# Aerosol Mixing State Focus Group Updates

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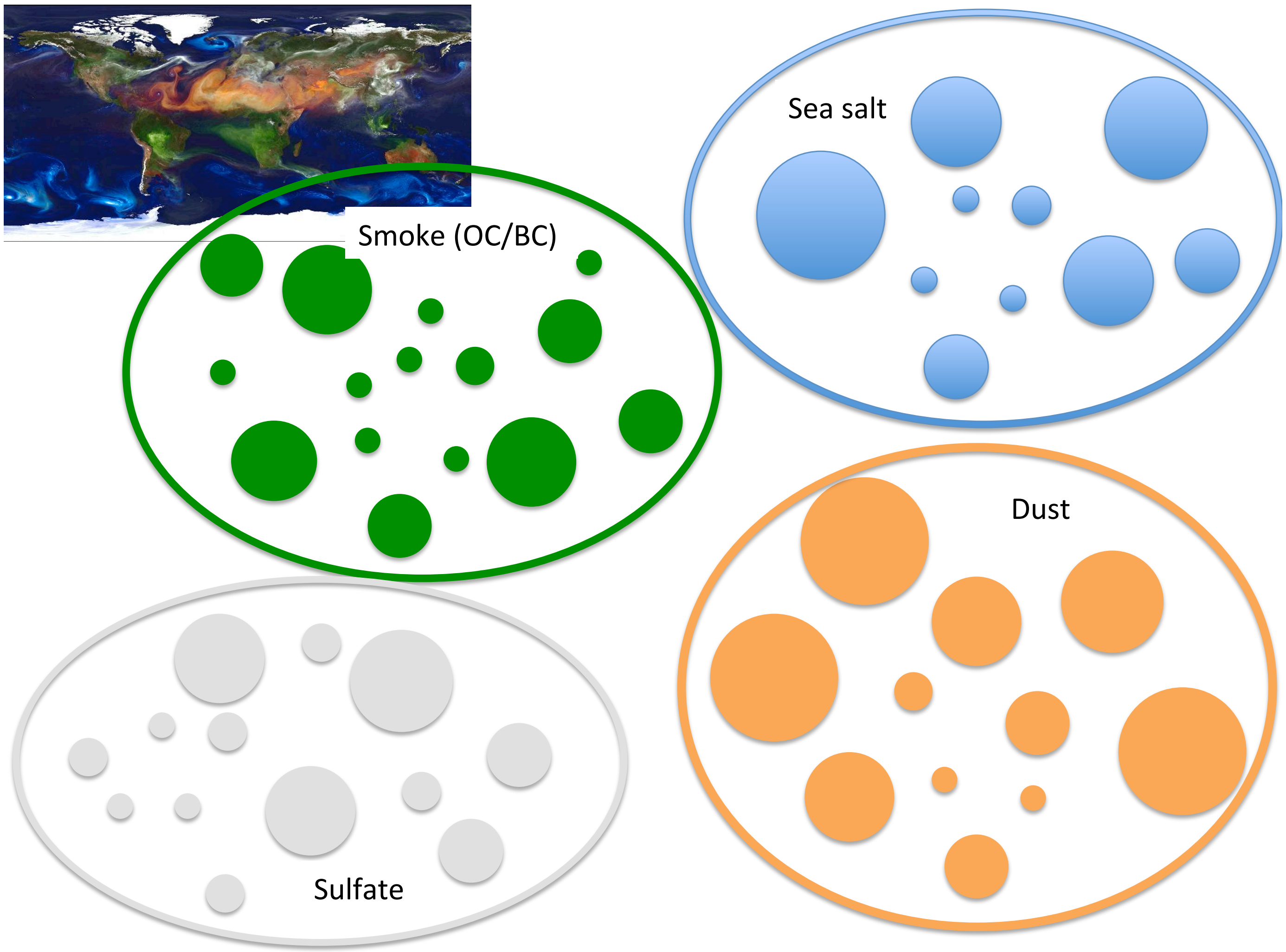
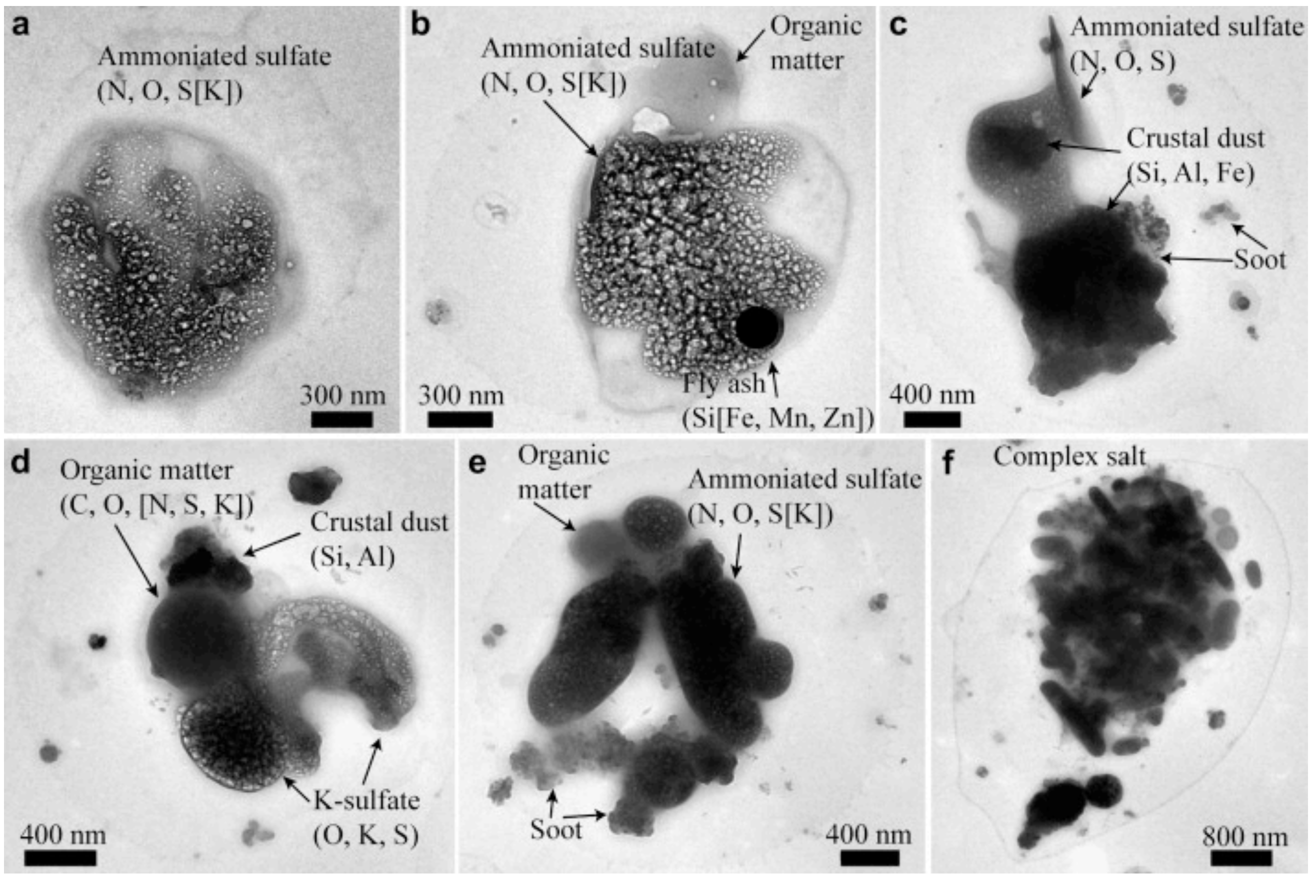
2015 ARM/ASR Joint User Facility PI Meeting
March 17, 2015

# Aerosol Populations in Current Models




External mixture of different aerosol types

# Real Particles in the Atmosphere




Li et al., Atmospheric Environment, 45, 2488-2495, 2011

How much detail is needed to capture aerosol impacts in large scale models?

# How important are these details?

Key question 1:

What is the impact of mixing state on CCN, IN, optical properties?

Key question 2:

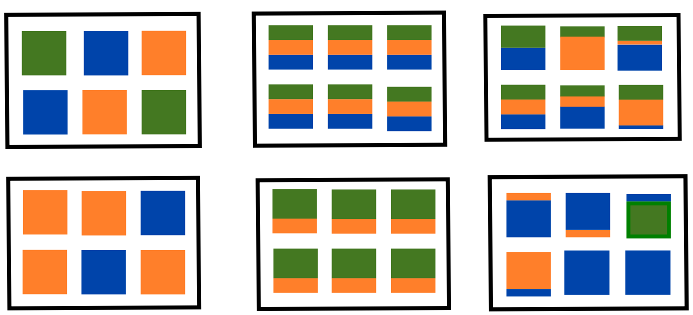
How should we include mixing state information in models that quantify aerosol climate impacts?

