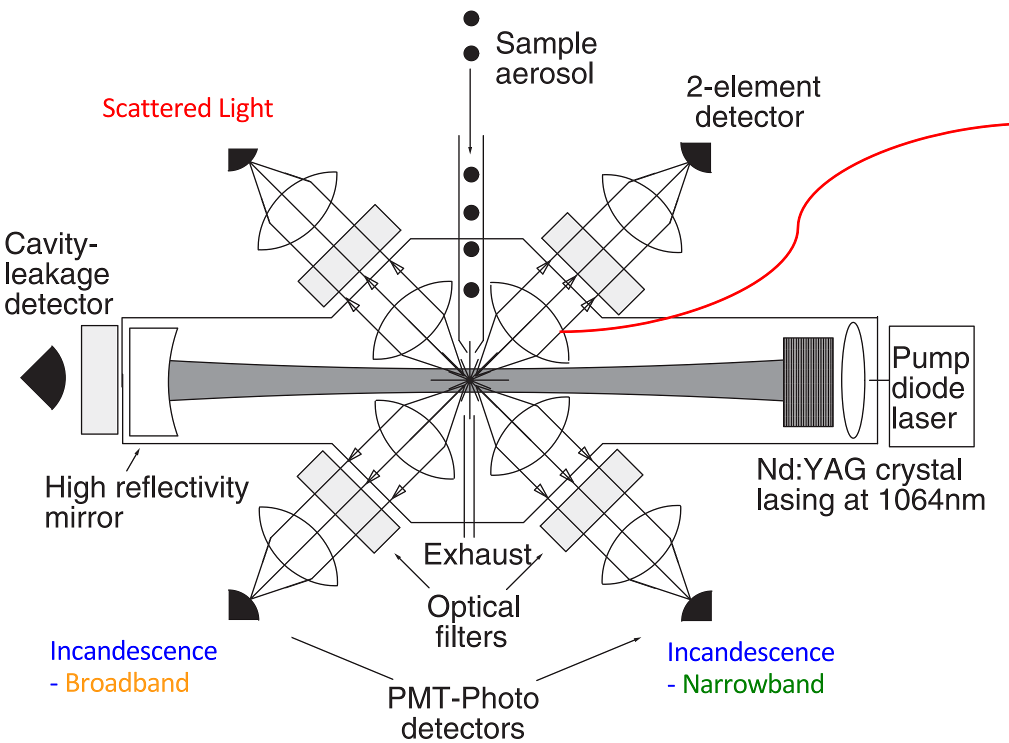
Evidence for both Particle types in the near Source

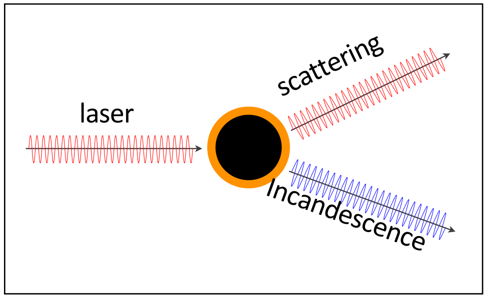


Next we probe the *potential* microphysical and optical implications of the derived rBC mixing state.

Probing Refractory Black Carbon (rBC) Mixing State

Detection of refractory black carbon (rBC) using laser-induced incandescence

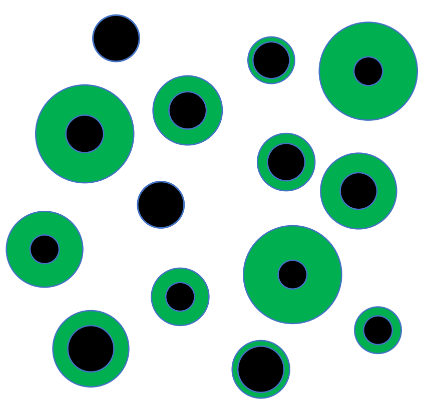




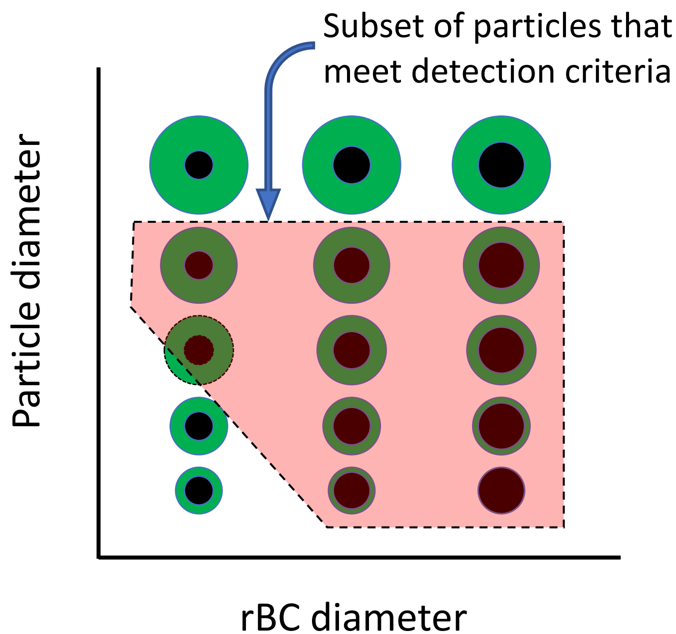
Schematic from
Schwarz et al., 2008

Refractory Black Carbon (rBC) Mixing State

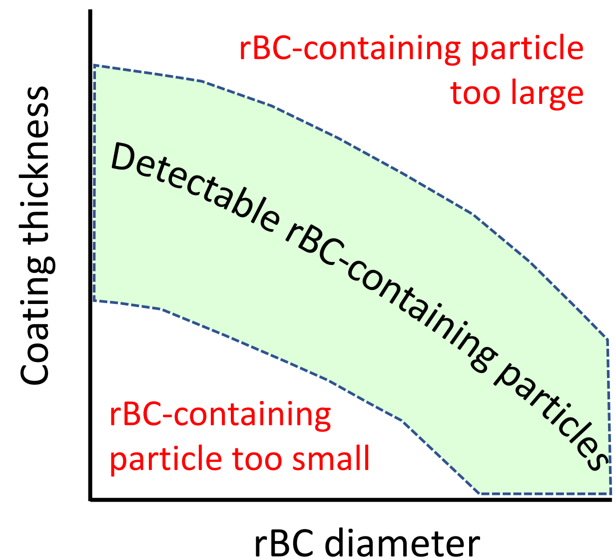
Use scattering and incandescence signals from individual rBC containing particles to probe rBC mixing state (i.e., coating thickness distribution as a function of rBC core diameter)



Distribution of rBC-containing particles



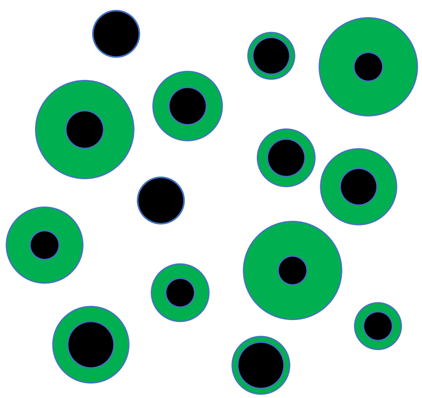
Classification of “total” particle diameter and core diameter using valid scattering & incandescence signals



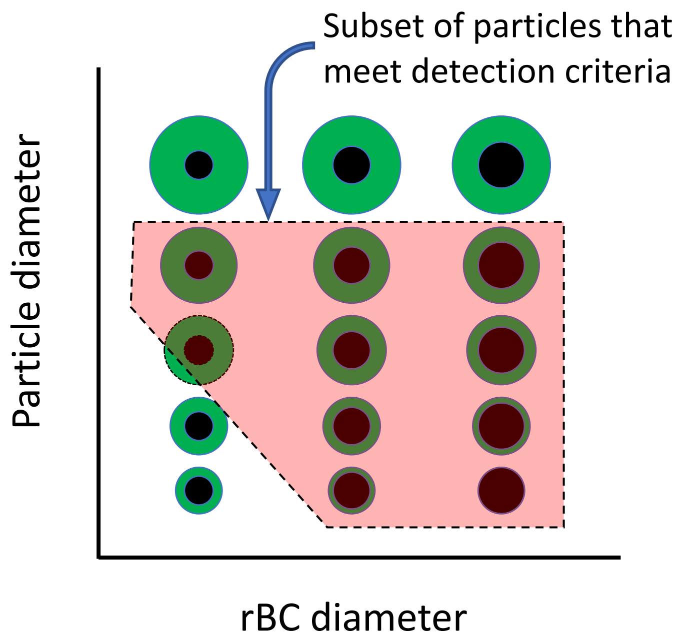
Plot a distribution of coating thicknesses for a given rBC core diameter

