Black Carbon, Air Quality and Climate

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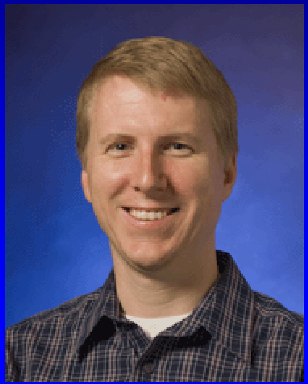
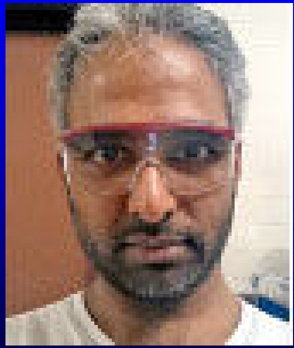

Peter Adams

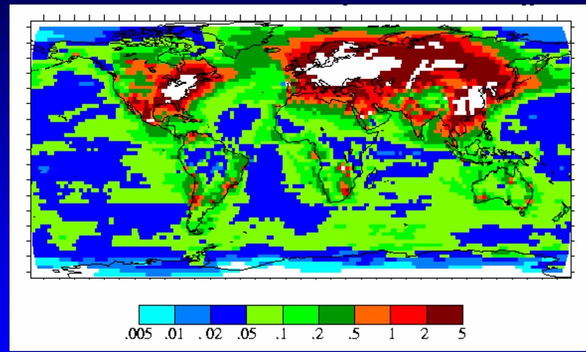

Rawad Saleh


R. Subramanian

Black Carbon, Air Quality and Climate

Sources of BC

?



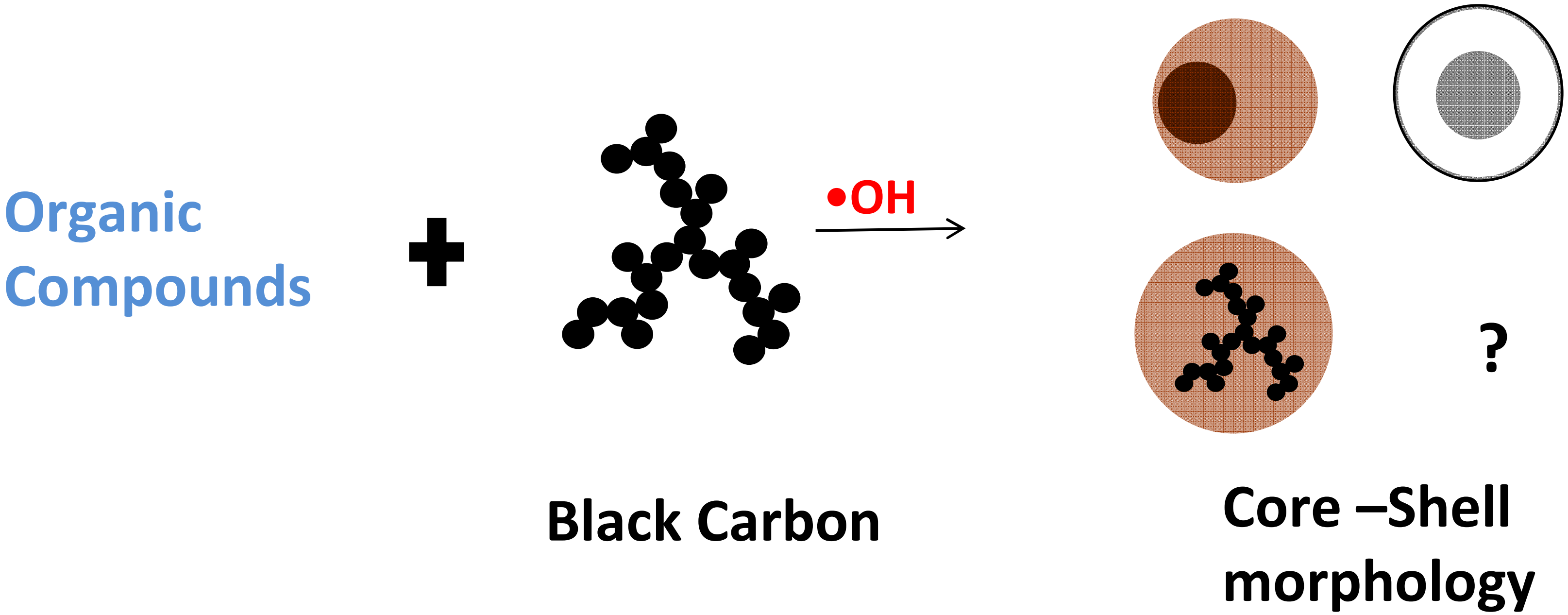
Objectives

- Improve our understanding of the optical properties of BC-containing particles and their evolution during their lifetime
- Link emissions of BC particles with particle number concentrations over the US
- Improve the ability of the existing regional models to simulate the BC mass and number concentrations
- Quantify effects of changes in BC emissions in PM and PN over the US