- What aerosol mixing states exist in different environments?
- How can we connect measurements (lab and field) to each other and to modeled mixing state information?
- What mixing state information should be measured in the field and in the lab?

# Two Definitions of “Mixing State”

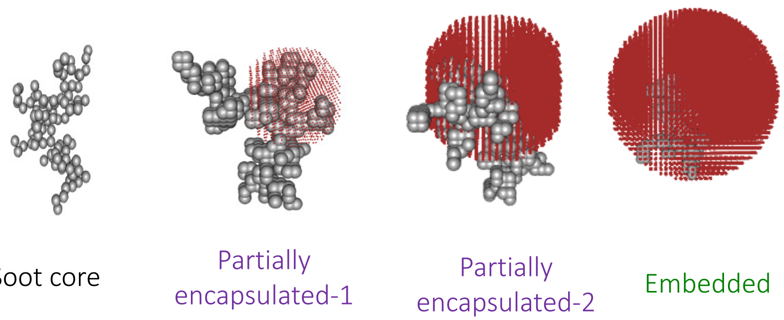
**Population mixing state:**

Distribution of chemical compounds across the particle population.

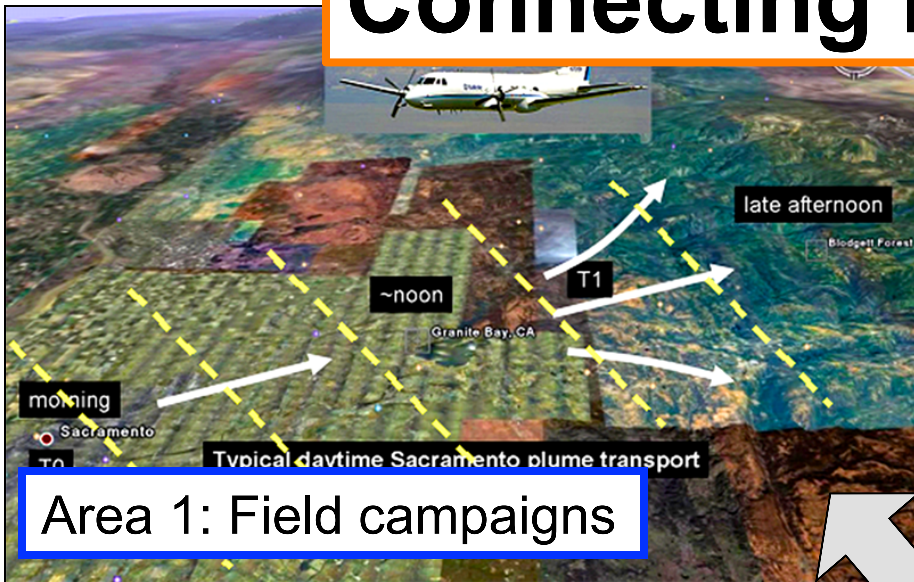


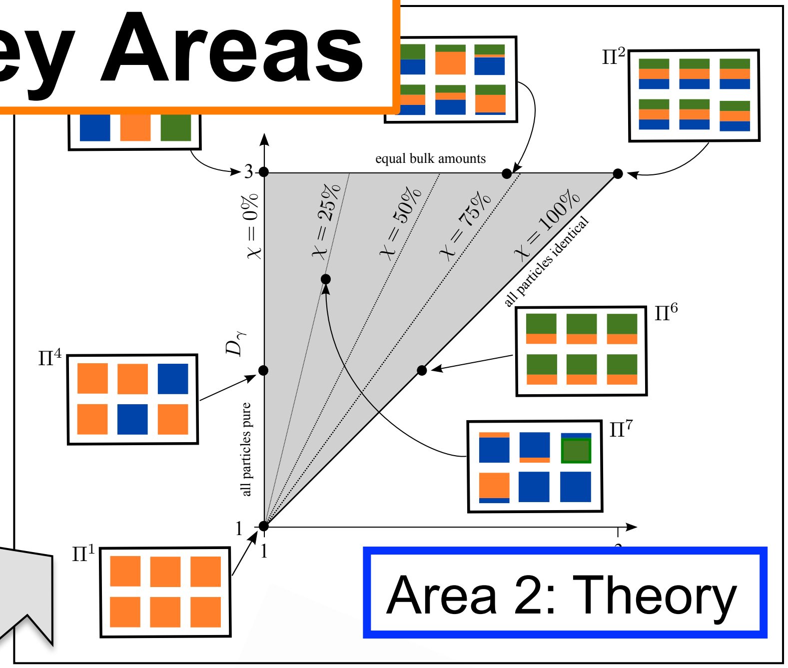
**Morphological mixing state**:

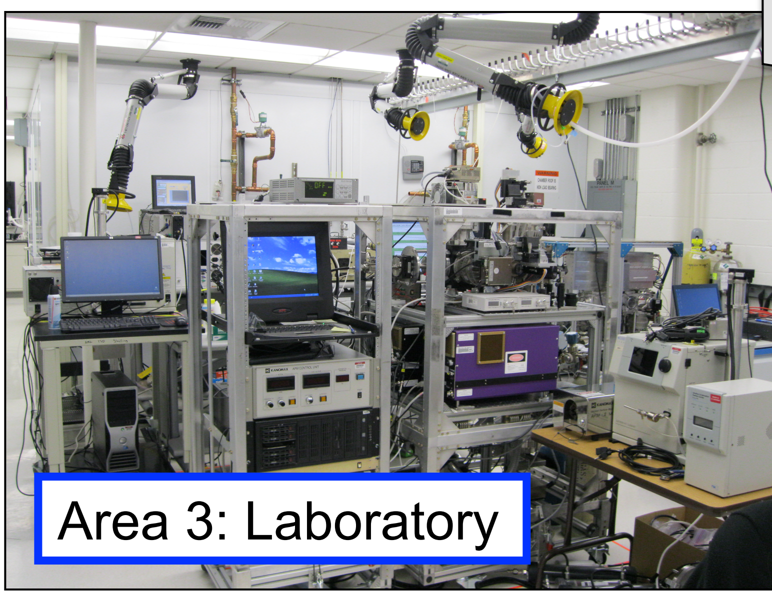
Distribution of chemical compounds within and on the surface of each particle.