Refractory Black Carbon (rBC) Mixing State

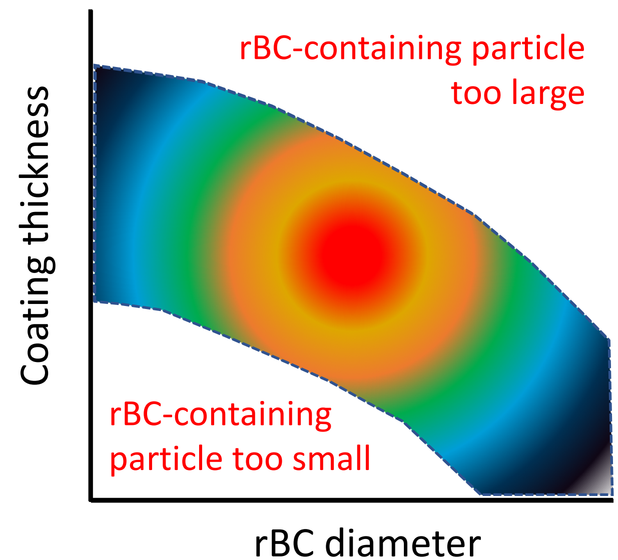
Use scattering and incandescence signals from individual rBC containing particles to probe rBC mixing state (i.e., coating thickness distribution as a function of rBC core diameter)



Distribution of rBC-containing particles



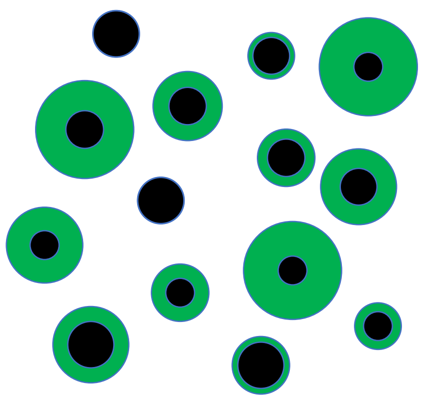
Classification of “total” particle diameter and core diameter using valid scattering & incandescence signals



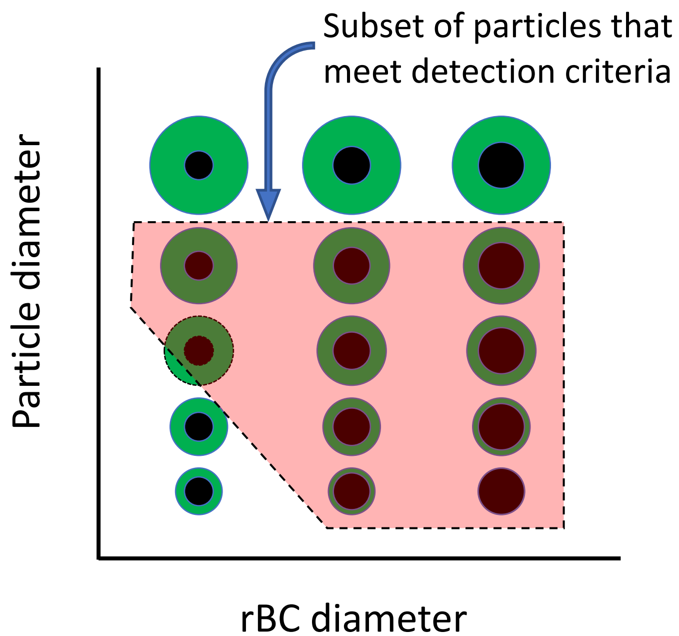
Plot a distribution of coating thicknesses for a given rBC core diameter

Refractory Black Carbon (rBC) Mixing State

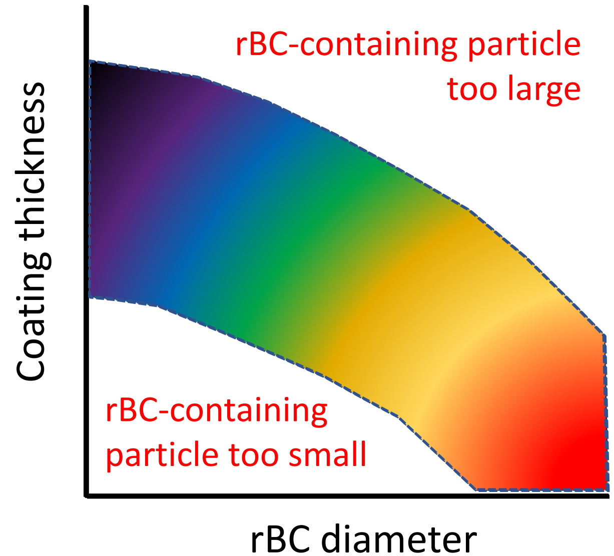
Use scattering and incandescence signals from individual rBC containing particles to probe rBC mixing state (i.e., coating thickness distribution as a function of rBC core diameter)



Distribution of rBC-containing particles



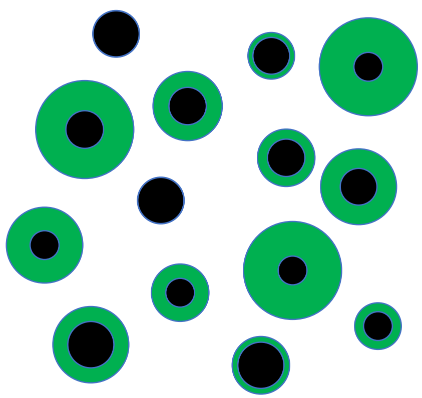
Classification of “total” particle diameter and core diameter using valid scattering & incandescence signals



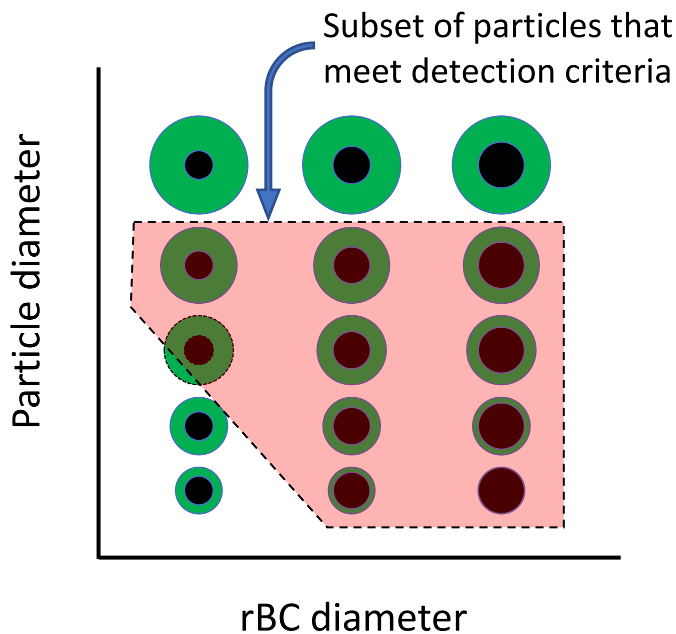
Plot a distribution of coating thicknesses for a given rBC core diameter

Refractory Black Carbon (rBC) Mixing State

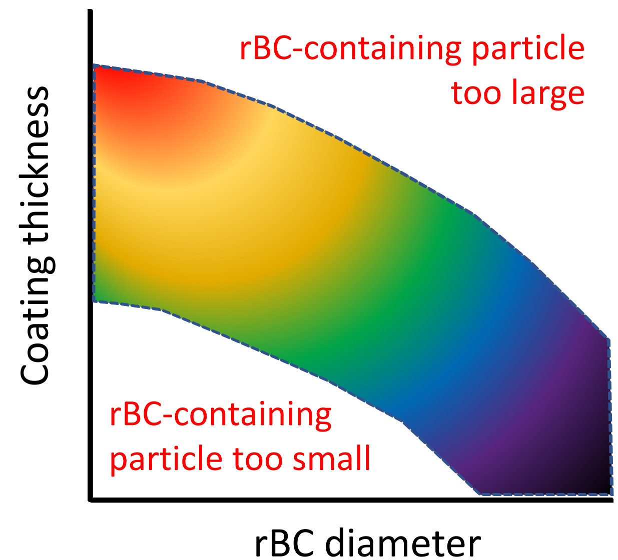
Use scattering and incandescence signals from individual rBC containing particles to probe rBC mixing state (i.e., coating thickness distribution as a function of rBC core diameter)