Chemical Aging and Optical Properties of BC Emissions

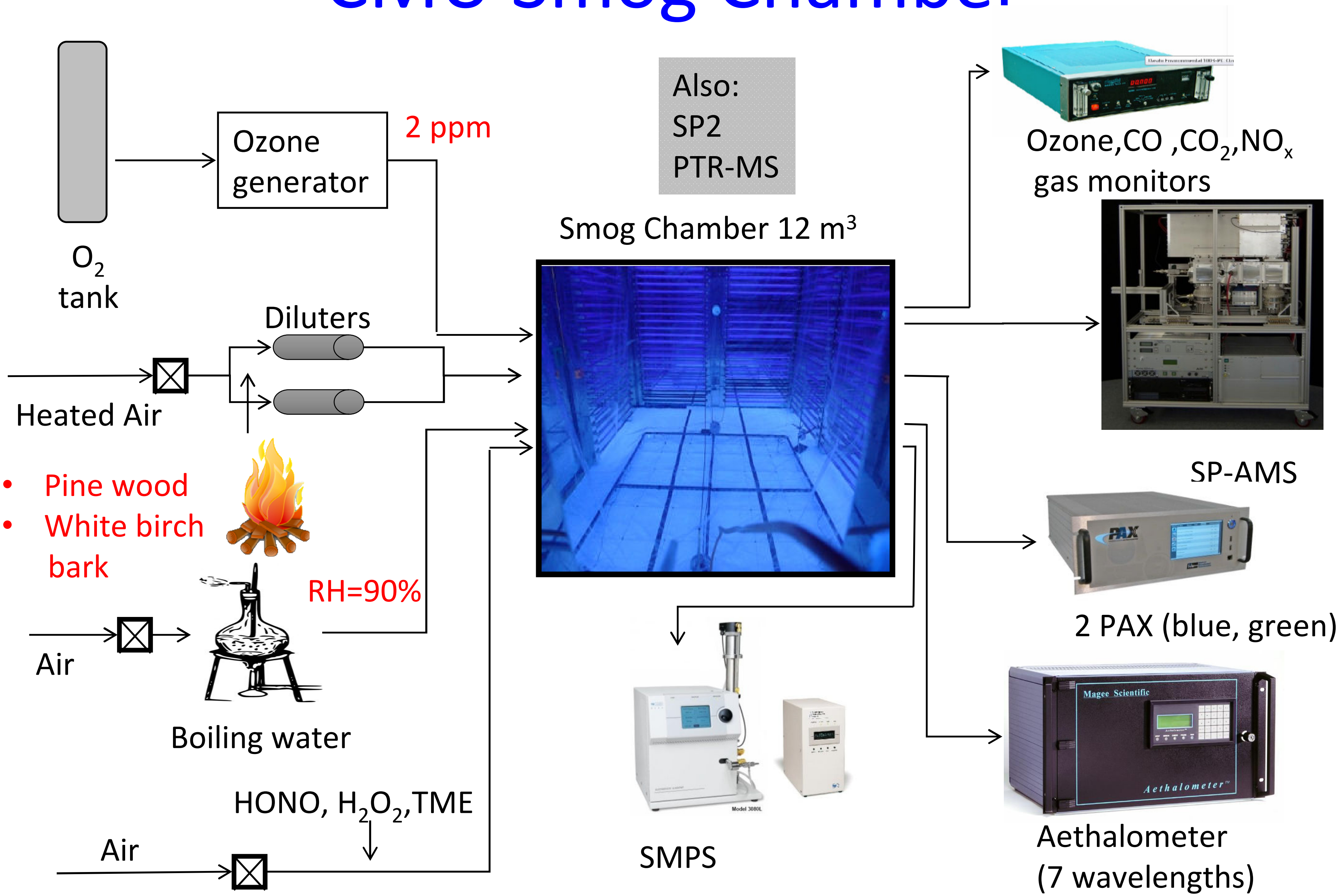
Combustion Emissions

BC particles act as condensation sites for OA



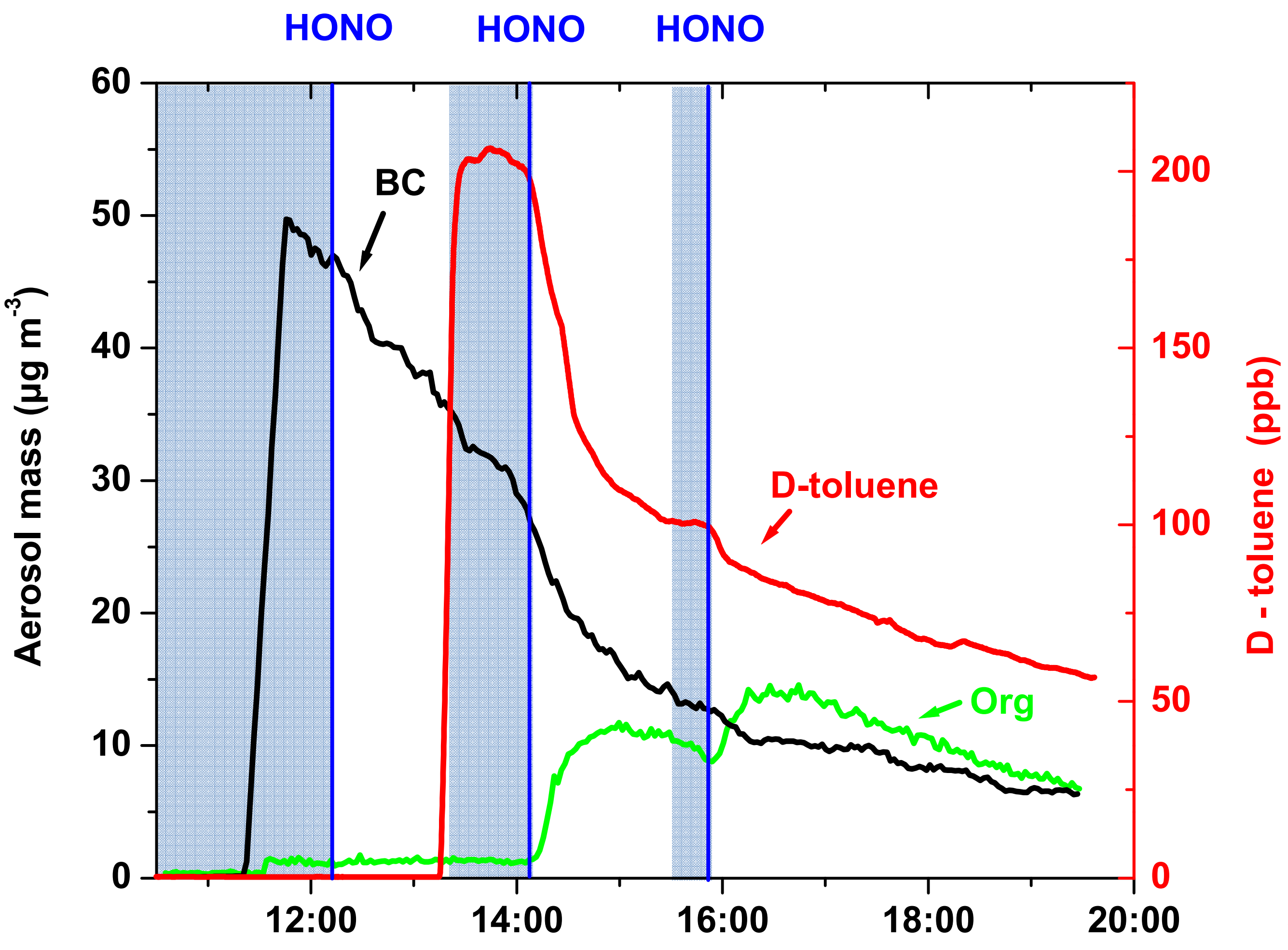
- **Brown Carbon?**
- **How does the condensation and chemical aging of OA affect the absorption of BC?**

CMU Smog Chamber



Coating of BC with D-toluene SOA

(fuel: White birch bark)