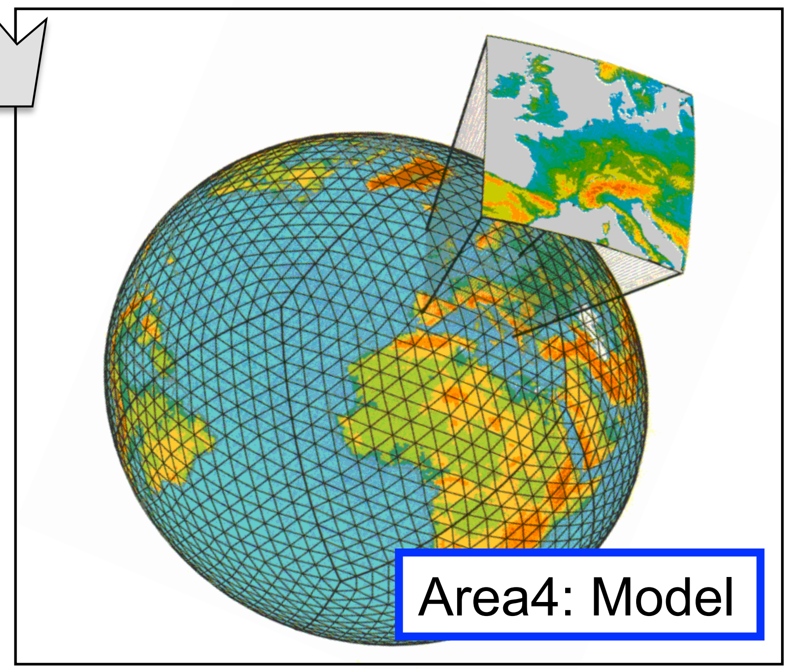


# Connecting Key Areas



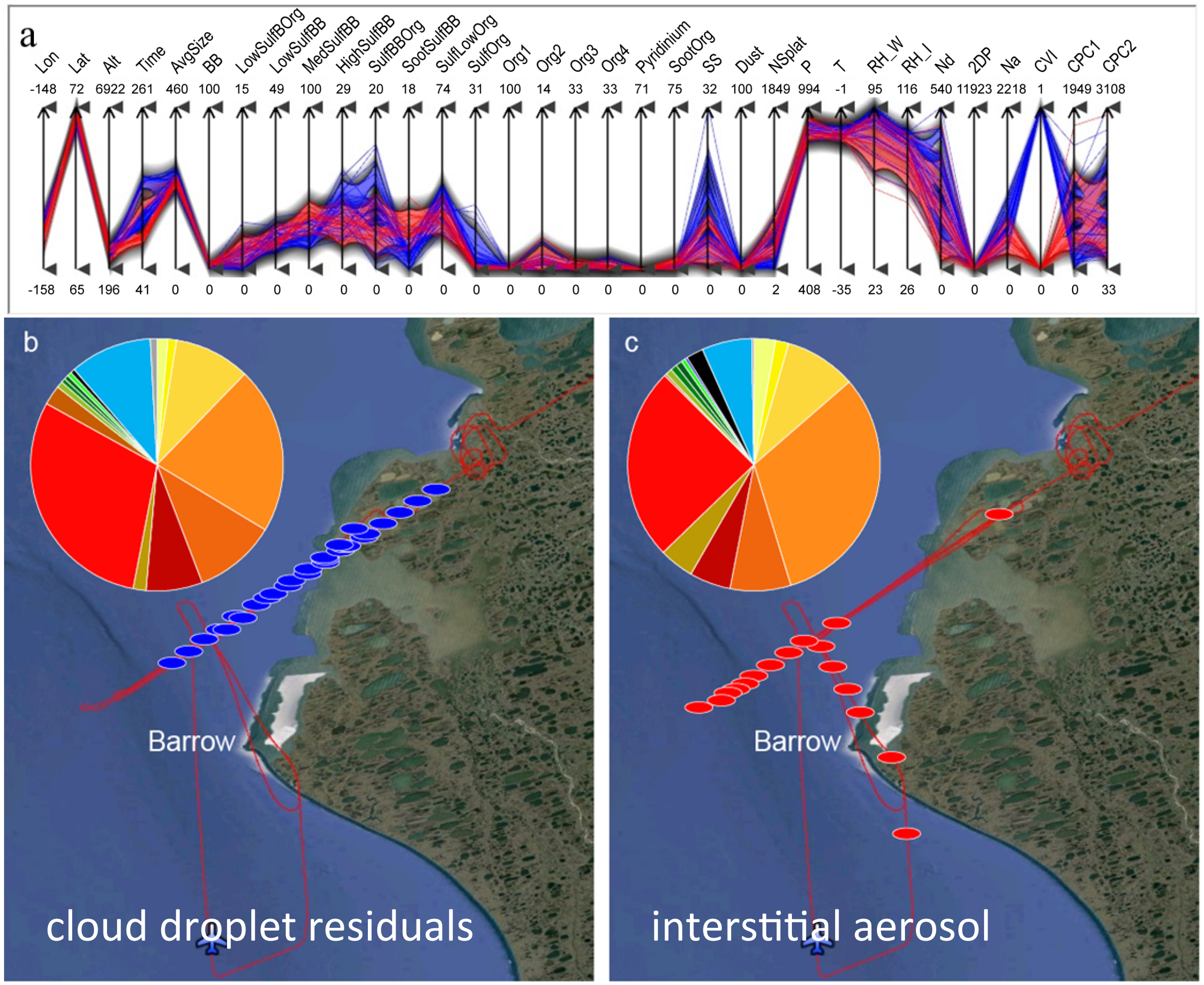



# Comparison of Mixing State of Cloud Droplet Residuals and Interstitial Particles






- Data collected by SPLAT II during ISDAC
- Used newly developed ND-Scope software.

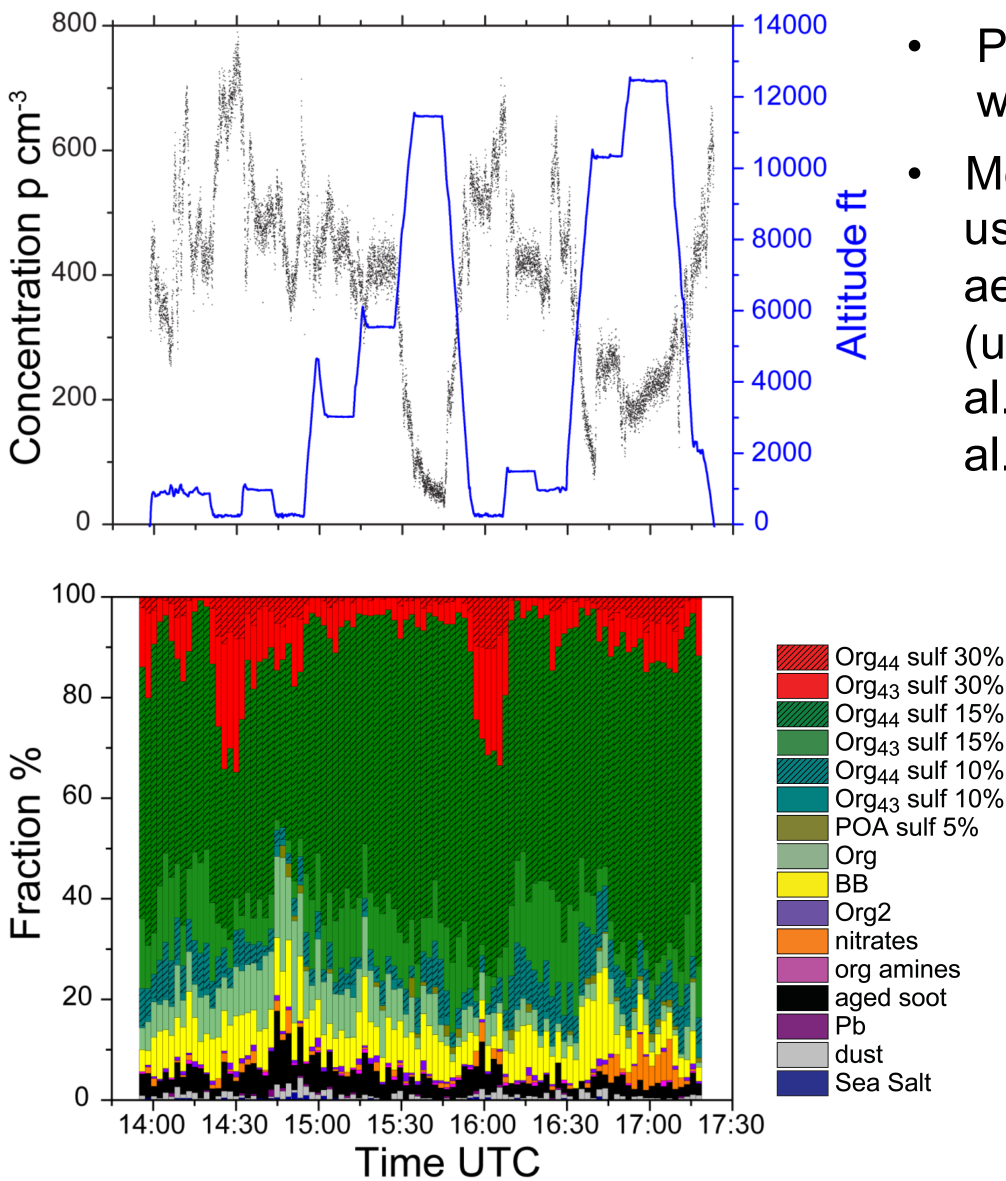
BB
BB_15%sulf
BB_40%sulf
BB_60%sulf
Sulfate

Organics
Piridinium
soot
Sea Salt
Dust

Zelenyuk at al. **(2015).** Airborne Single Particle Mass Spectrometers (SPLAT II & miniSPLAT) and New Software for Data Visualization and Analysis in a Geo-Spatial Context. *Journal of The American Society for Mass Spectrometry* 26:257-270.

# Mixing State of Aerosol Characterized during TCAP Field Campaign

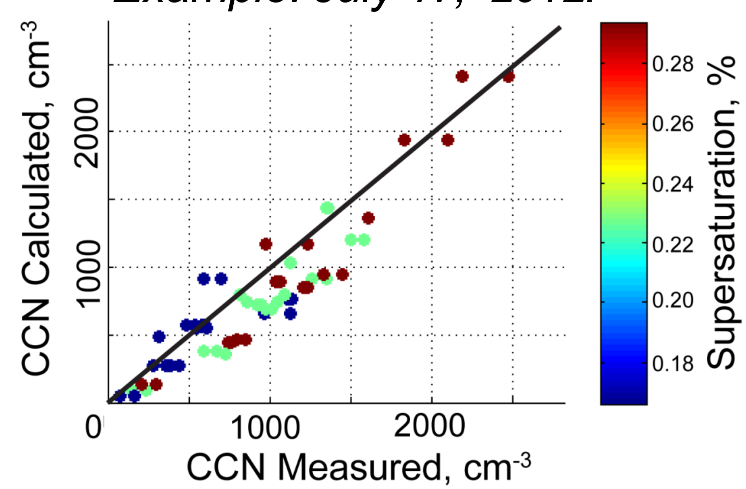





- Particle size and compositions change with time, altitude and location.
- Measured particle size and mixing state used for aerosol CCN closure and aerosol optical properties closure (using 4STAR (Segal-Rosenheimer et al. (2014) JGR), and HSRL-2 (Berg et al.; Chand et al.)).