Distribution of rBC-containing particles

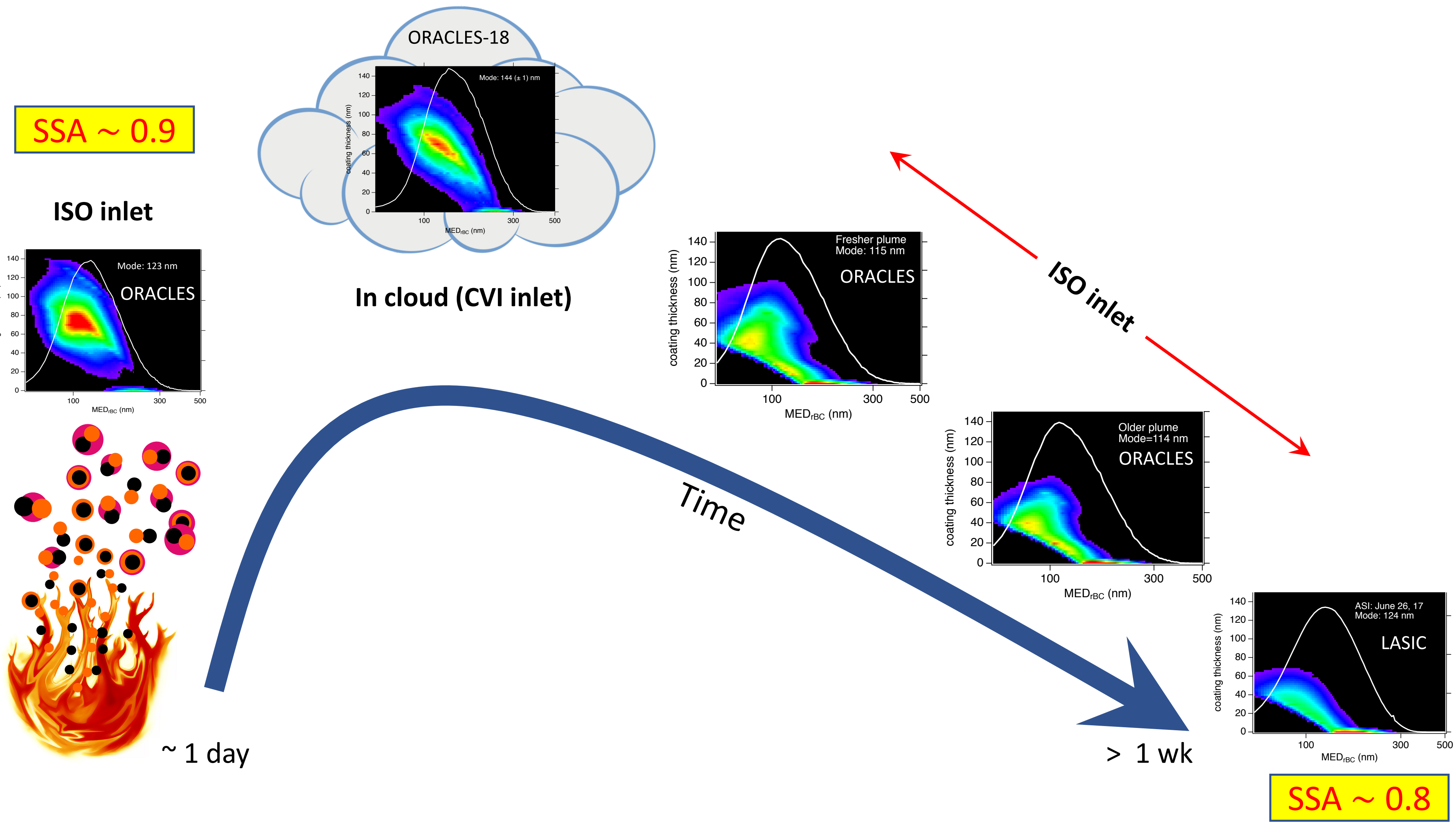


Classification of “total” particle diameter and core diameter using valid scattering & incandescence signals



Plot a distribution of coating thicknesses for a given rBC core diameter

rBC Particle Mixing States from LASIC & ORACLES

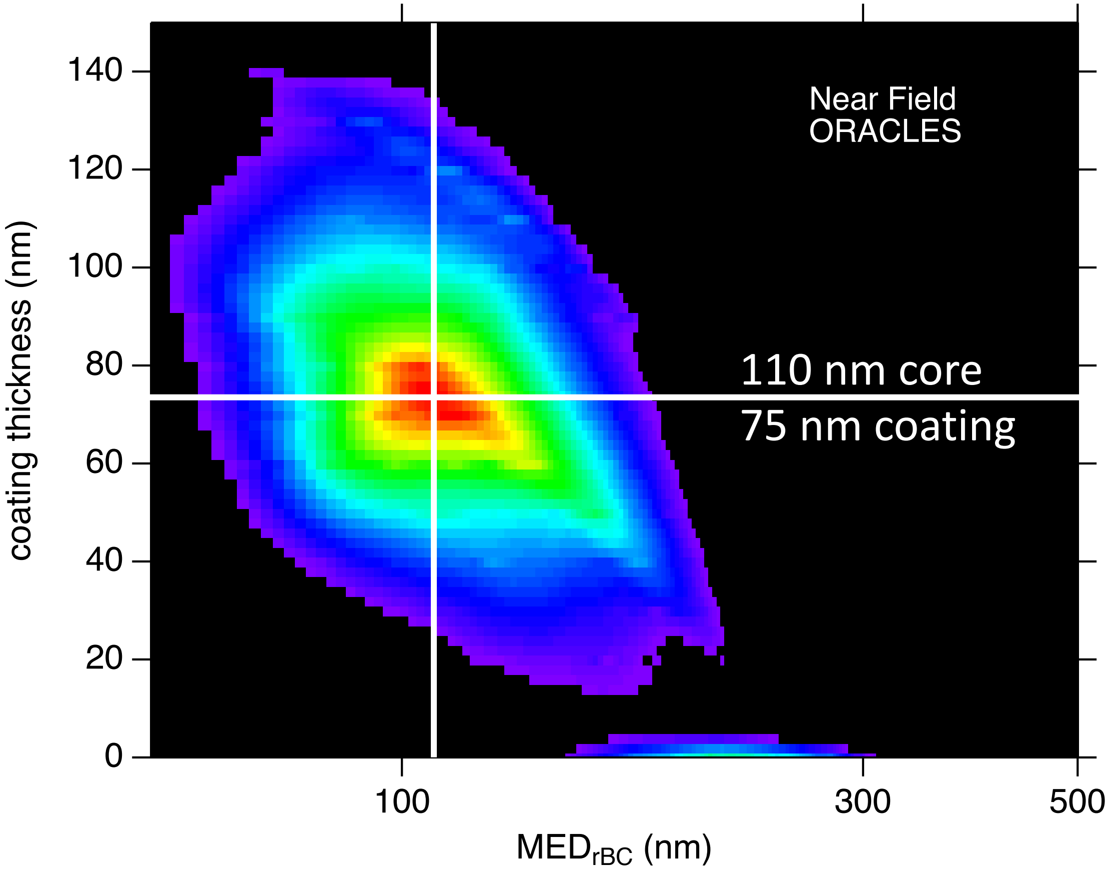


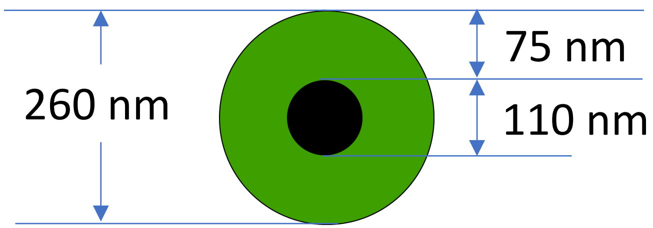
What drives the observed changes in mixing state and the SSA behavior?

rBC Particles are Thickly-Coated Nearest the African Coast

1) BC-coated particle is in core-shell configuration.