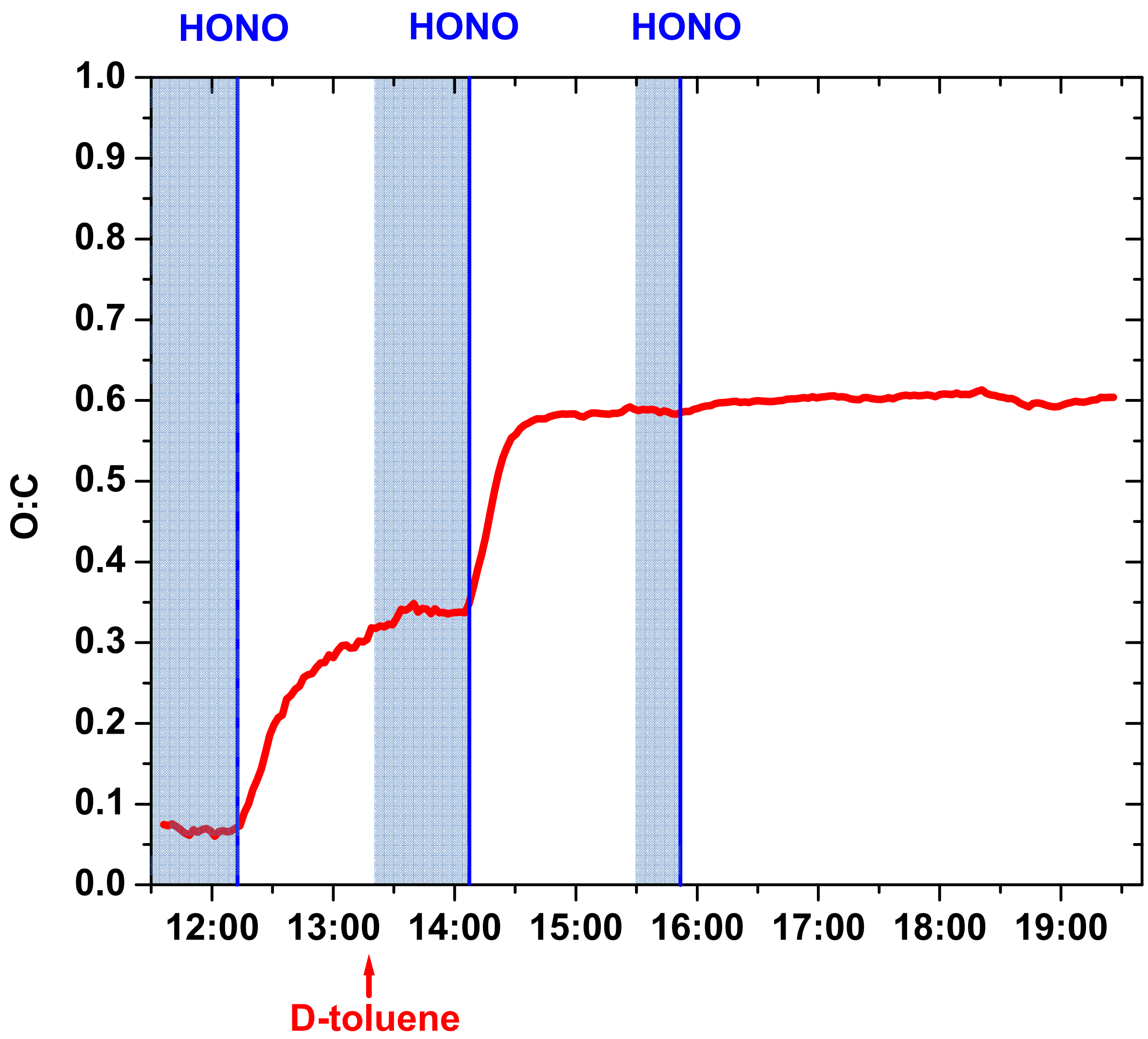


CE_{BC}= 0.4

CE_{org}=1

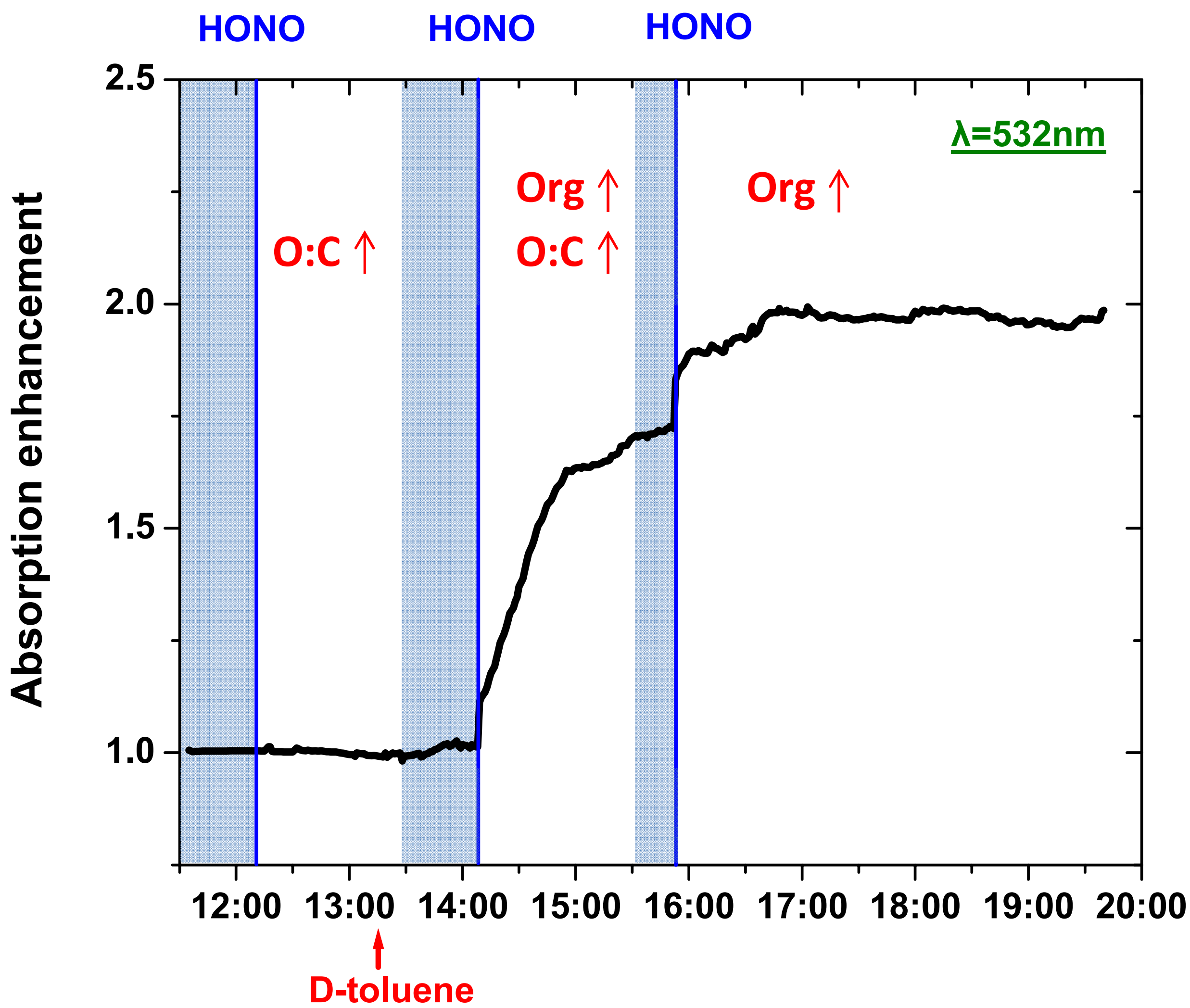
O/C during D-toluene SOA formation

(fuel: White birch bark)

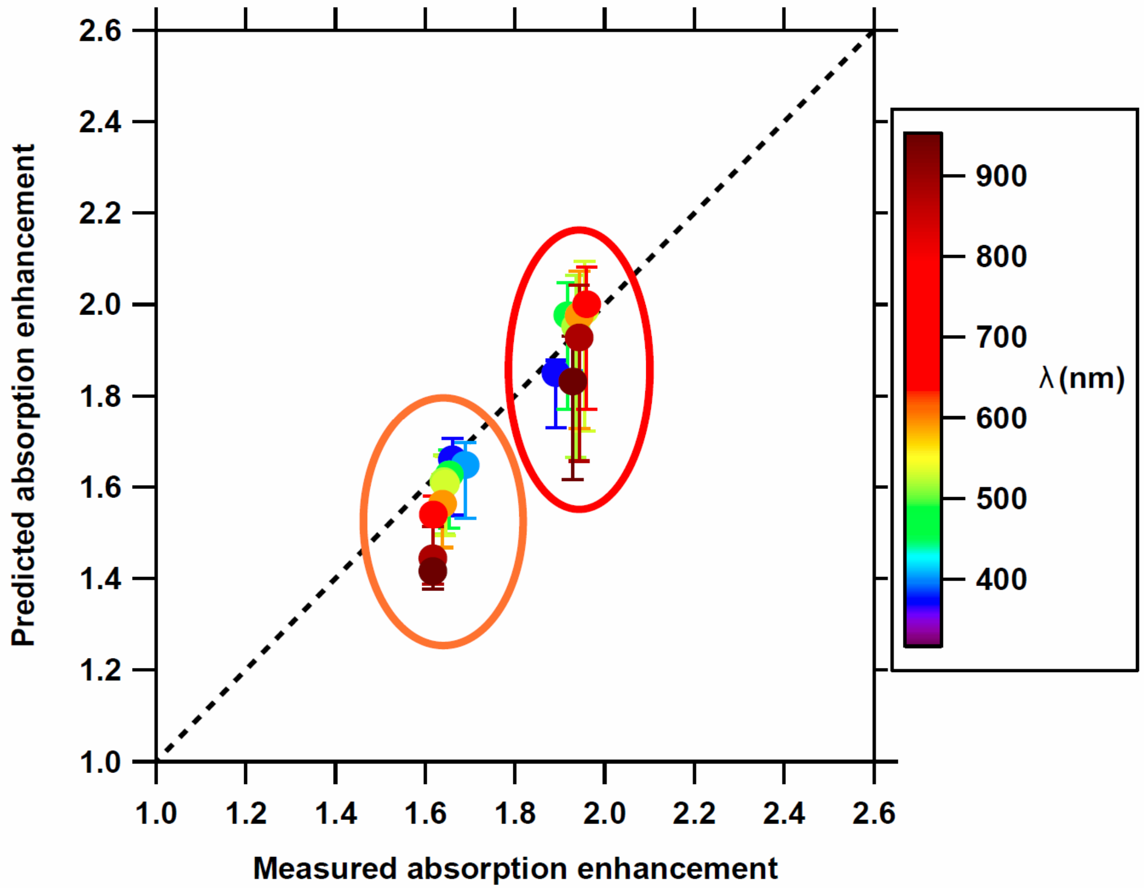


Absorption during D-toluene SOA formation

(fuel: White birch bark)



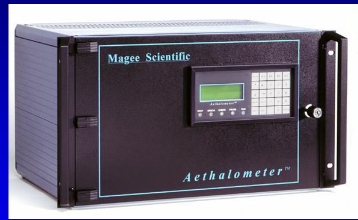
Comparison of Mie theory with measurements



The D-toluene SOA - soot particles have core shell morphology and their absorption is consistent with Mie theory predictions.

OA and BC Formation and Aging (FLAME III and IV)

Aethalometer



OA/BC from biomass burning

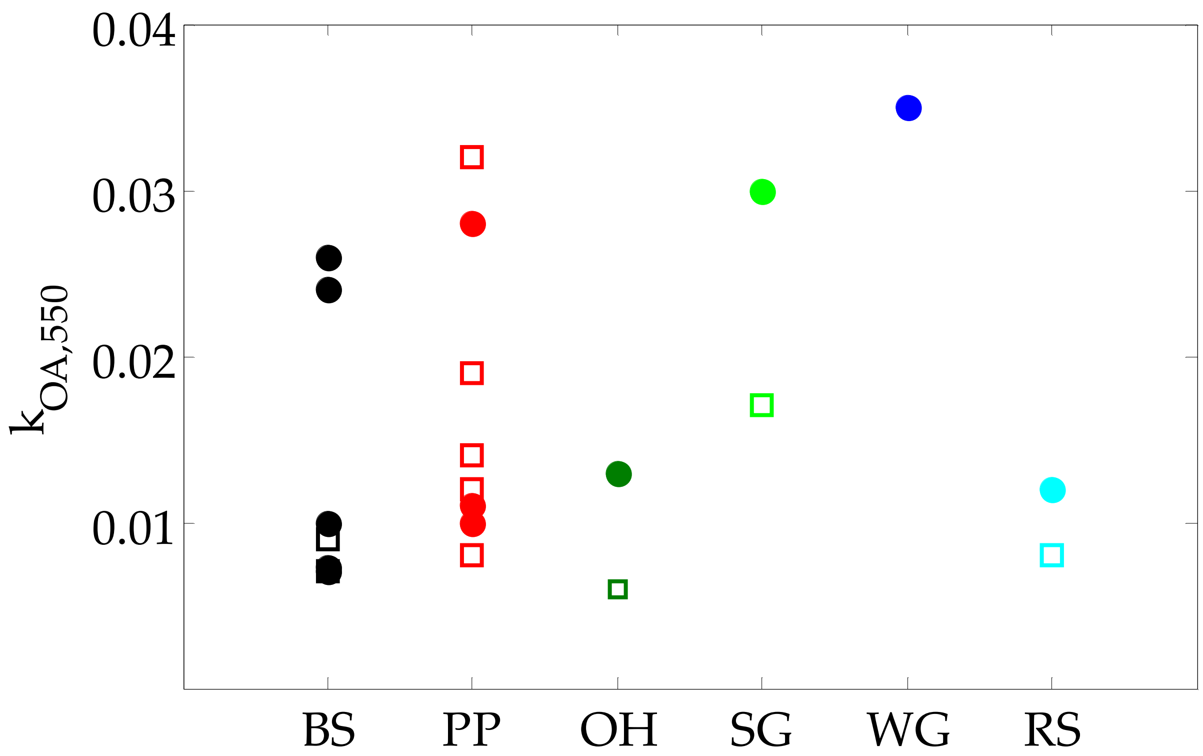
SP2

PASS-3

HR-SP-AMS

OA Absorption: The Chaos Returns !