Slide courtesy of Alla Zelenyuk

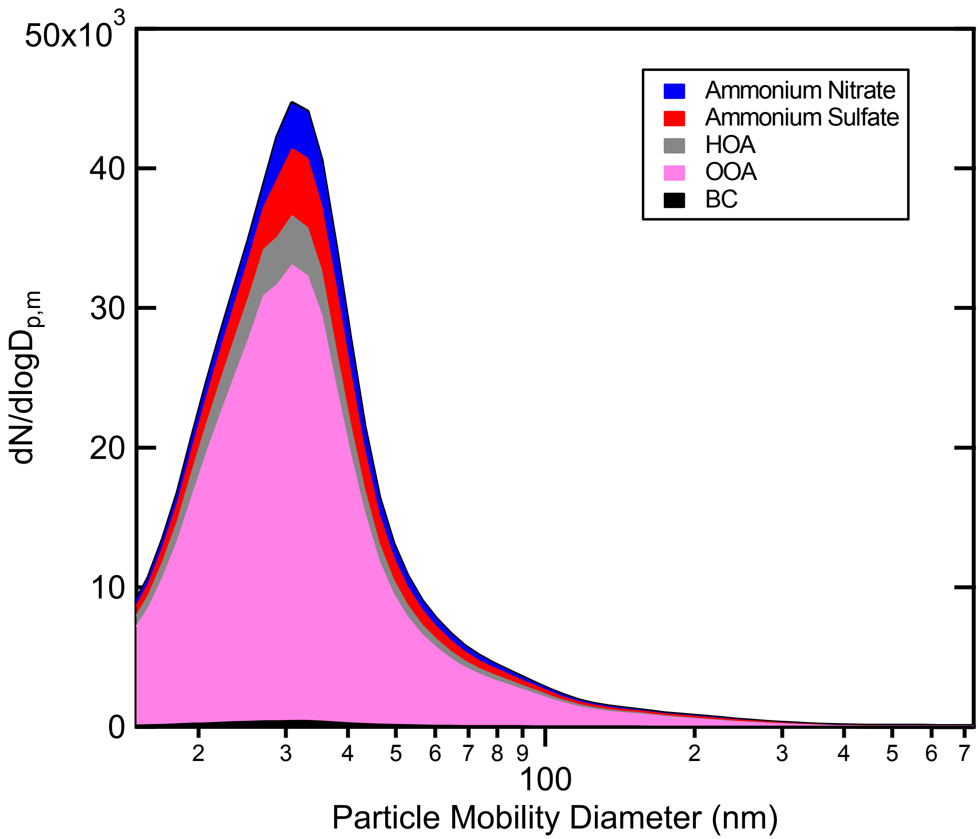
*Example: July 17, 2012.*



# Size-dependent composition mixing state and particle hygroscopicity

**Atkinson, Radney, Lum, Kolesar, Cziczo, Pekour, Zhang, Setyan, Zelenyuk and Cappa**

- Measurements of *f*(RH) from CARES were used to determine hygroscopic growth factors for submicron oxygenated organic aerosol and supermicron particles at the T0 and T1 sites
- Influence of particle mixing state (internal vs. external and size-dependent vs. size-independent composition) was assessed.




**<u>OOA</u>**

$k_{mean}$ = 0.08-0.16

GF(85%) = 1.11-1.22

**<u>Supermicron</u>**

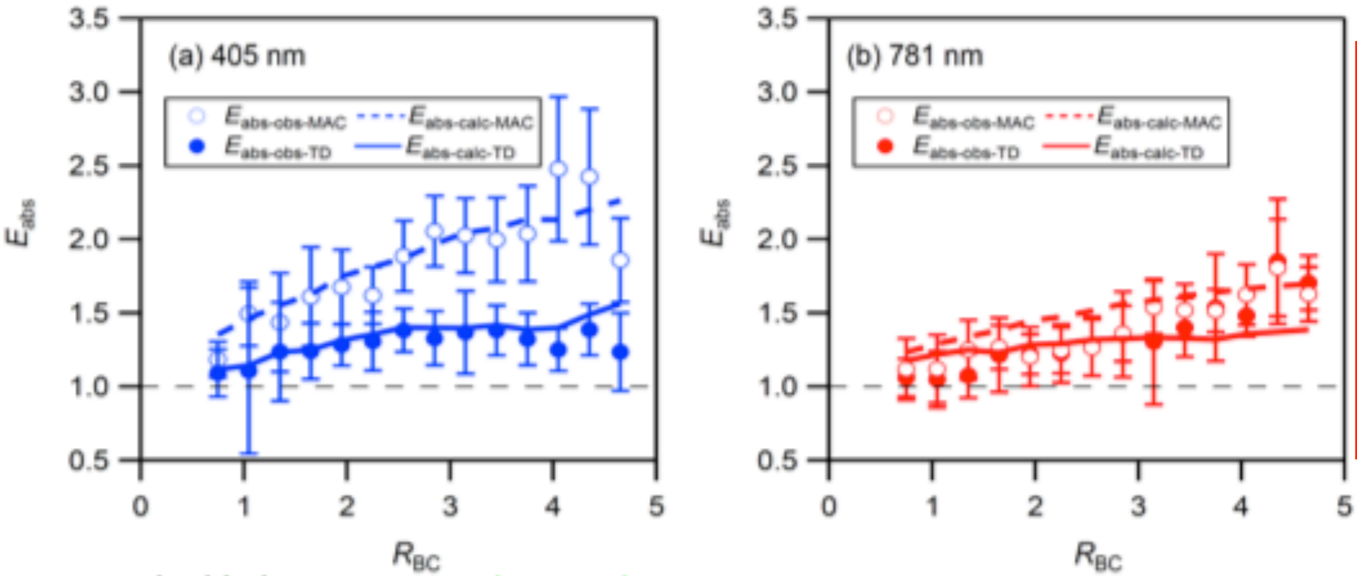
$k_{mean}$ = 0.8-1.2

GF(85%) = 1.75-1.97

- Overall, moderate influence of mixing state assumption on optically-relevant hygroscopicity (not considering absorption)
- Compositional variability in supermicron found important, yet under-characterized