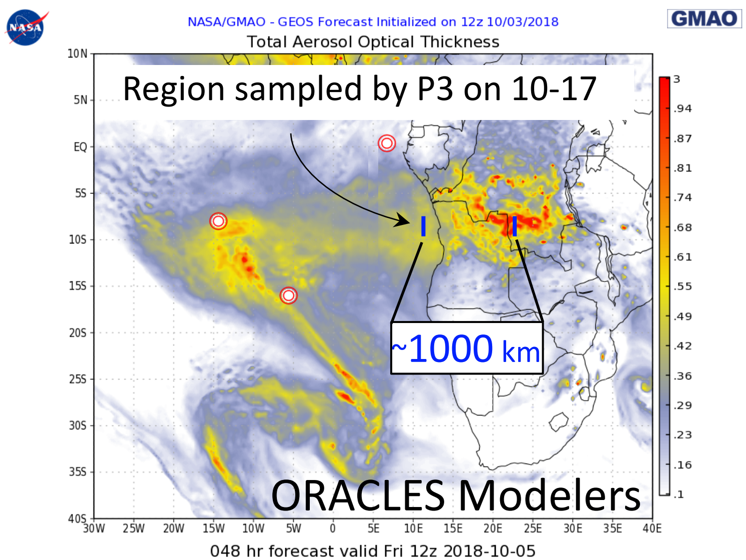
Assumption supported by large coating thicknesses.

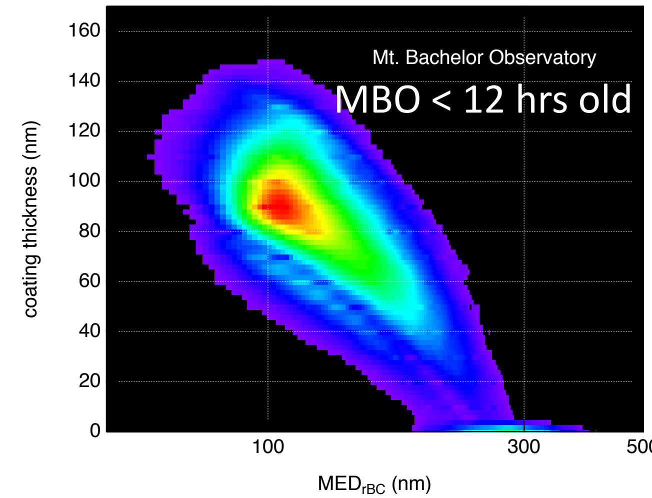


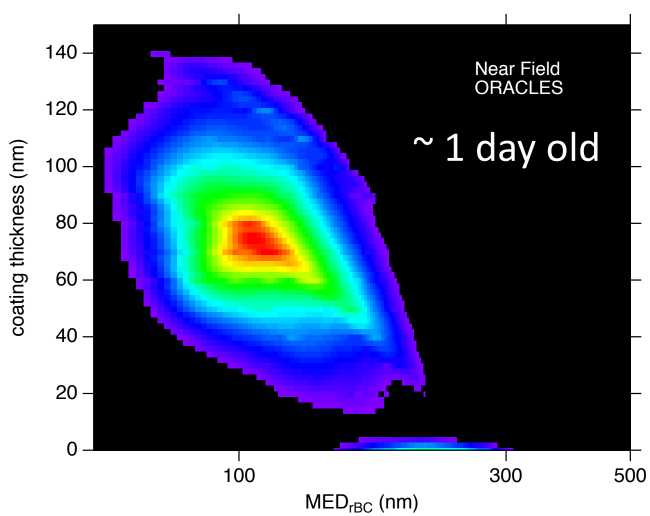


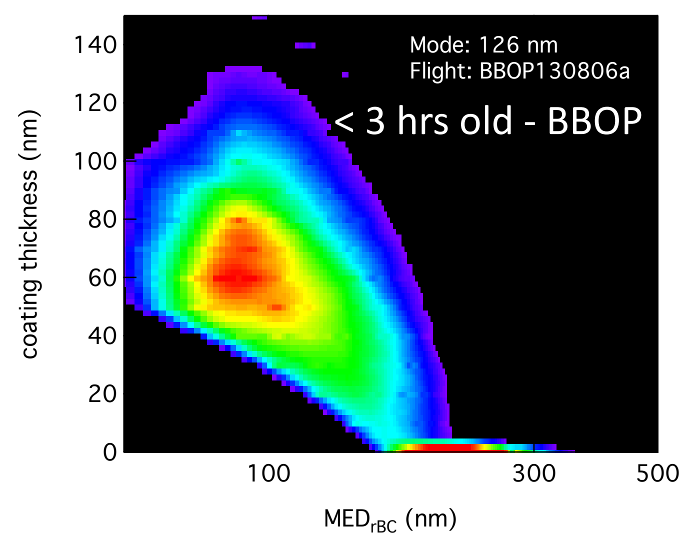
$$\frac{\text{Coating mass}}{\text{rBC mass}} = 13$$

Thickly-coated rBC particles in Near Source





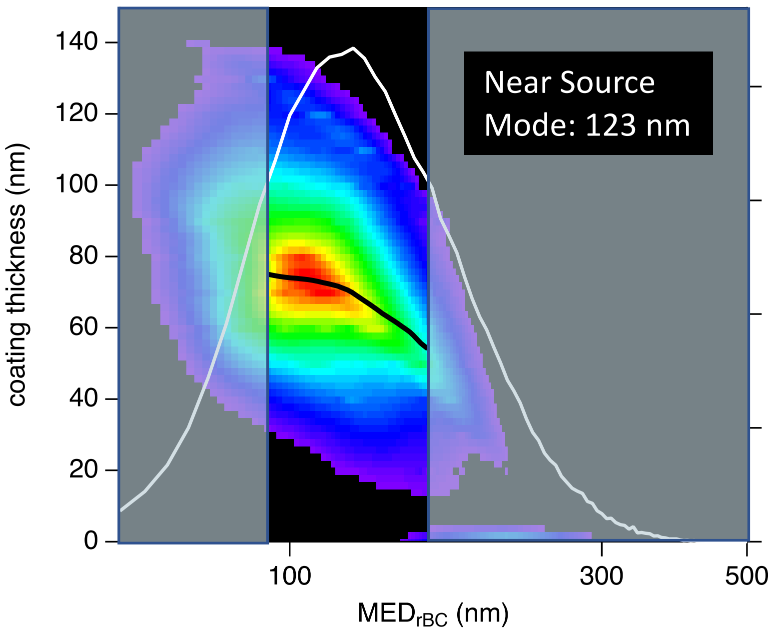


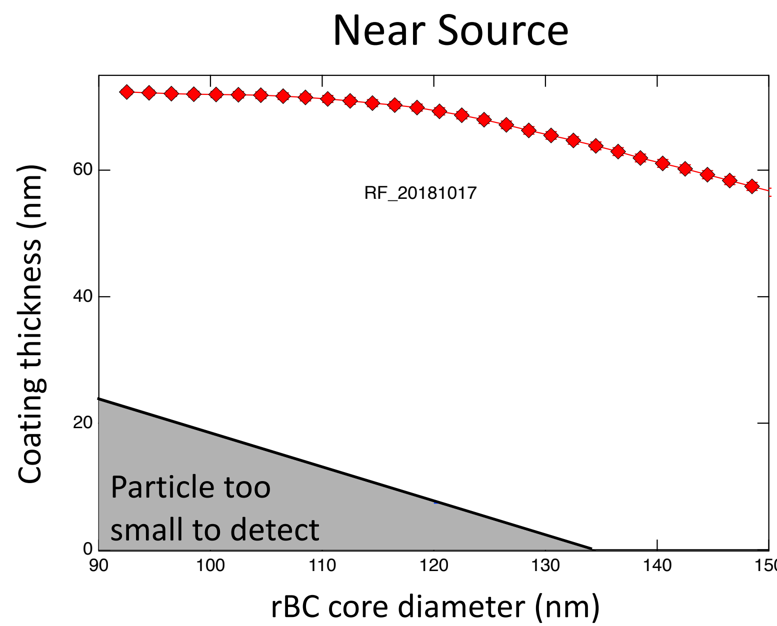


Near source rBC mixing suggests negligible coating loss rBC particles from origin to P3 sampling point.