R. Saleh

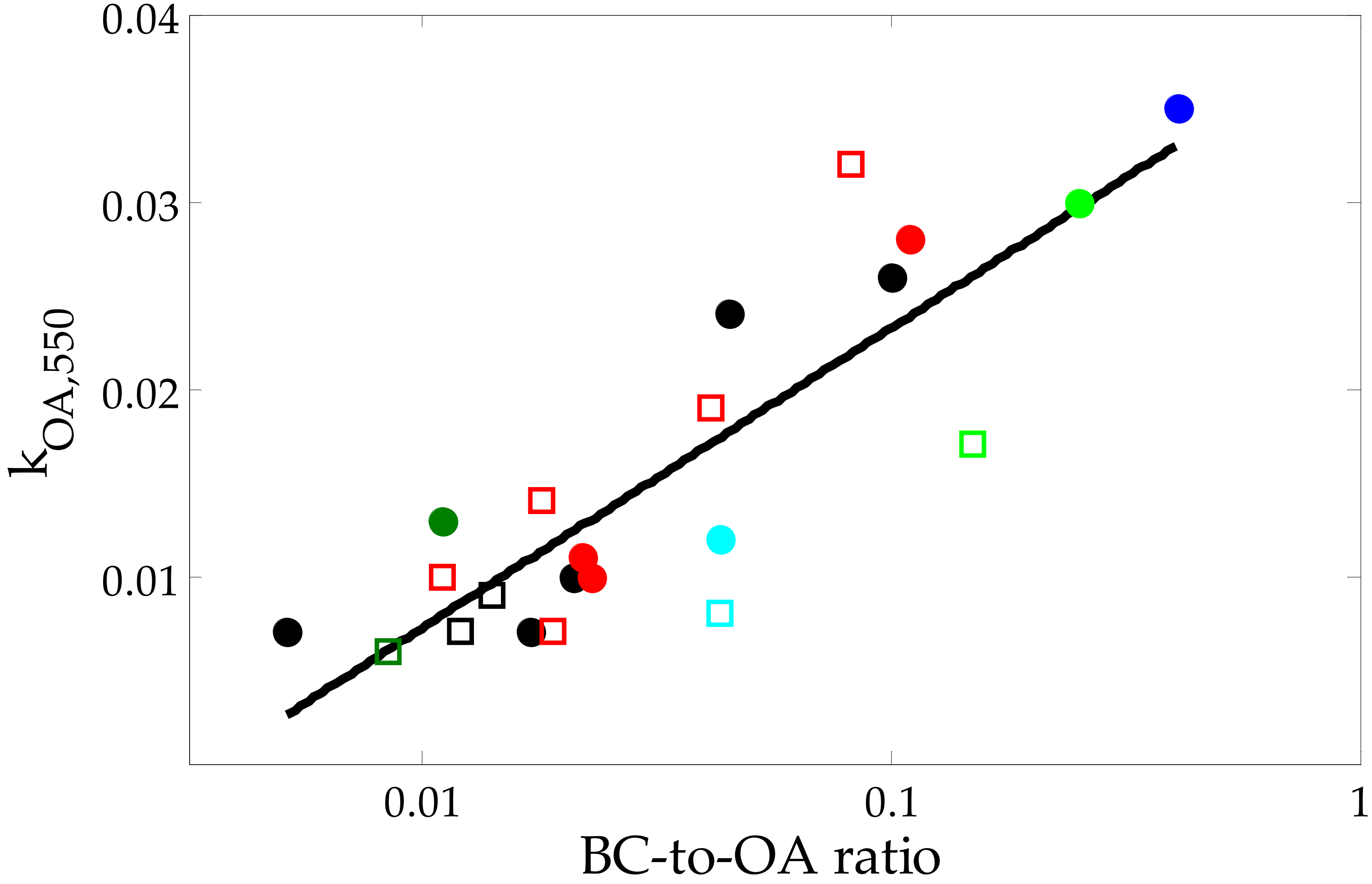


FLAME IV

BS: black spruce
PP: ponderosa pine
OH: organic hay
SG: saw grass
WG: wire grass
RS: rice straw
Closed symbols: fresh
Open symbols: aged

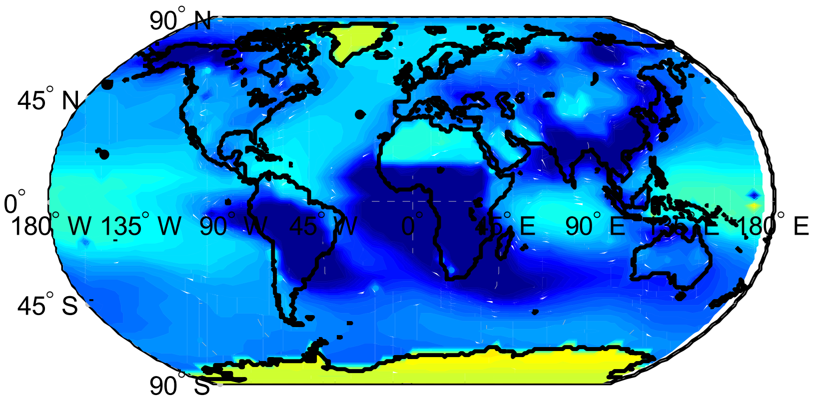
- A lot of variability across fuels, and even within the same fuel.
- Similar to previous work.

Some Order: The Role of BC/OA

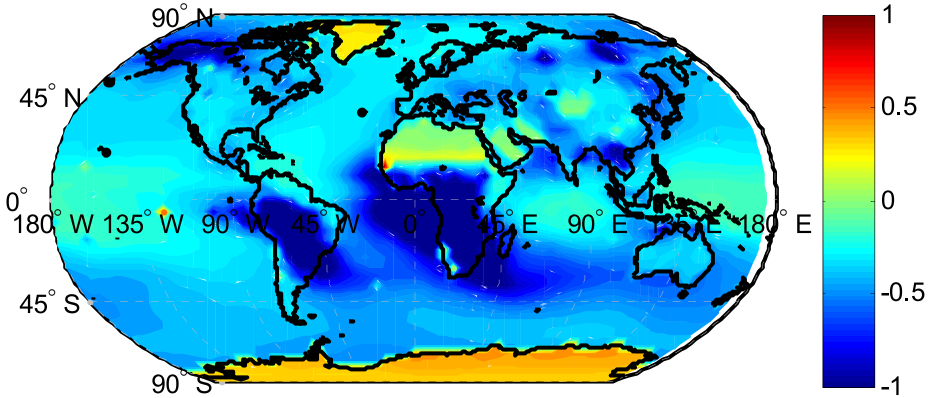


Direct Radiative Effect of Biomass Burning Emissions

non-absorbing OA | **absorbing OA**



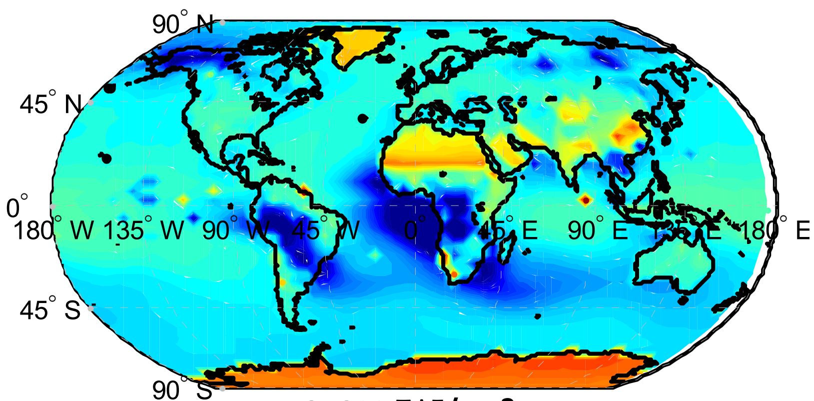
-0.64 W/m²



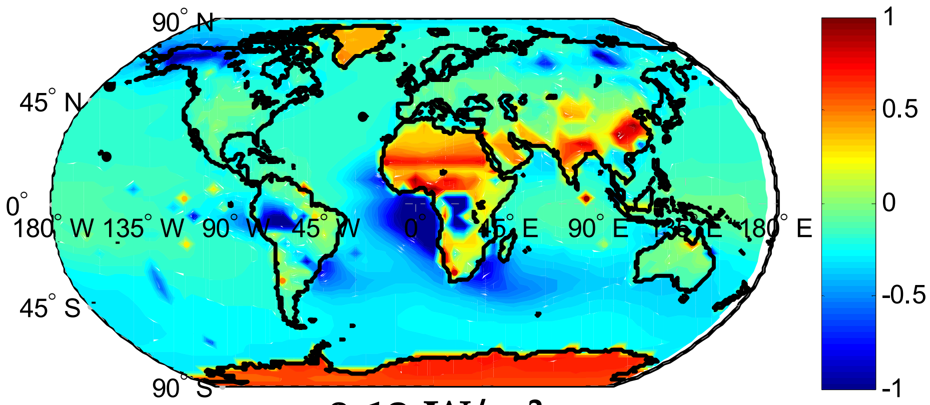
-0.43 W/m²

externally-mixed

internally-mixed



-0.25 W/m²



-0.12 W/m²

BC Measurement

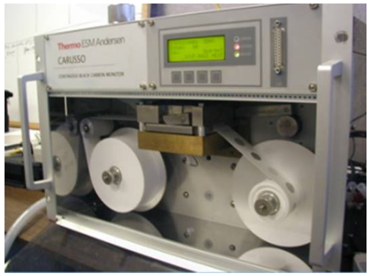
BC related instruments

Optical instruments

$$b_{abs} \xrightarrow{MAC} eBC$$

1) Aethalometer

2) Multiangle Absorption Photometer (MAAP)