# BC absorption enhancement high in Detling (Clearflo) and low in Sacramento (CARES): Mixing State Effect

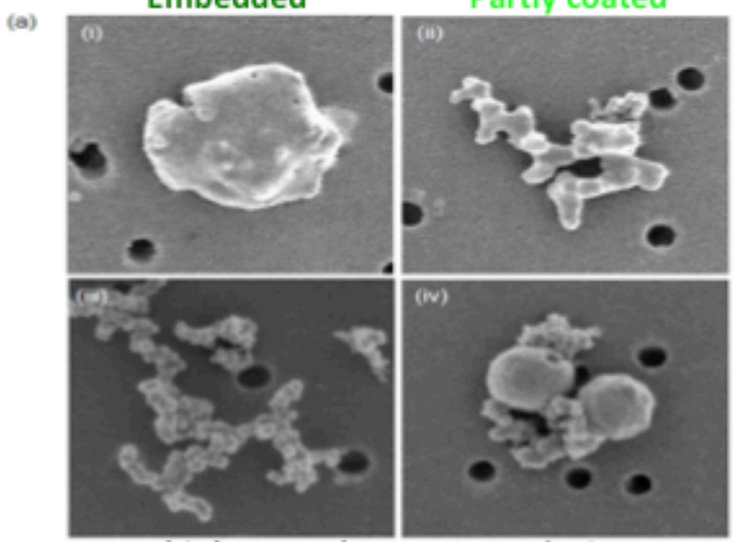
T-Denuding
SP2, PASS3,
SP-AMS




*Liu et al submitted Nat. Geo 2015*
*Dubey et al Poster 2015*

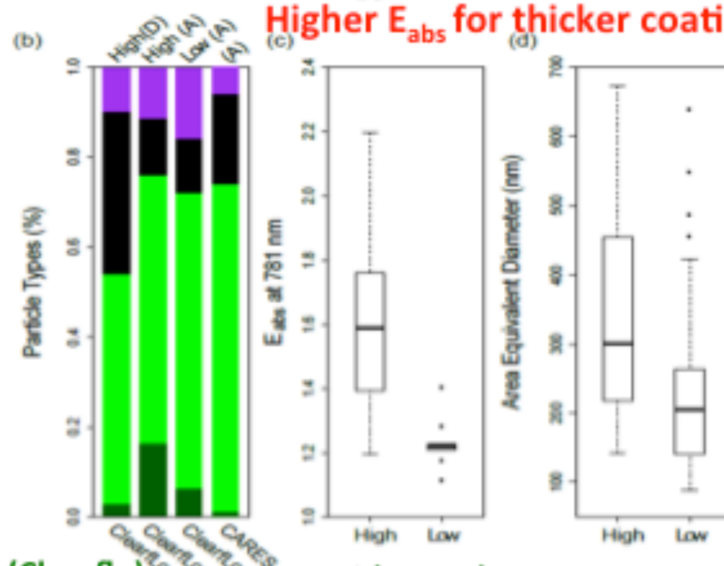
SEM

Embedded
Partly coated




Thinly coated
Inclusion

Higher $E_{abs}$ for thicker coating

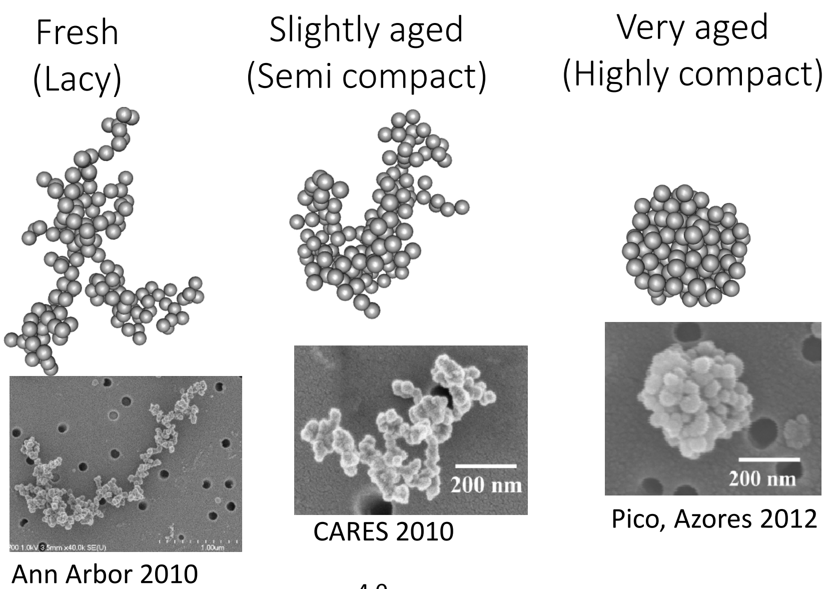



12% (Clearflo) Embedded 1% (CARES)

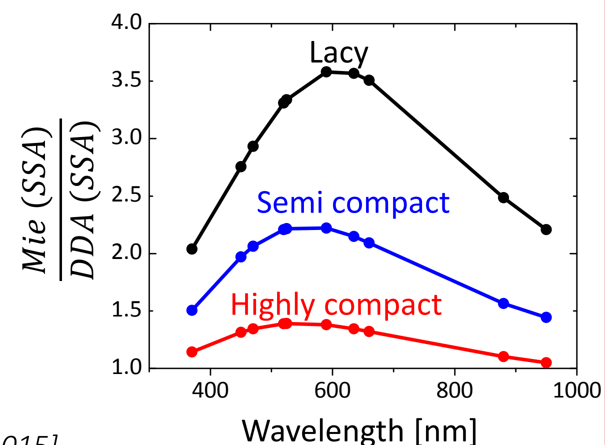
# Morphological effects on BC optical properties

S. China, B. Scarnato, C. Mazzoleni et al.

## Shape

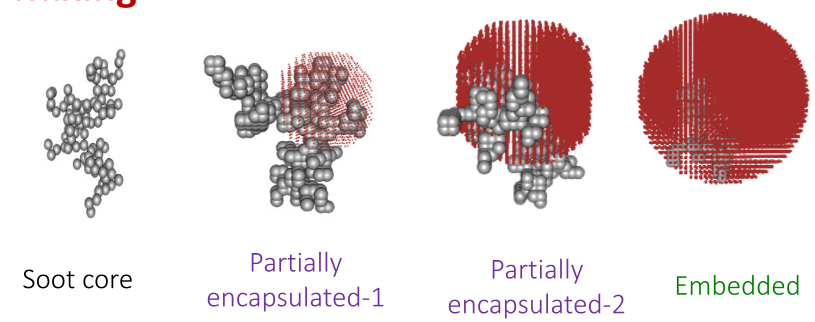


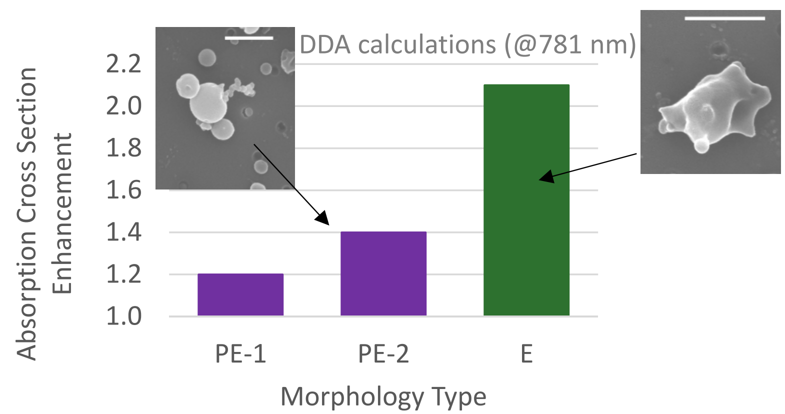

*DDA:*
*Discrete*
*Dipole*
*Approximation*



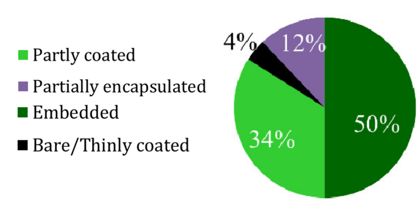

*[China et al. GRL, 2015]*

## Mixing




Example: Las Conchas Fire, NM



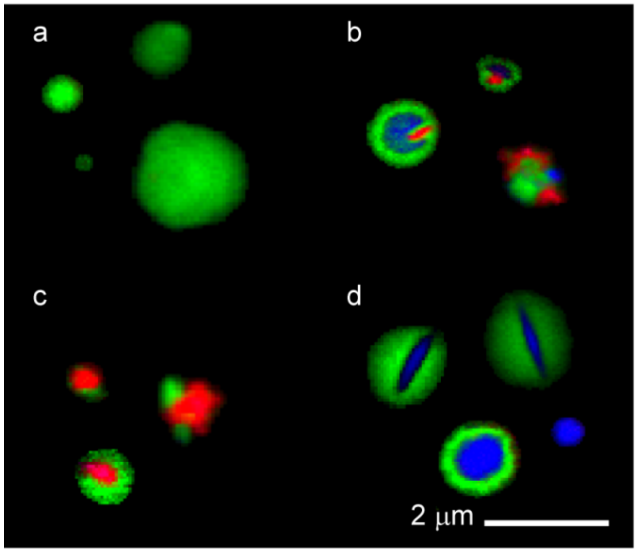

Total estimated absorption enhancement

$E_{abs}=1.59$

*[China et al., AGU 2014]*

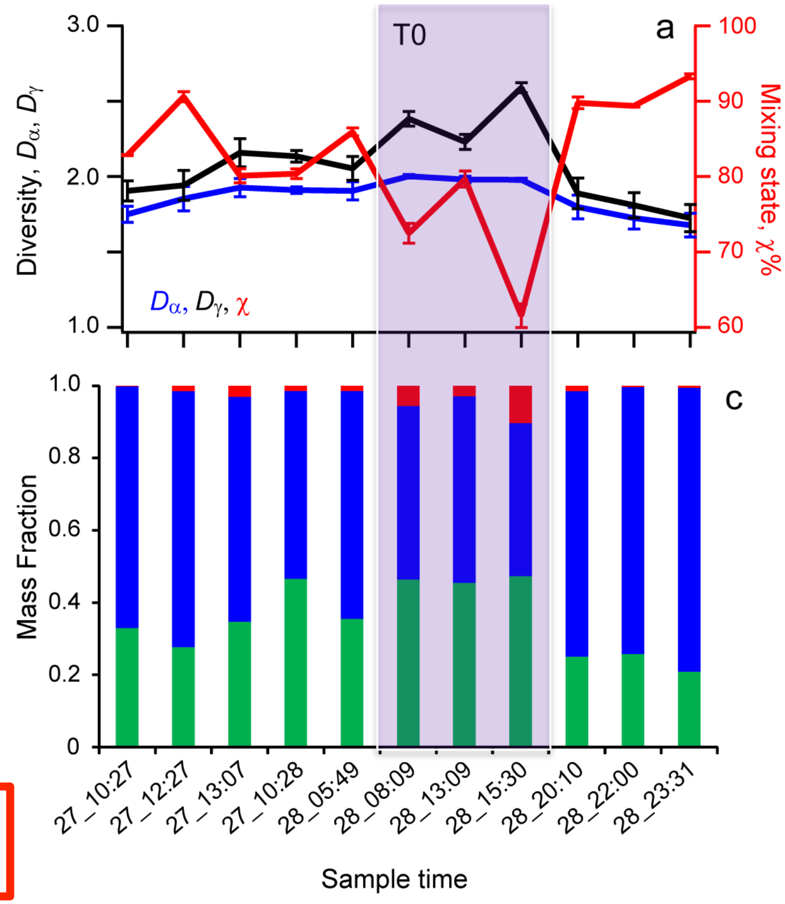
*See poster 32, session 1: Mazzoleni et al.*