Quantifying rBC Mixing State

To better quantify the rBC-containing particle mixing state, only a subset of coating thicknesses are used so as to avoid detection limit issues discussed earlier.



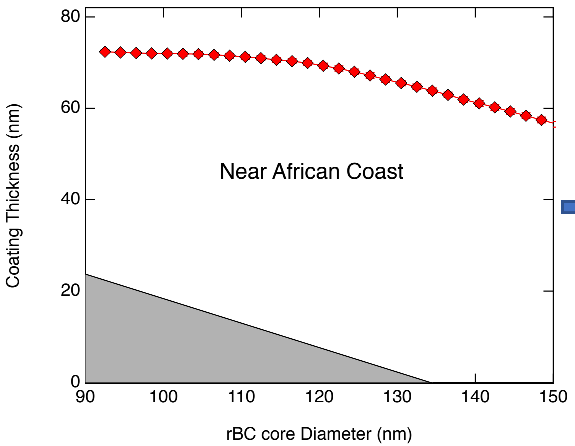


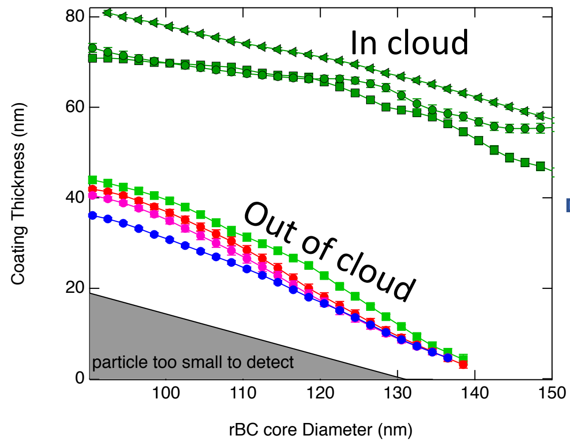
For SP2 colleagues, the LEO methodology was employed in this mixing state analysis

Variability in Derived rBC Coating Thicknesses

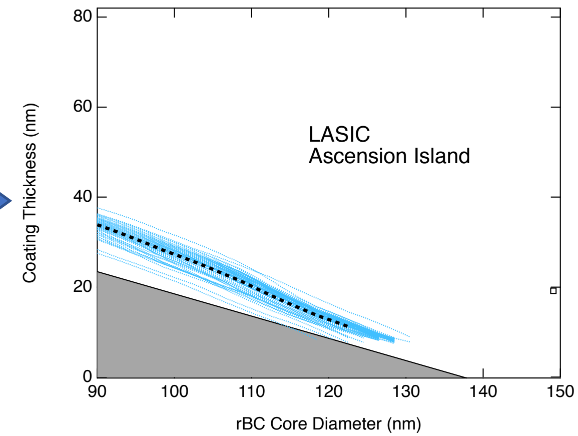
- Except for near source transect, all **ISO** inlet gives coating thicknesses < 40 nm for rBC dia=100 nm
- **CVI** inlet data spans the range from most thickly-coated particles to that observed in **ISO** inlet
- rBC particles analyzed from LASIC (Ascension island) exhibit the thinnest coatings

Near Source





Far Source



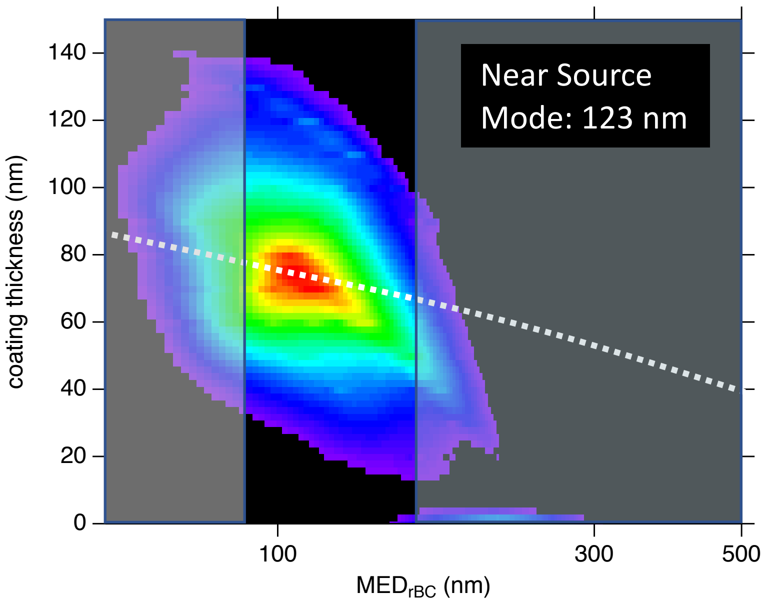
Cloud processing leaves behind only residual particles (i.e., smaller total diameter)

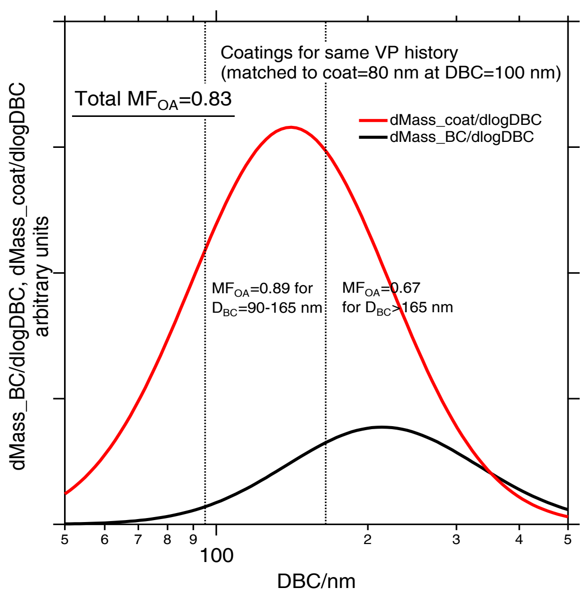
Reconstructing Coated Core Mass Distribution

Coated particles experienced the same vapor-pressure history;
thus, a given (coat, D_{BC}) determines coating for any D_{BC}.

Assumption supported by:

1) constant VP history line near max contours
2) steepness of contour lines
3) agreement of calculating mass fraction (MF_{OA}) in region where SP2 determines both coat and D_{BC}: 0.93 by integration, 0.89 by constant VP line