3) Photoextinciometer -PAX

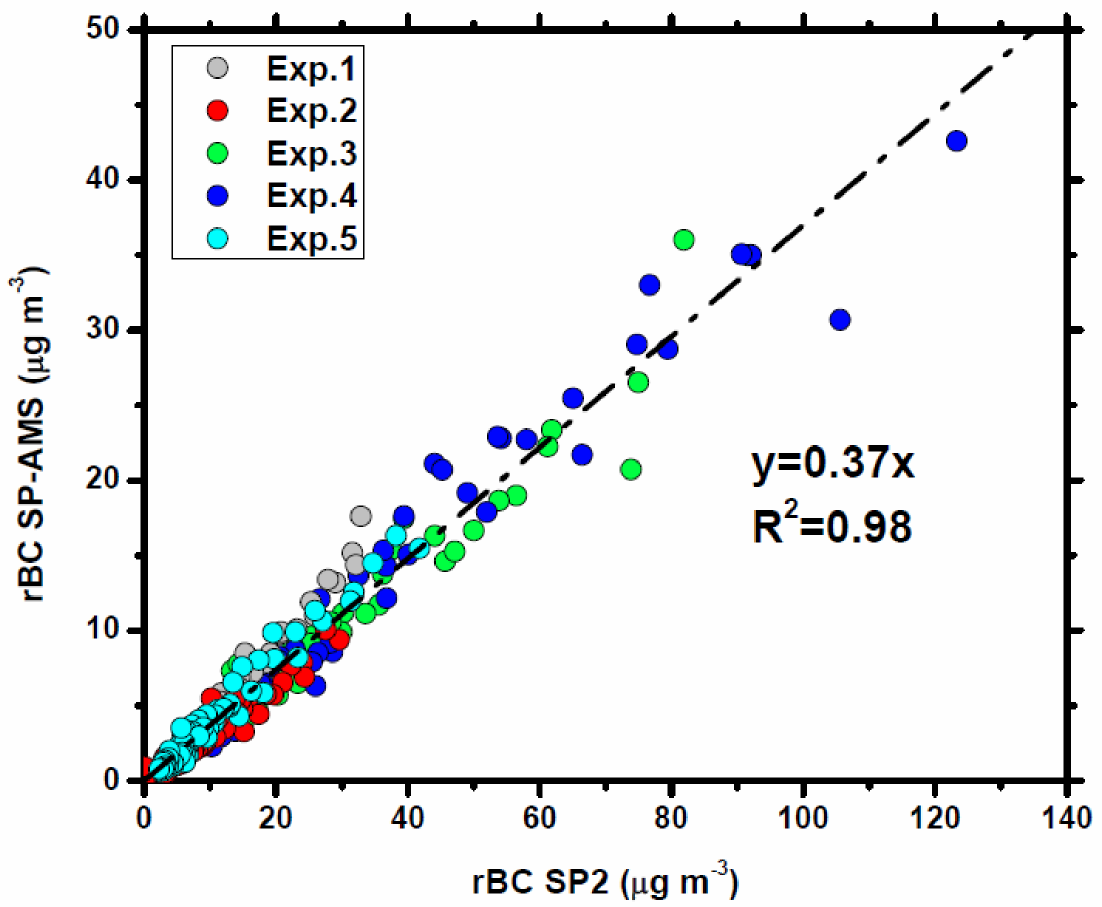
Instruments using incandescence methods (rBC)

1) Single Particle Soot Photometer(SP2)

2) Soot Particle Aerosol
Mass Spectrometer(SP-AMS)

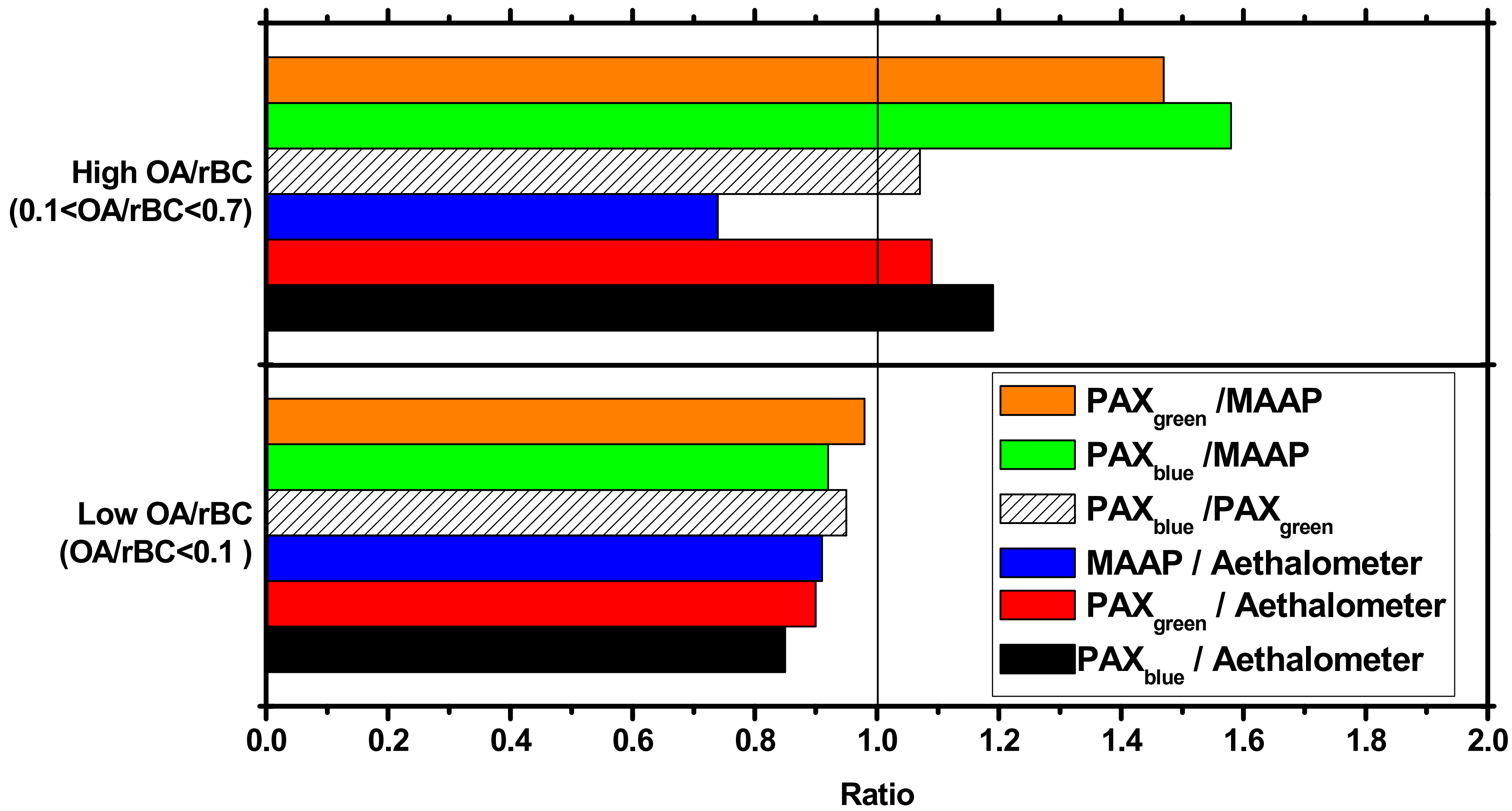
Comparison of rBC measurements

(OA/rBC =0.02 - 0.33)