# Carbonaceous Mixing State via X-Ray Microscopy




Organic (OC)
Inorganic (IN)
Soot/Black Carbon (BC)

Rachel O'Brien, Ryan Moffet, Mary Gilles, Alex Laskin, Bingbing Wang




Organic build-up event → decreasing mixing state

Increase in soot-dominated particles

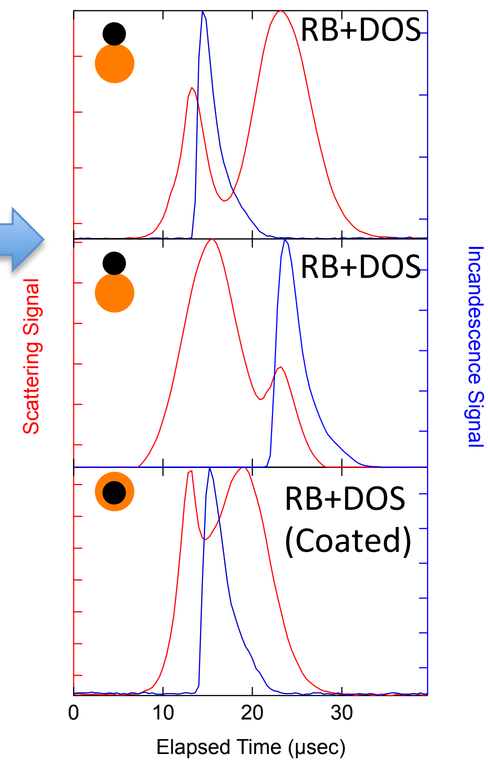
- Mixing state parameterization of imaged atmospheric particles
- Particle size, composition, and location of inclusions
- Cares 2010: evidence of local emissions (T0) and organic condensation (T1)

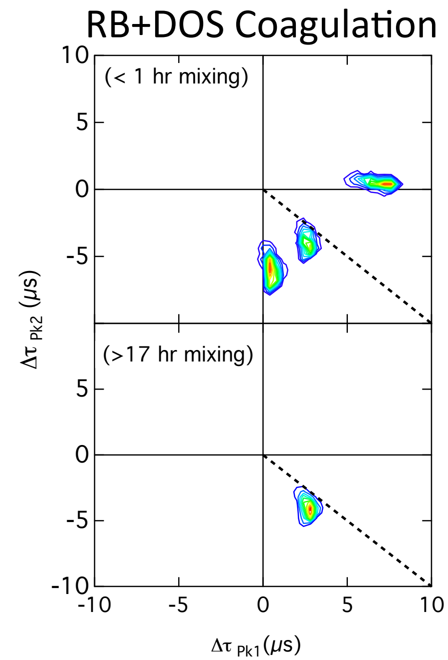
# Change in SP2 Scattering Signal with Plume Age

**Sedlacek,**
**Lewis, Onasch, Kleinman,**
**Lambe, Davidovits**

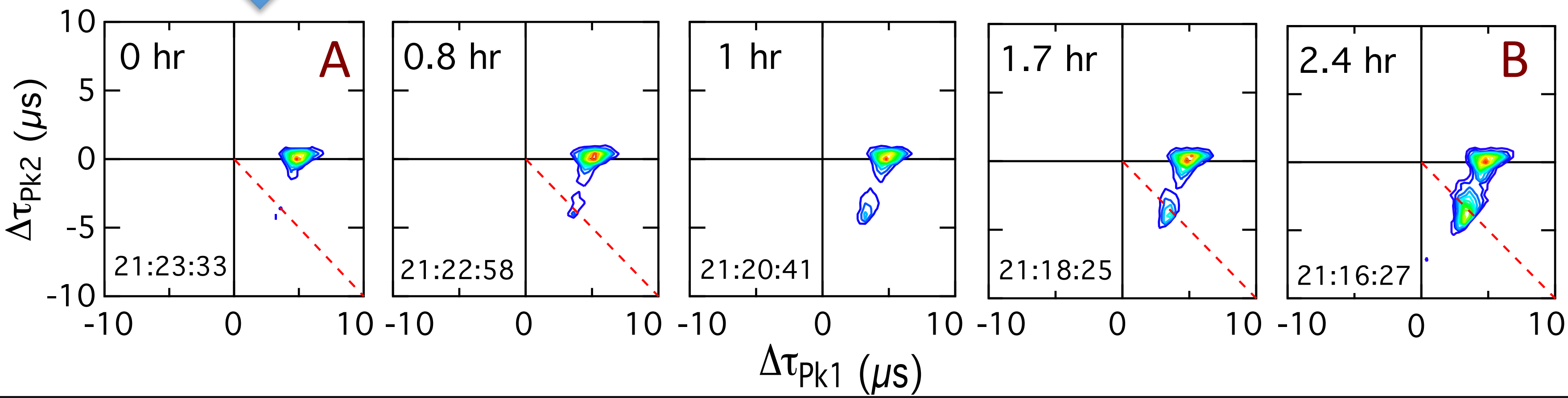
BC3: Systematic experiments suggest that SP2 scattering signal may provide probe of rBC-containing particle morphology.

BBOP: Centerline transect provides opportunity to examine rBC particle morphology as a function of plume age




$\tau_{Pk1}$: First scat peak to Incand Peak
$\tau_{Pk2}$: Second Scat Peak to Incand Peak



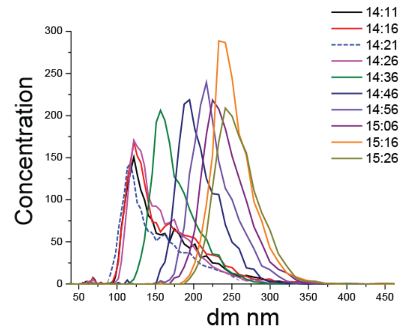

Fragmentation pattern suggests increase in thickly-coated rBC-containing particles with time

# Soot Aerosol Aging Study (SAAS)

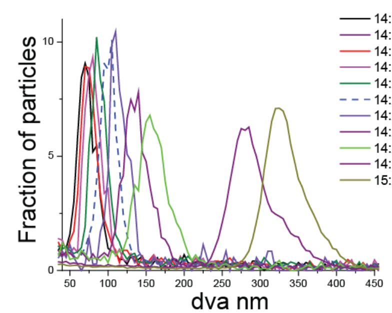
How do soot mixing state and morphology evolve due to condensation and coagulation with SOA and how this aging effects optical properties, CCN, and IN?