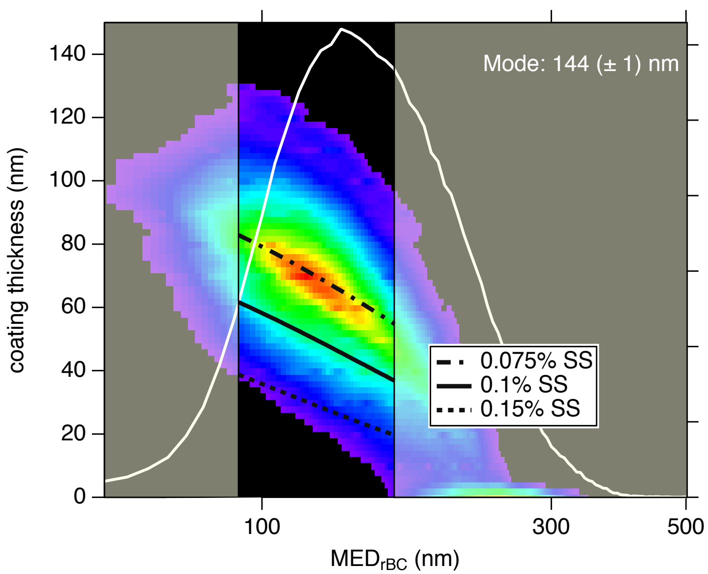




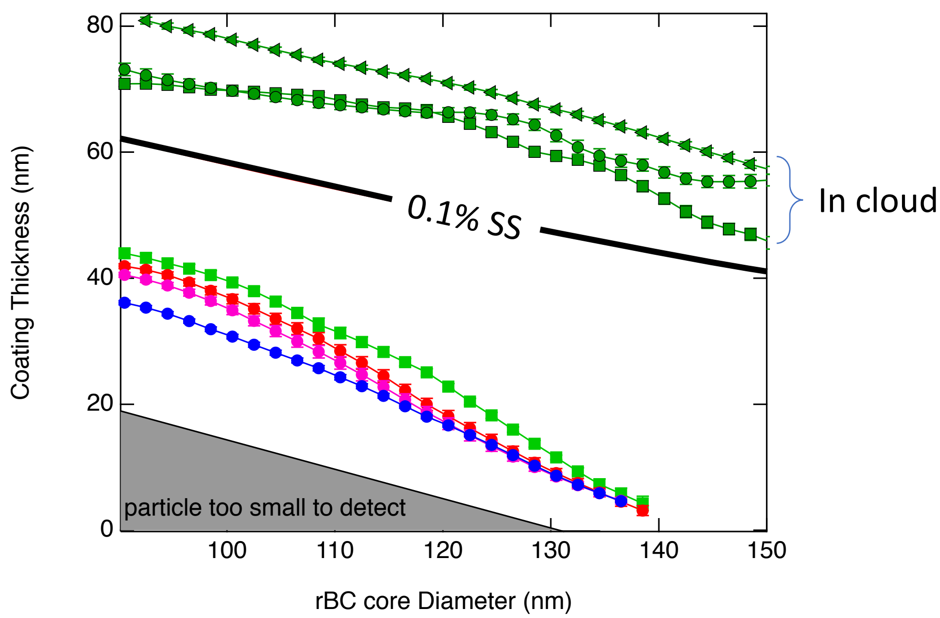
Activation of Coated rBC Cores

Activation: Kohler for core-shell (depends mainly on D_{tot})

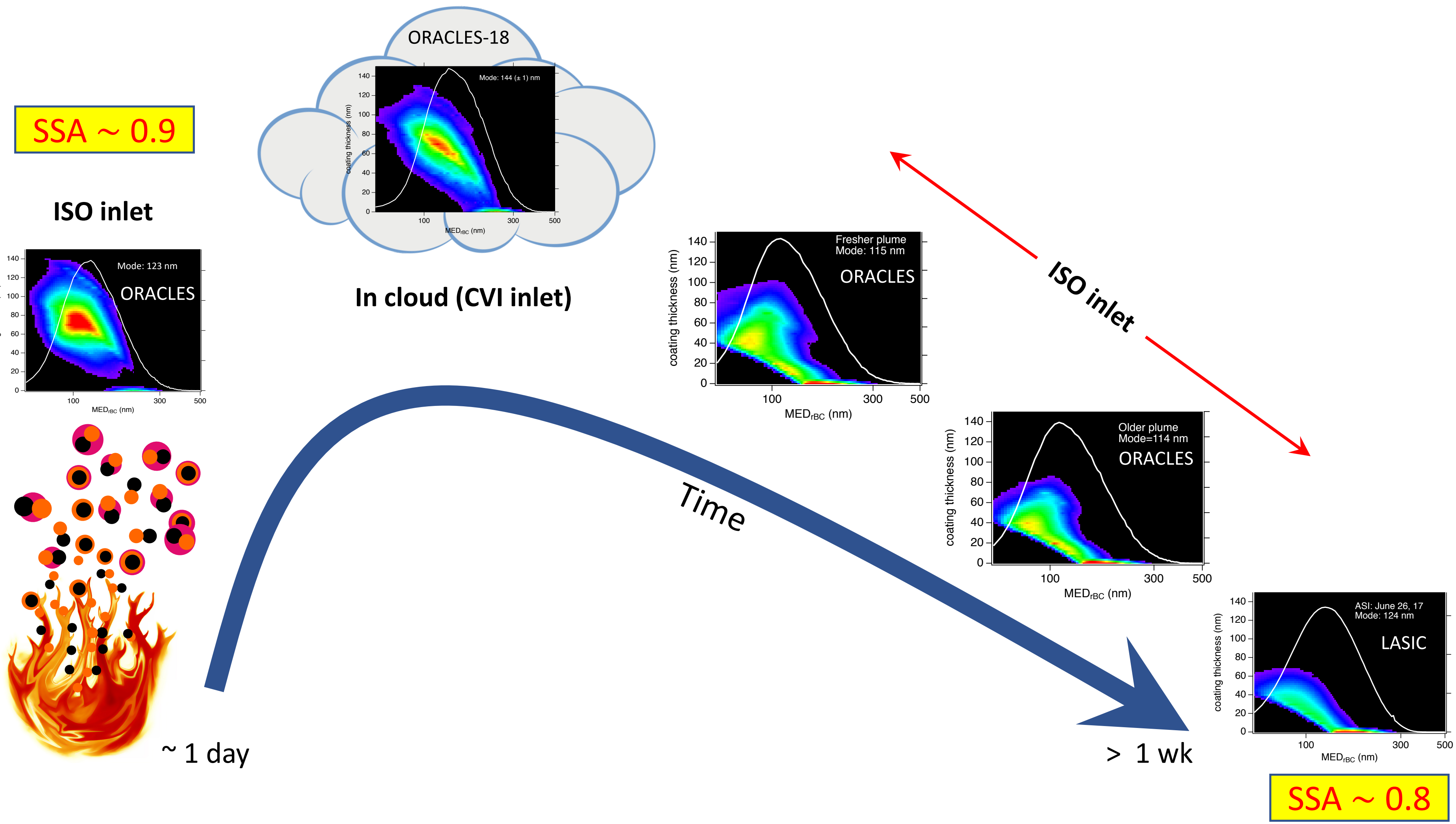
- κ = 0.15 (typical for organics)
- s_{crit} = 0.1% (insensitive, coupled to κ)



Green symbols from differing flights



rBC Particle Mixing States from LASIC & ORACLES

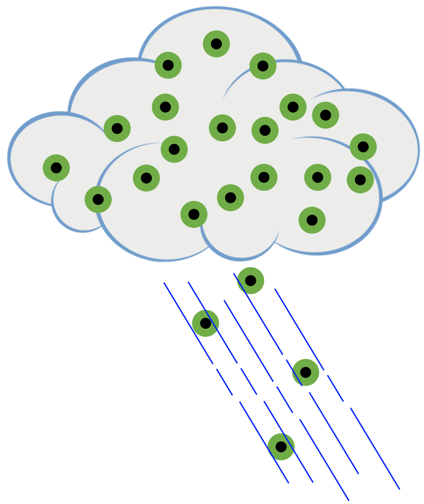


What drives the observed changes in mixing state and the SSA behavior?

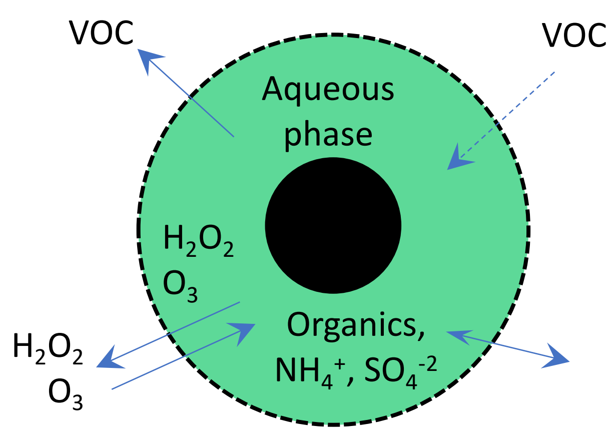
Activation and/or cloud processing

How Might Activation Promote Coating Loss

- Wet deposition through precipitation



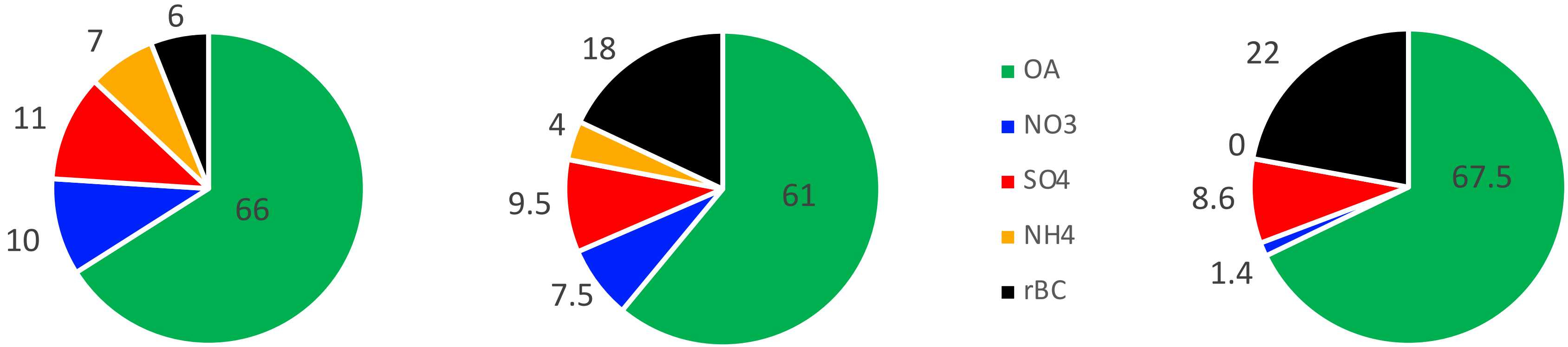
- Aqueous phase chemistry (molecular fragmentation → higher volatility species)