- **CE≈Es=0.37**

Intercomparison of optical instruments

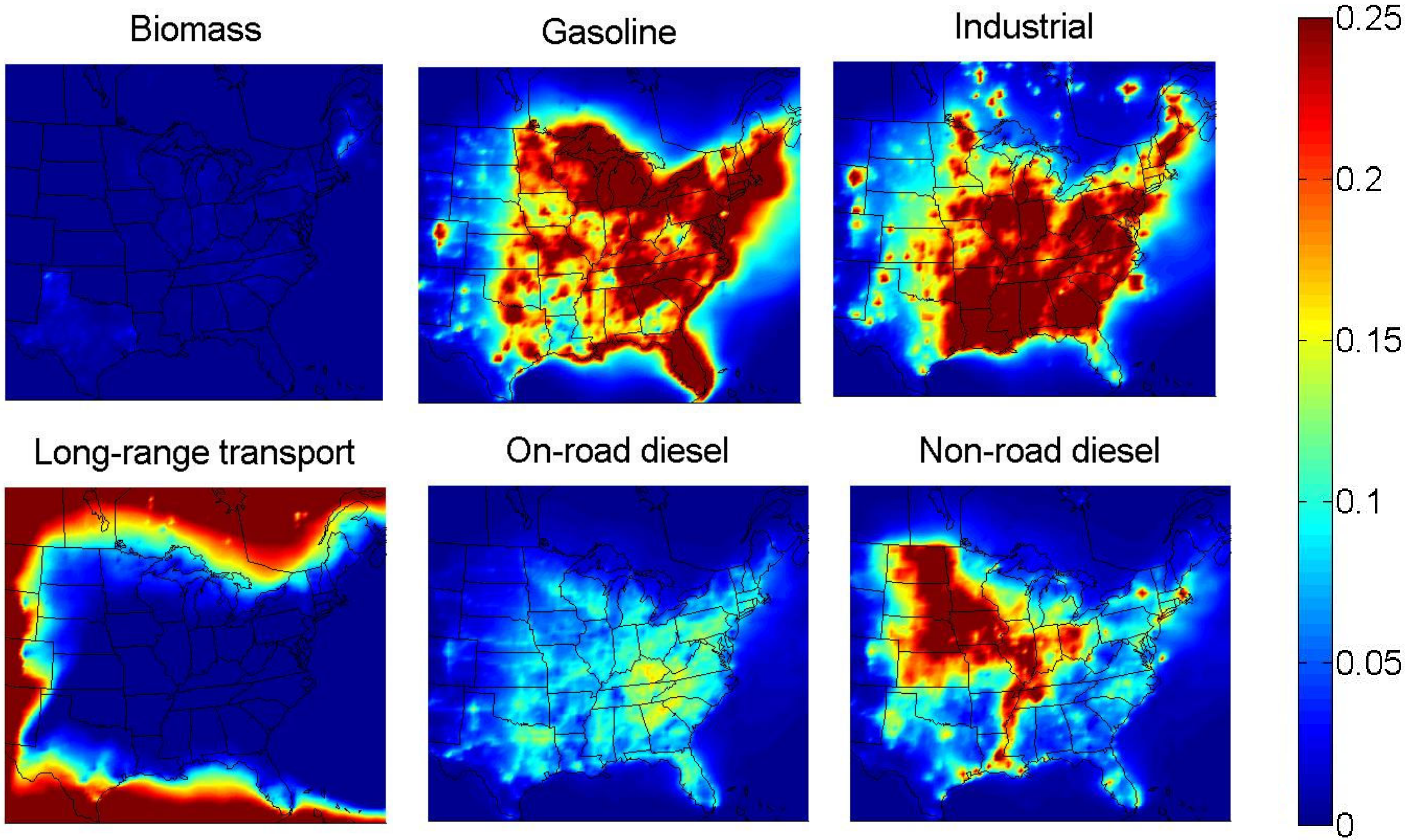


For low OA/rBC: 5%-15% discrepancies

For high OA/rBC: 20%-60% discrepancies

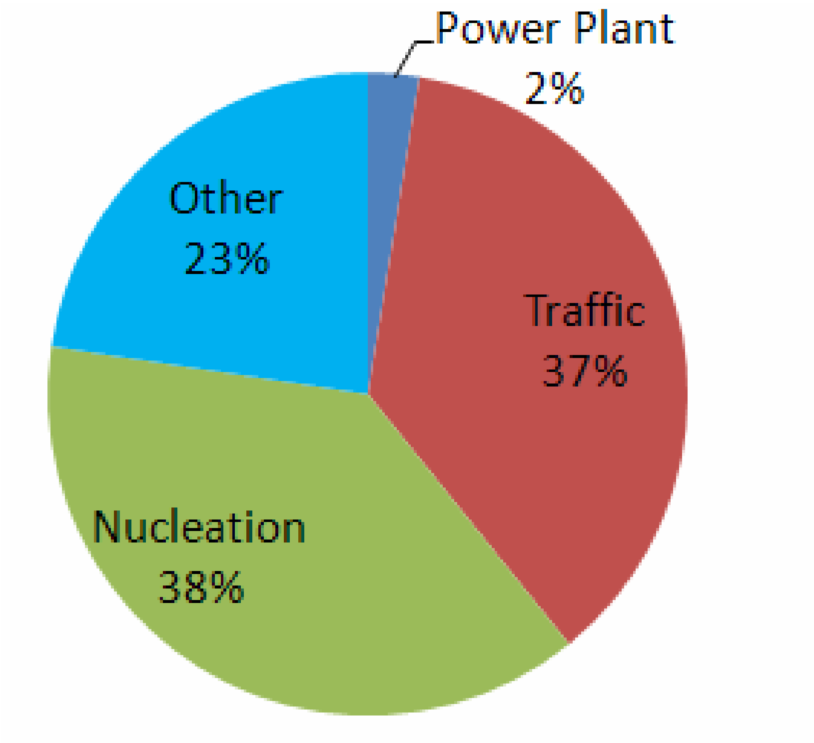
BC and Aerosol Number Concentrations

Total primary particle number fractional source contributions



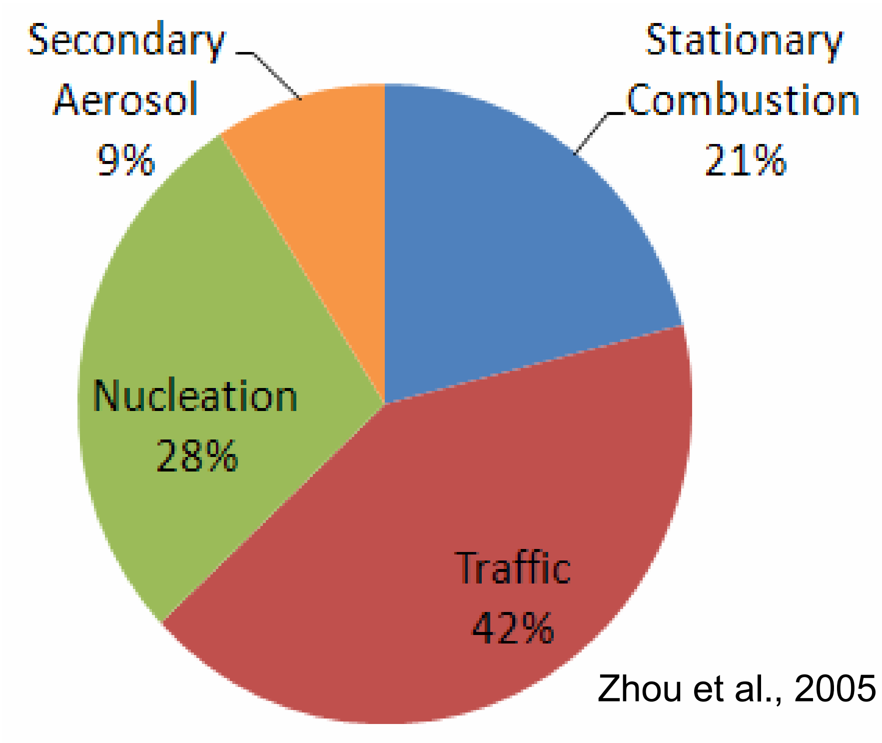
Sources of Measureable (>3 nm) Particle Number in Pittsburgh