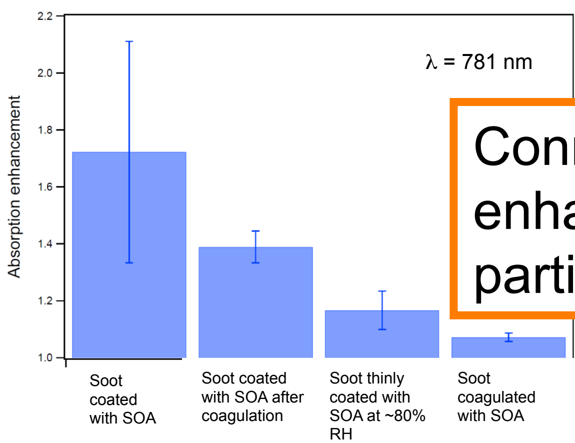
Aerosol mixing state data in controlled laboratory conditions

SMPS



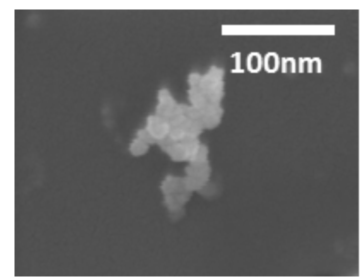

SPLAT




Soot before coating

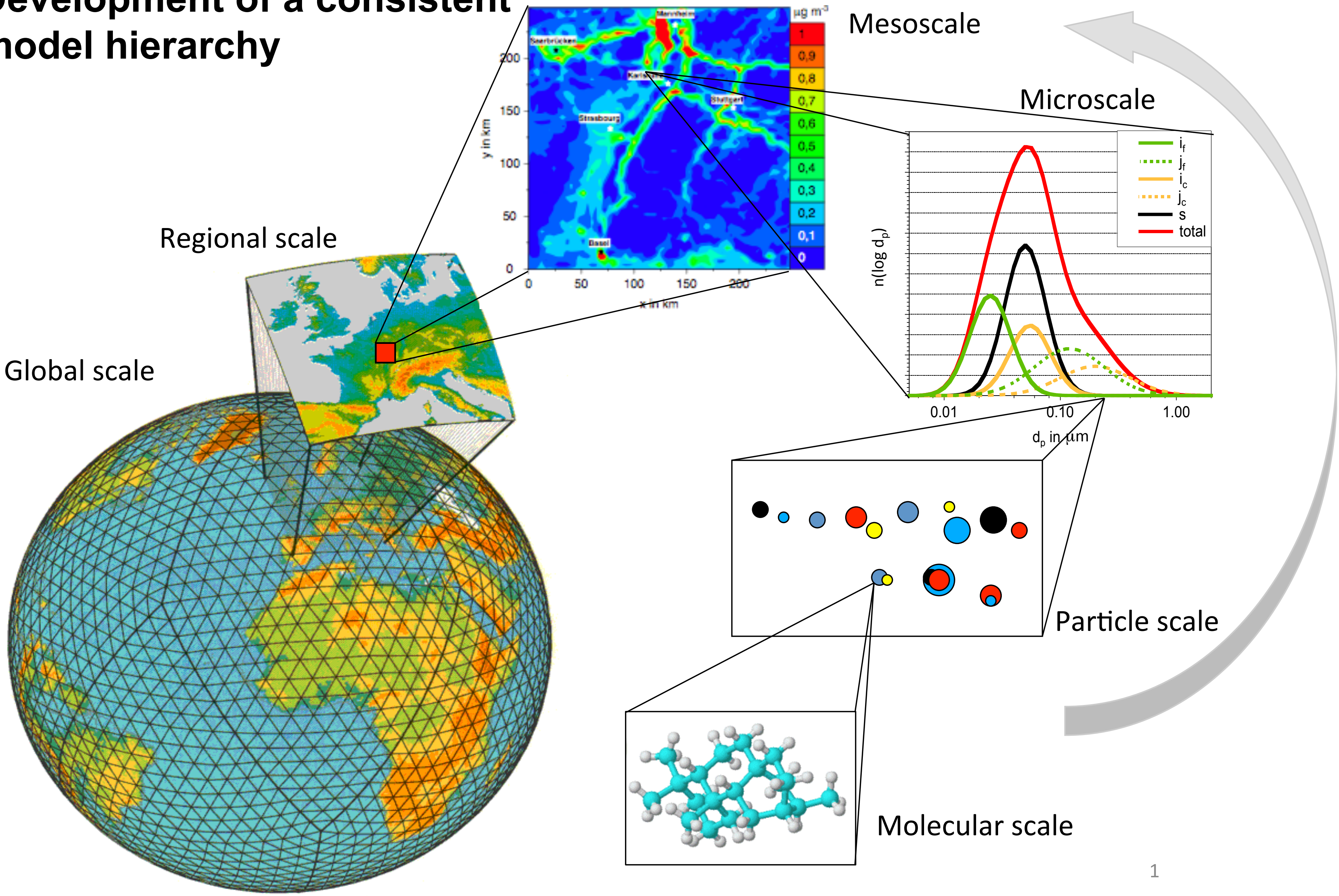
After Coating




Connecting absorption enhancement with single-particle mixing state data

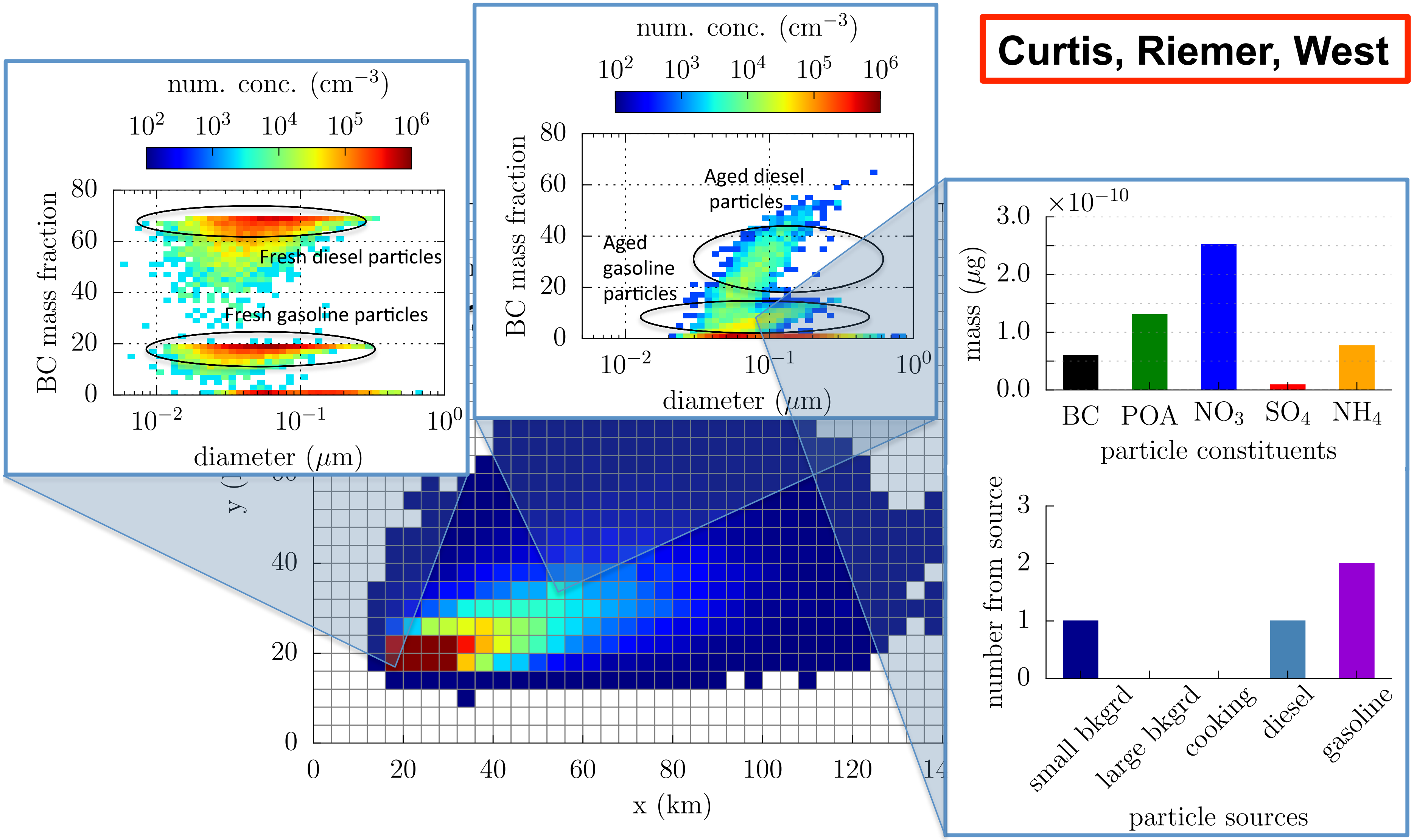
**R.A. Zaveri, J.E. Shilling, M. Pekour, G. Kulkarni, D. Chand, J. Wilson, A. Zelenyuk-Imre, A. Laskin, S. Liu, A. Aiken, M. Dubey, R. Subramanian, N. Sharma, S. China, C. Mazzoleni, A. Sedlacek, T.B. Onasch, R. Sellon, M.K. Gilles, and R. Moffet**