Comparison of Model and Measurement of BB SSA

- rBC: RI=1.8 – 0.8**i*; BrC: RI=1.5-0.01**i* (imaginary part restricted by initial SSA ~0.9)

Near Source§ | Older* | LASIC‡



SSA_{calc}: 0.94
SSA_{obs}: ~0.93

SSA_{calc}: 0.88
SSA_{obs}: ~0.85

SSA_{calc}: 0.76
SSA_{obs}: ~0.80

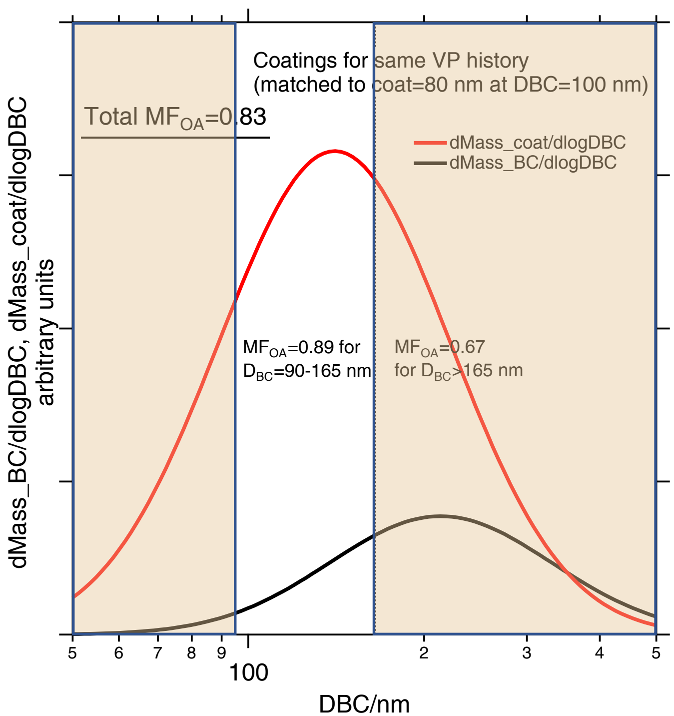
~ 1 day — Time — > 1 wk

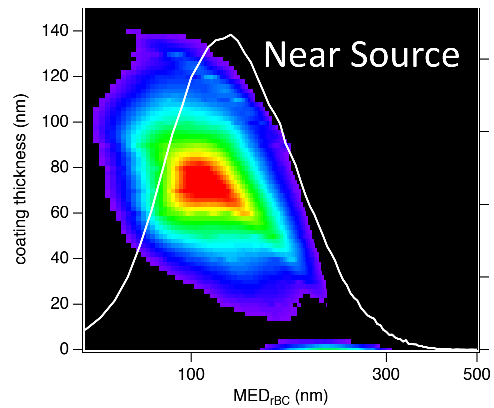
Greater proportion of rBC with time

§Dobracki 2019 private communication; *Dobracki et al., AMS 2019; ‡Aiken et al., DOE-ASR Sci. Team Mtg, 2018

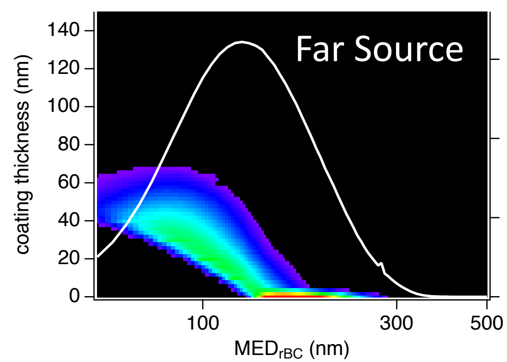
Model Prediction: Large Fraction of NR-Material is Coating

- Experimental measurement indicate the rBC cores are thickly coated
- Model presented here provides a way of estimating relative quantities (mass ratio)





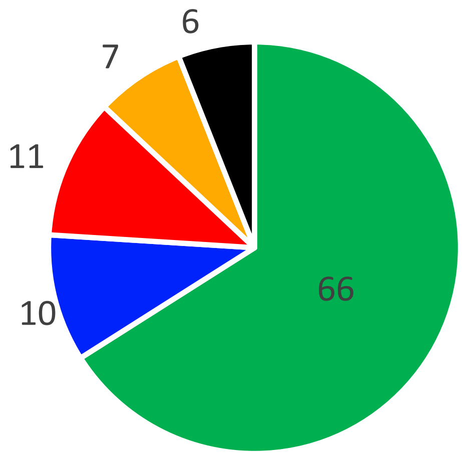
$$\frac{\text{Coating mass}}{\text{rBC mass}} = 13$$



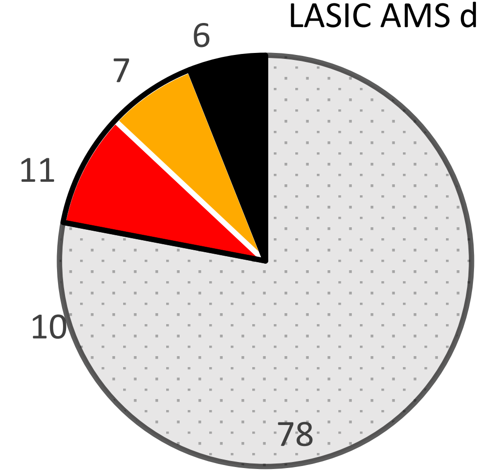
$$\frac{\text{Coating mass}}{\text{rBC mass}} = 2.8$$

How Much OA is Bound with rBC?

ORACLES AMS data courtesy of A. Dobracki
LASIC AMS data courtesy of A. Atkin et al.

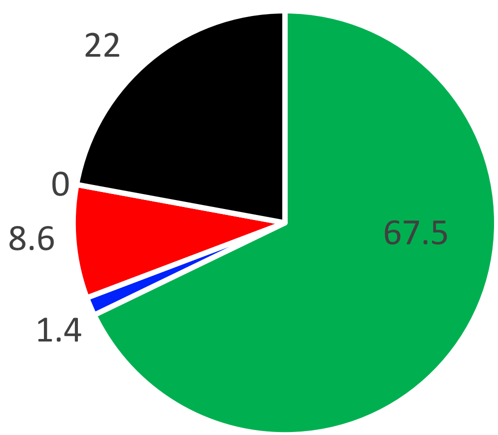


Near African Coast
(Near source)