PMCAMx



Predicted: 29,000 cm^{-3}

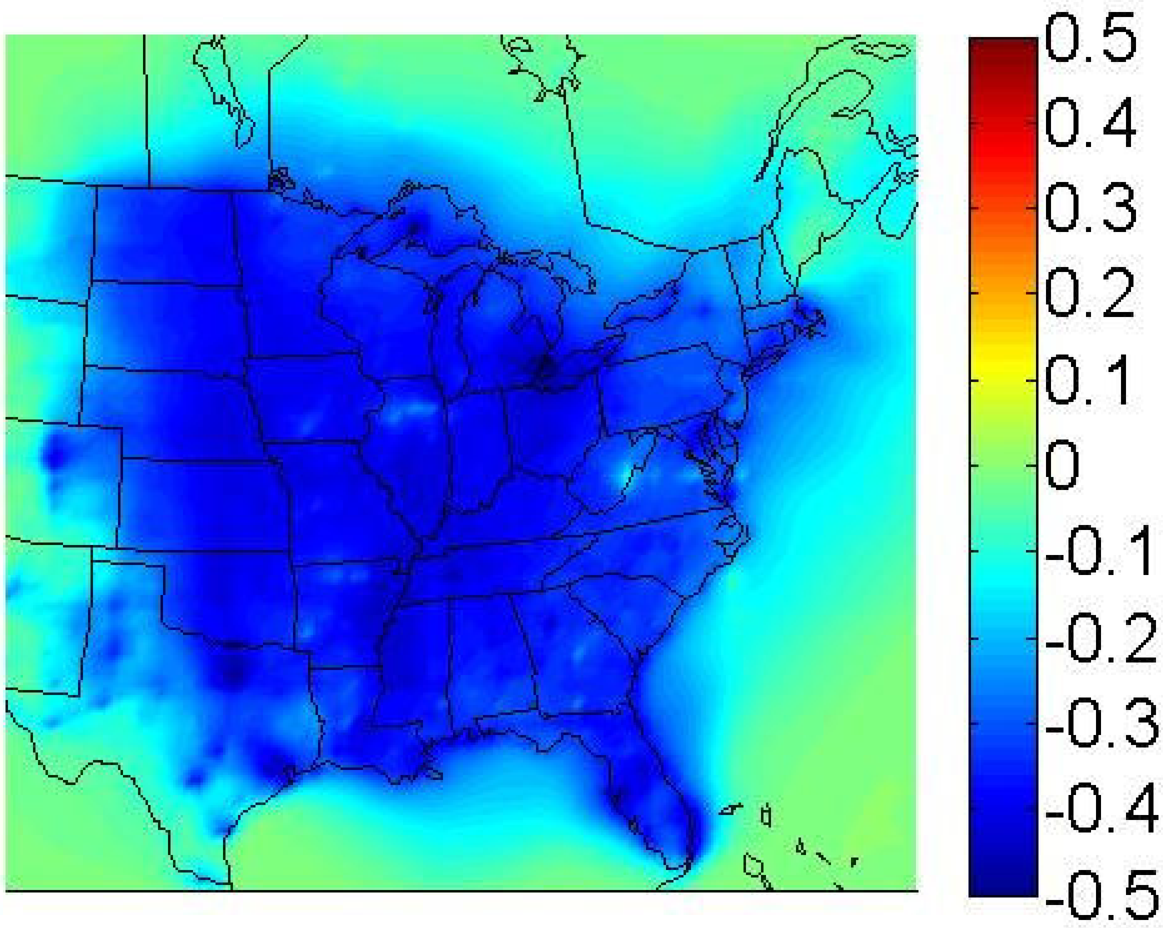
Calculated from Measurements



“Measured”: 26,000 cm^{-3}

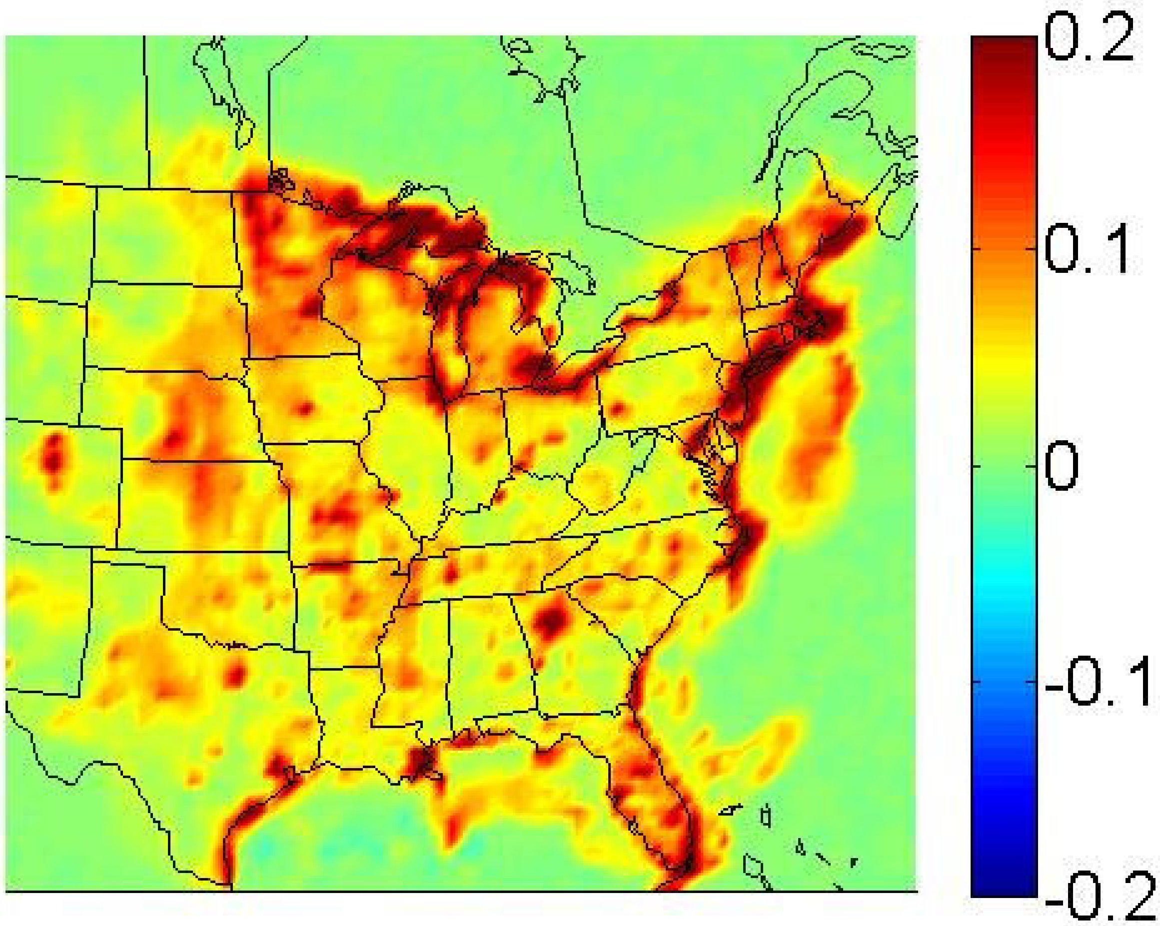
Effects of Controls of Diesel Particulate Emissions (-50% Scenario)

Fractional Change of EC



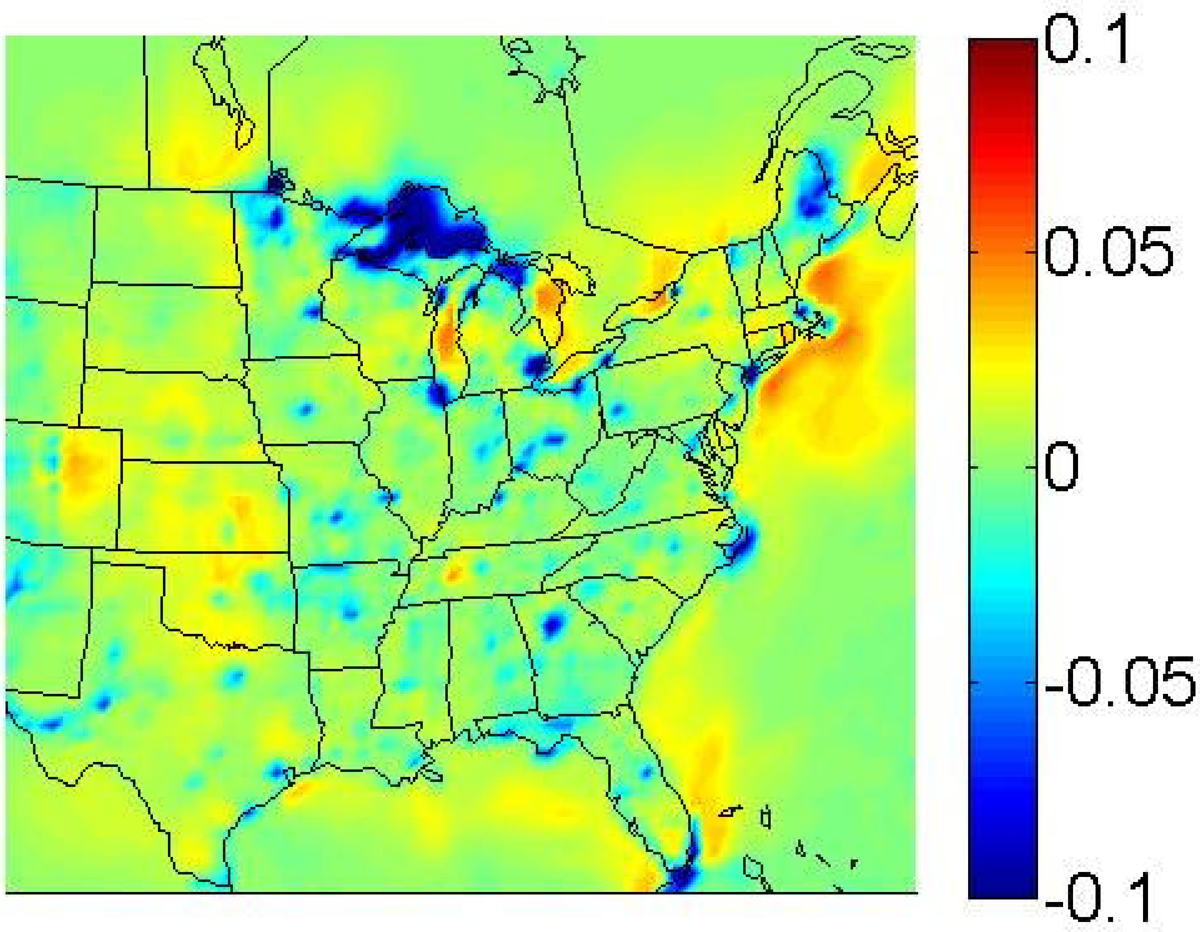
Average $PM_{2.5}$ reduction around 3%.

Fractional Changes of N_{1-10}



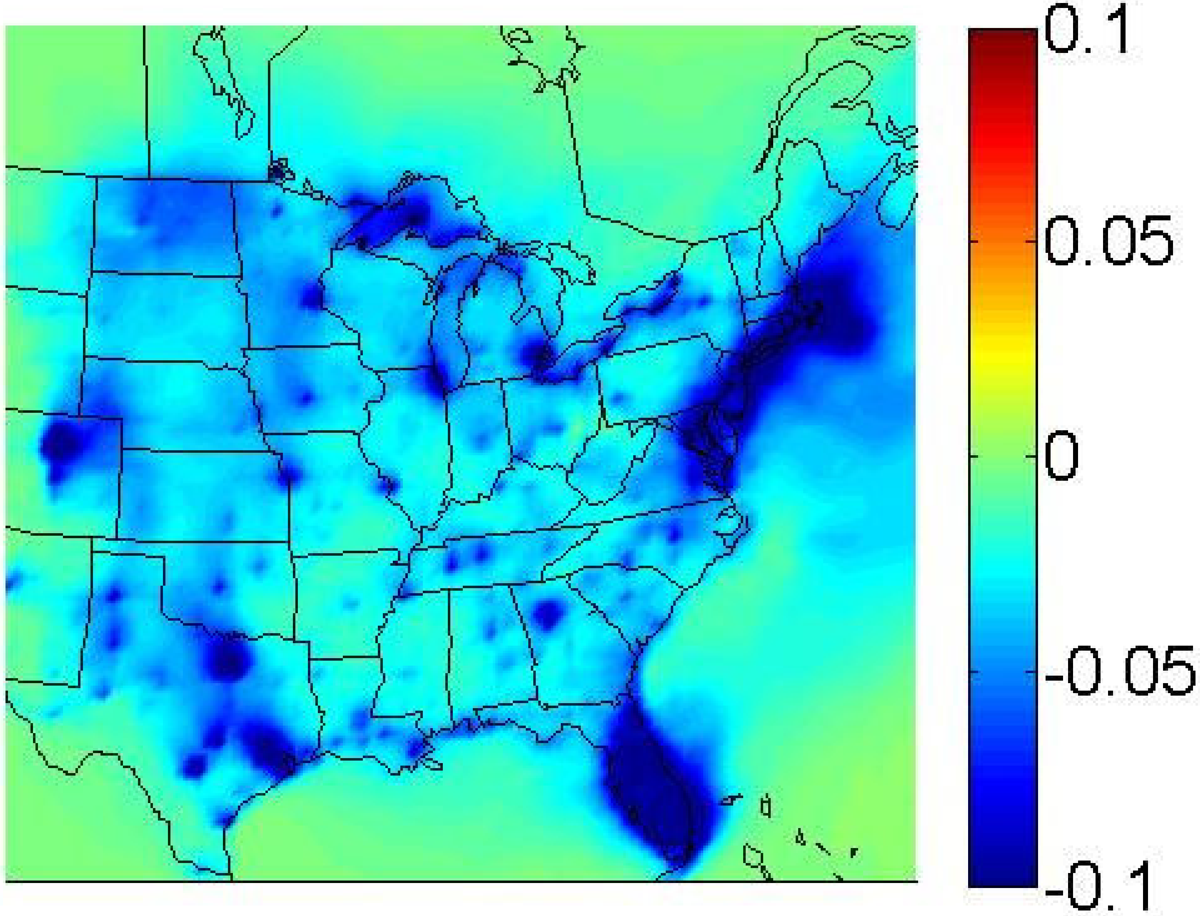
Nucleation increases, creating more smaller particles due to the decrease in the condensation sink.