# Development of a consistent model hierarchy



**WRF-PartMC-MOSAIC**. Particle-resolved physics and chemistry coupled with 3D dynamics

Curtis, Riemer, West




**Processes:** coagulation, gas chemistry, aerosol-gas interactions, advection and diffusion.

**Computation:** 40 x 30 x 60 domain, 10,000 particles per grid cell (250 million total).

**Runtime:** 12 wall hours on 60 cores to simulate 2 model hours.

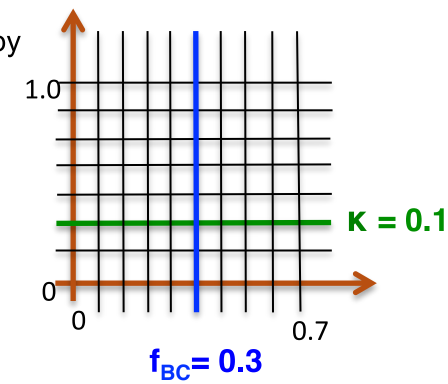
# Black Carbon Mixing State Modeling

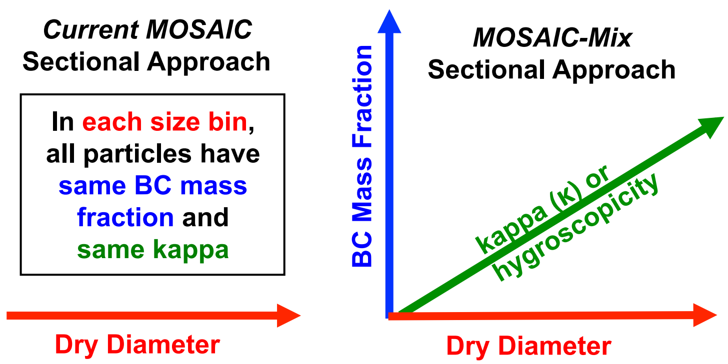
J. Ching. R. Zaveri, R. Easter, N. Riemer, J. Fast – Submitted soon to *J. Geophys Res.*

Pacific Northwest
NATIONAL LABORATORY

**Internal mixture assumption for BC in sectional aerosol models can lead to significant errors in the predicted CCN concentrations and optical properties. Thus, our goal is to develop a computationally efficient mixing state representation for black carbon in the sectional aerosol model MOSAIC to improve predictions of CCN concentrations and optical properties.**

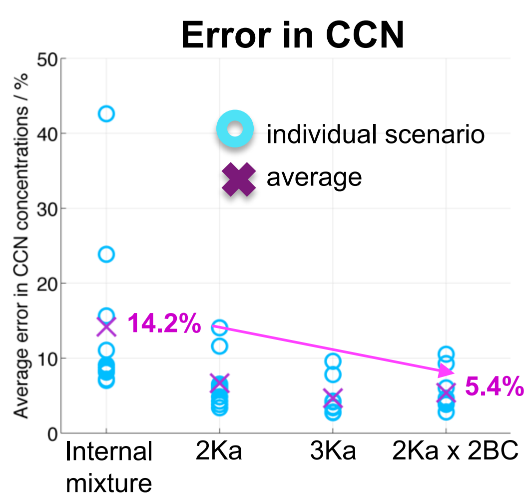
- 10 scenarios simulated by MOSAIC-Mix
- Over 2000 combinations of $f_{BC}$ and κ boundaries tested
- Optimum $f_{BC}$ and κ bin boundaries located, that give the **minimum overall error**

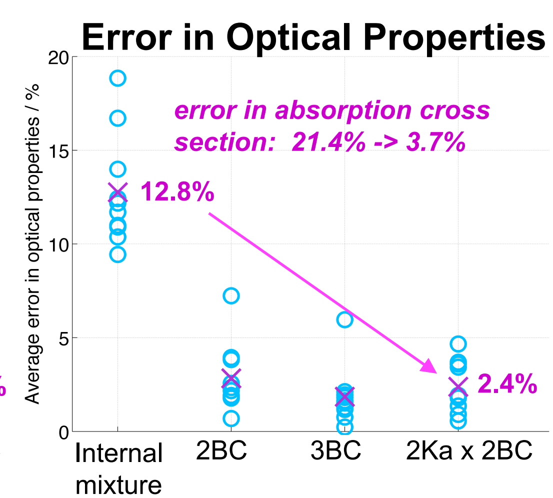



A novel 3-dimensional mixing state representation for aged black carbon particles is developed that is different from "brute force" approaches or ad-hoc choices of bin boundaries. A more systematic approach is used to determine best bin boundaries.

- A simple ***2 $f_{BC}$ x 2 κ bins achieves* smaller total error** (CCN + optical) than 2 *or 3* $f_{BC}$ bin only or 2 *or 3* κ bin only configurations.
- MOSAIC-Mix will be implemented in WRF-Chem to evaluate its performance using field measurements and assess the impact of BC mixing state on direct and indirect radiative forcing.

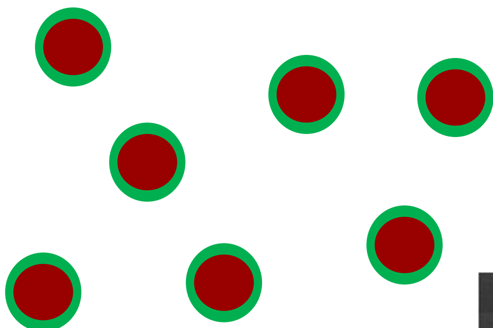


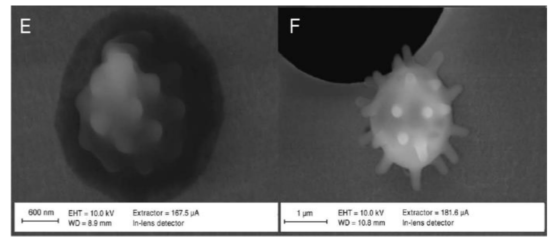



# Ice Nucleation?
# Breakout session on Thursday 8:00-10:00AM

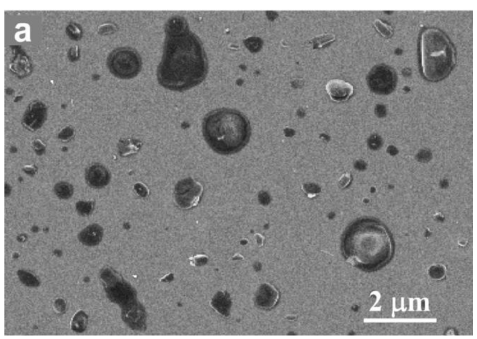
## How Does Mixing state Impact Ice Nucleation?

Air mass consisting of internally mixed brown and green particles:

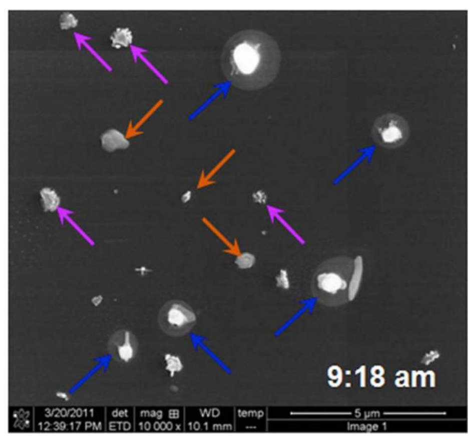




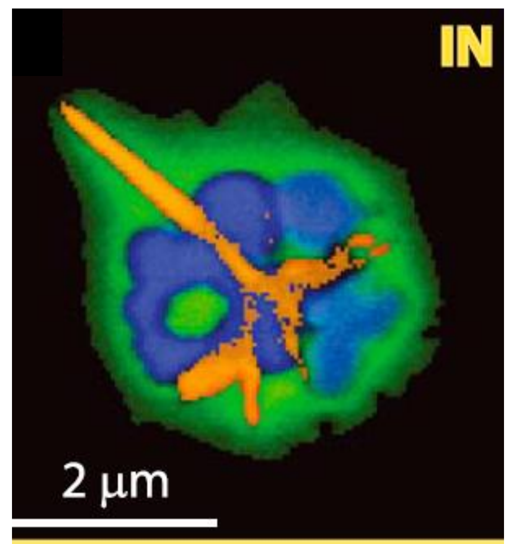

Pöschl et al., Science, 2010




Knopf et al., GRL, 2010




Laskin et al., JGR, 2012




Knopf et al., JGR, 2014

Slide courtesy of Daniel Knopf

# Progress and Outcomes

- Cross-cutting collaborations between key areas, enabled by having a focus group.
- Unified conceptual framework for quantifying aerosol population mixing state based on per-particle species mass fractions and diversity metrics.
- On our way to developing a consistent model hierarchy.
- Controlled chamber experiments on aerosol mixing state evolution.

# Current Priorities and Opportunities

- Confronting particle morphology:
  - Metrics for morphology
  - Process-level understanding of how particle morphology evolves
- Quantitative population mixing state information from experimental data, using per-particle species mass fractions.
- Information on population mixing state of emissions is bottleneck for modeling.
- Population mixing state inter-comparison studies between global, regional, and particle-resolved aerosol models.