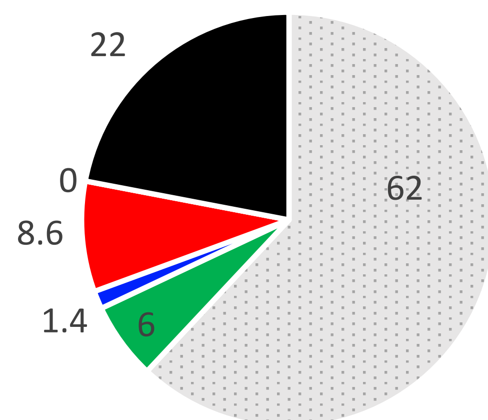


$$\frac{\text{Coating mass}}{\text{rBC mass}} = 13$$

OA NO3 SO4 NH4 rBC nR coating on rBC core



Ascension Island



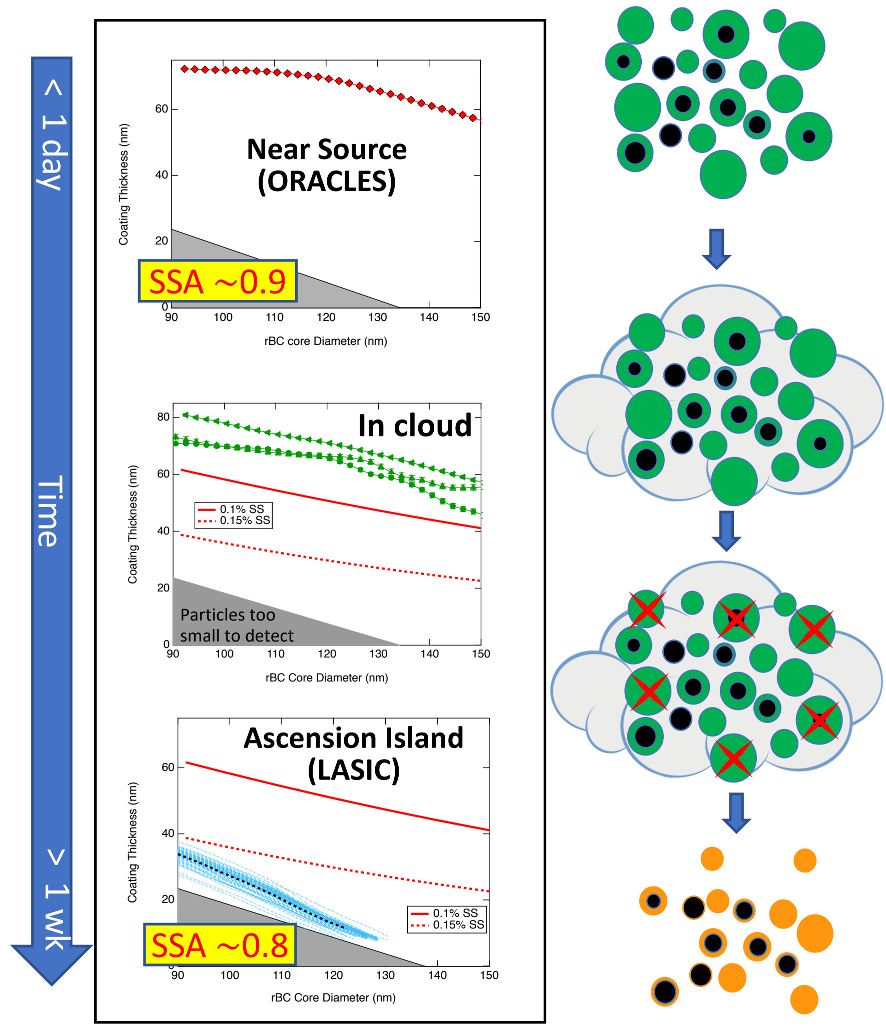
$$\frac{\text{Coating mass}}{\text{rBC mass}} = 2.8$$

A substantial fraction of non-refractory material is bound with rBC!

Activation/Cloud Processing Drives BB Optical Property Evolution

Take Home Messages

- BC from African biomass burns is thickly-coated in the near source and thinly-coated in the far field. (figs 1/3)
- SSA *decreases* from near source to far source. (figs 1/3)
- Thickly-coated BC particles are preferentially activated, enhancing their removal. (fig 2)
  - Reduction in light scattering
  - Decrease in SSA
  - Strong effect
- Brown Carbon (BrC) bleaching
  - Reduction in light absorption; little effect on scattering
  - Increase in SSA
  - Weak effect
- Much of the non-refractory material is bound with BC
- BrC plays secondary role to BC in determining SSA



Aerosol-cloud interactions in BB plumes drives BC to center stage

Research Associate Position in Aerosol Microphysics

Research Associate for Aerosol Microphysics

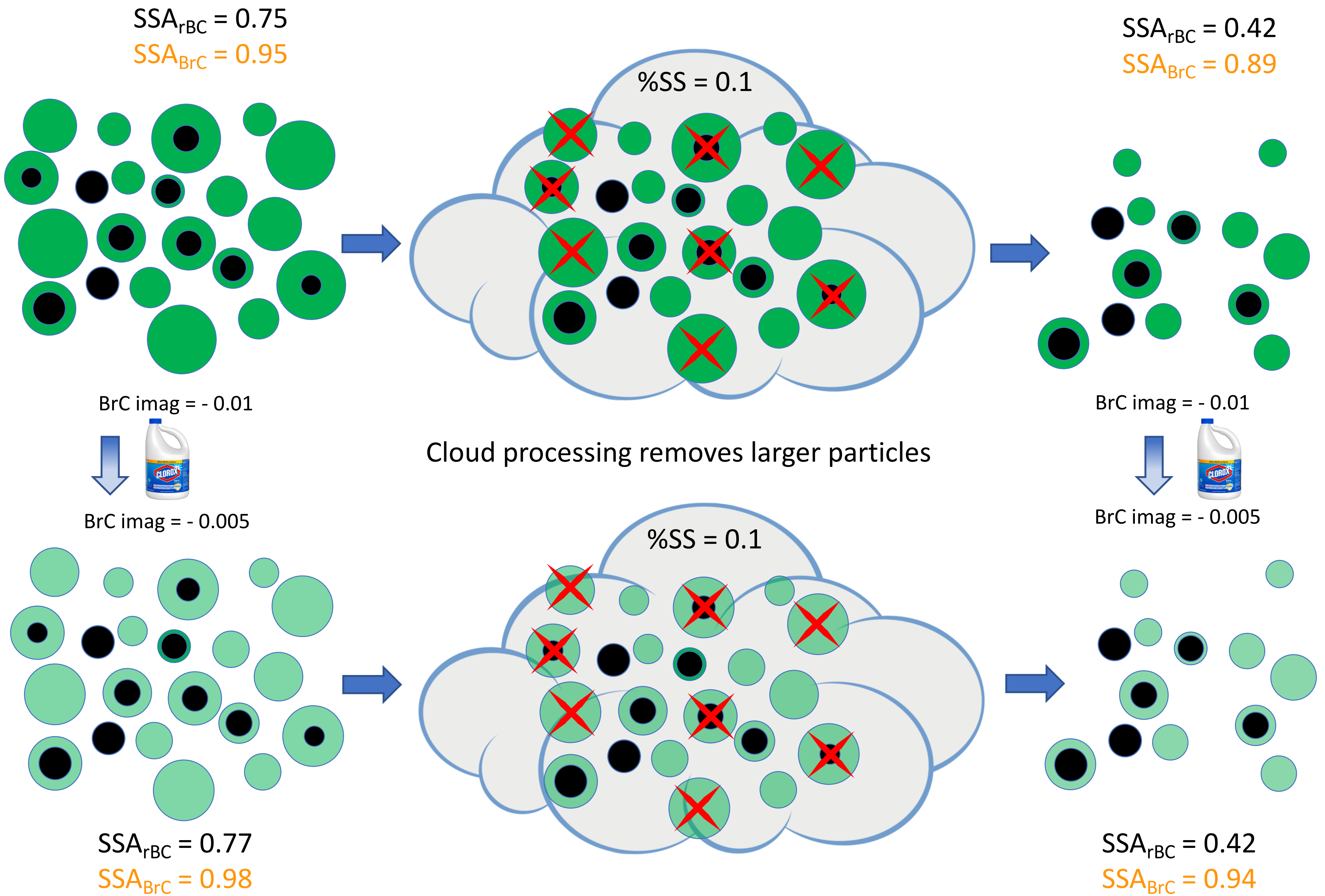
Research into the microphysical, optical, hygroscopic, and cloud-nucleating properties of aerosols, specifically light-absorbing aerosols, and how these properties affect radiation transmission through the atmosphere, including aerosol-cloud interactions.

Our laboratory is outfitted with several state-of-the-art instruments that include the Single Particle Soot Photometer (SP2), Centrifugal Particle Mass Analyzer (CPMA), BNL-designed Photothermal Interferometer (PTI) for measurement of light absorption, and Cloud Condensation Nuclei (CCN) counter, along with core aerosol instrumentation (e.g., particle counters, scanning mobility particle sizer, particle generation).

https://jobs.bnl.gov/job/upton/research-associate-for-aerosol-microphysics/3437/11754016

Extra slides

Model Prediction of Cloud Processing of Absorbing Aerosols



Microphysical Implications of rBC Mixing State

5) Optics: Mie code for core-shell to calculate SSA

- rBC: RI=1.8 – 0.8*i
- BrC: RI=1.5-0.01*i (imaginary part can't be larger because of initial SSA (~0.9))