Fractional Changes of N_{10-50}



Particles in this size range are typically emitted or grown from nucleated particles, so they see increases (from nucleation) and decreases elsewhere.

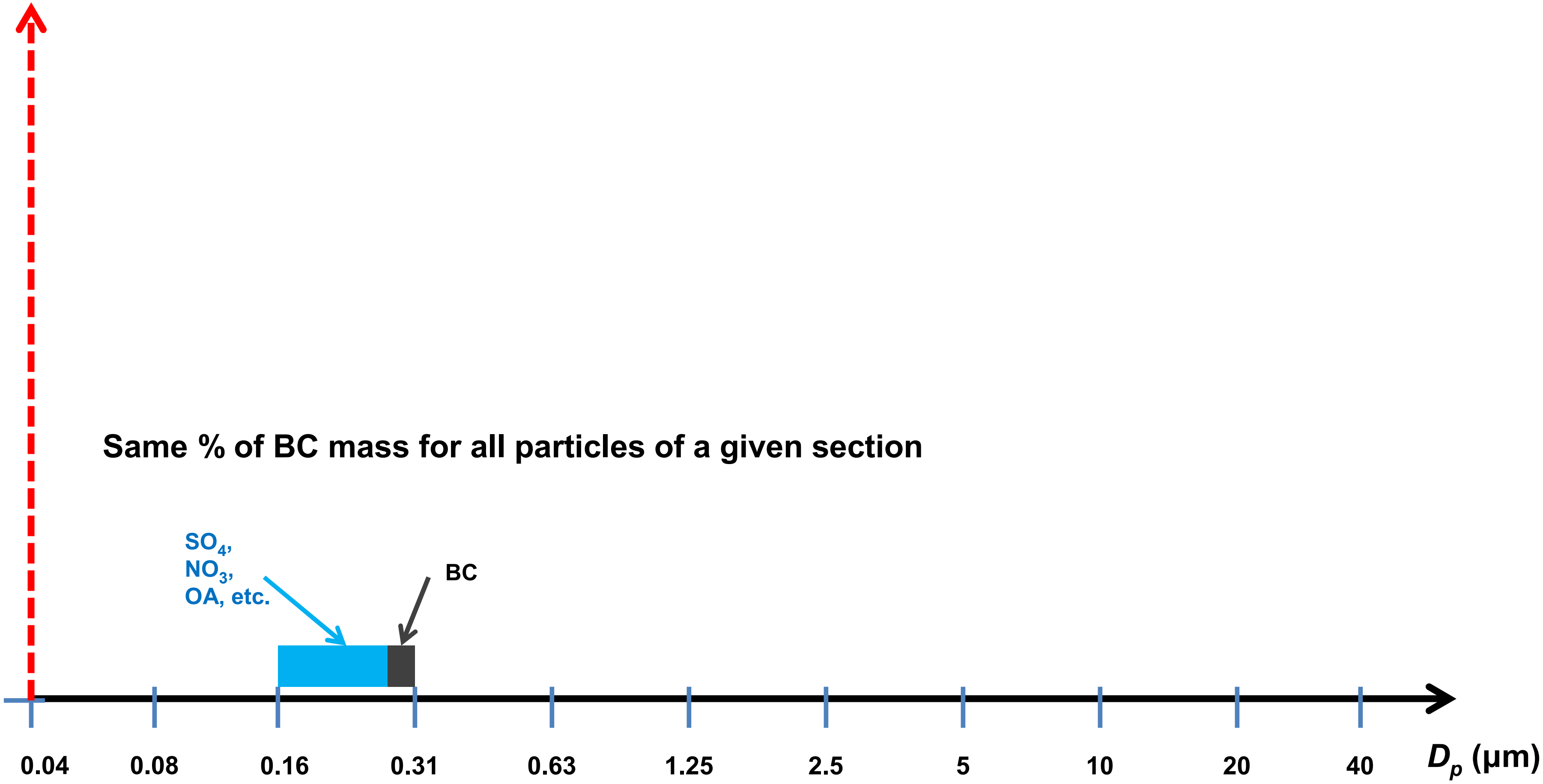
Fractional changes of N_{100}



Improving Regional Scale BC Models

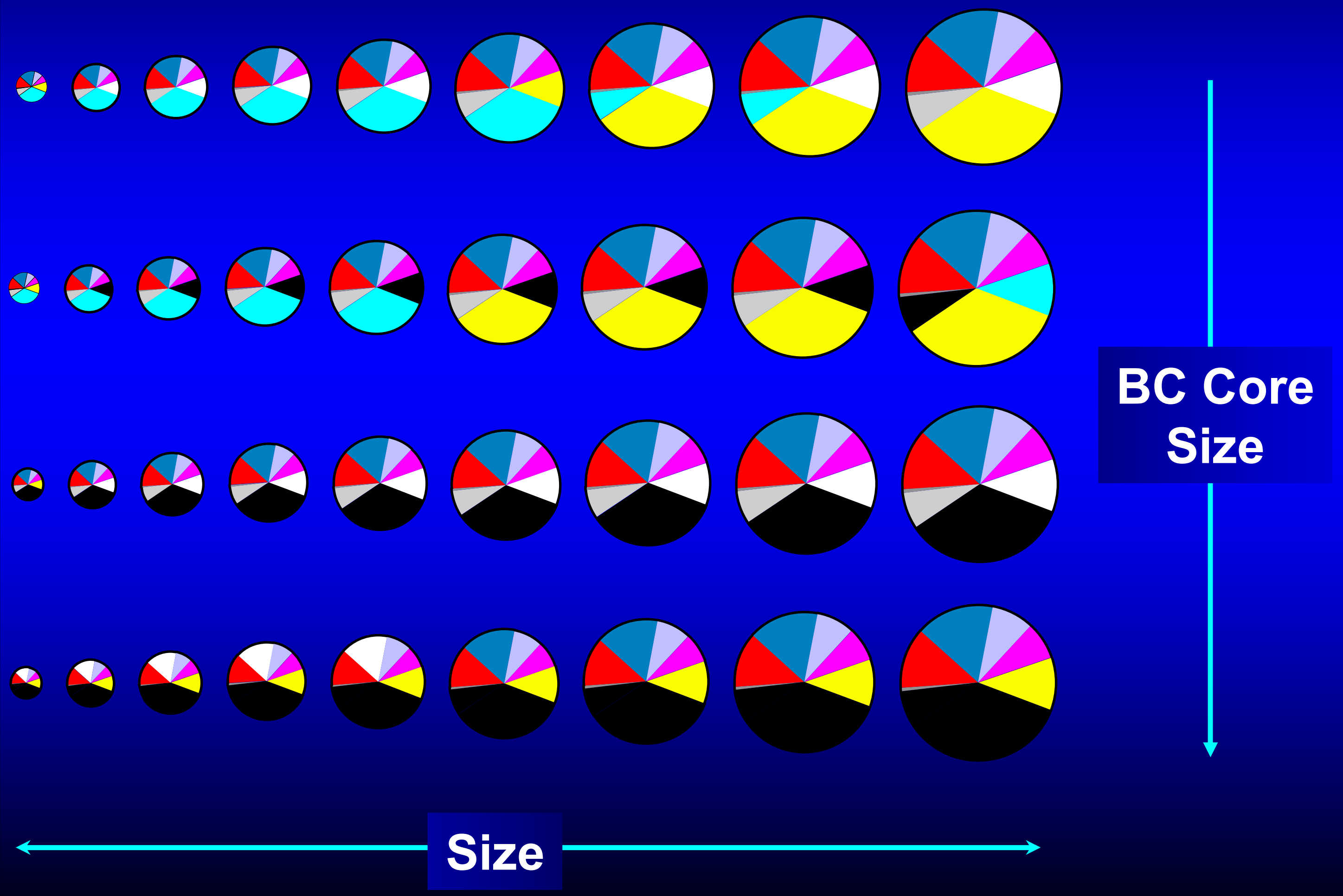
PMCAMx

Current approach: 1 particle distribution-Internal Mixing



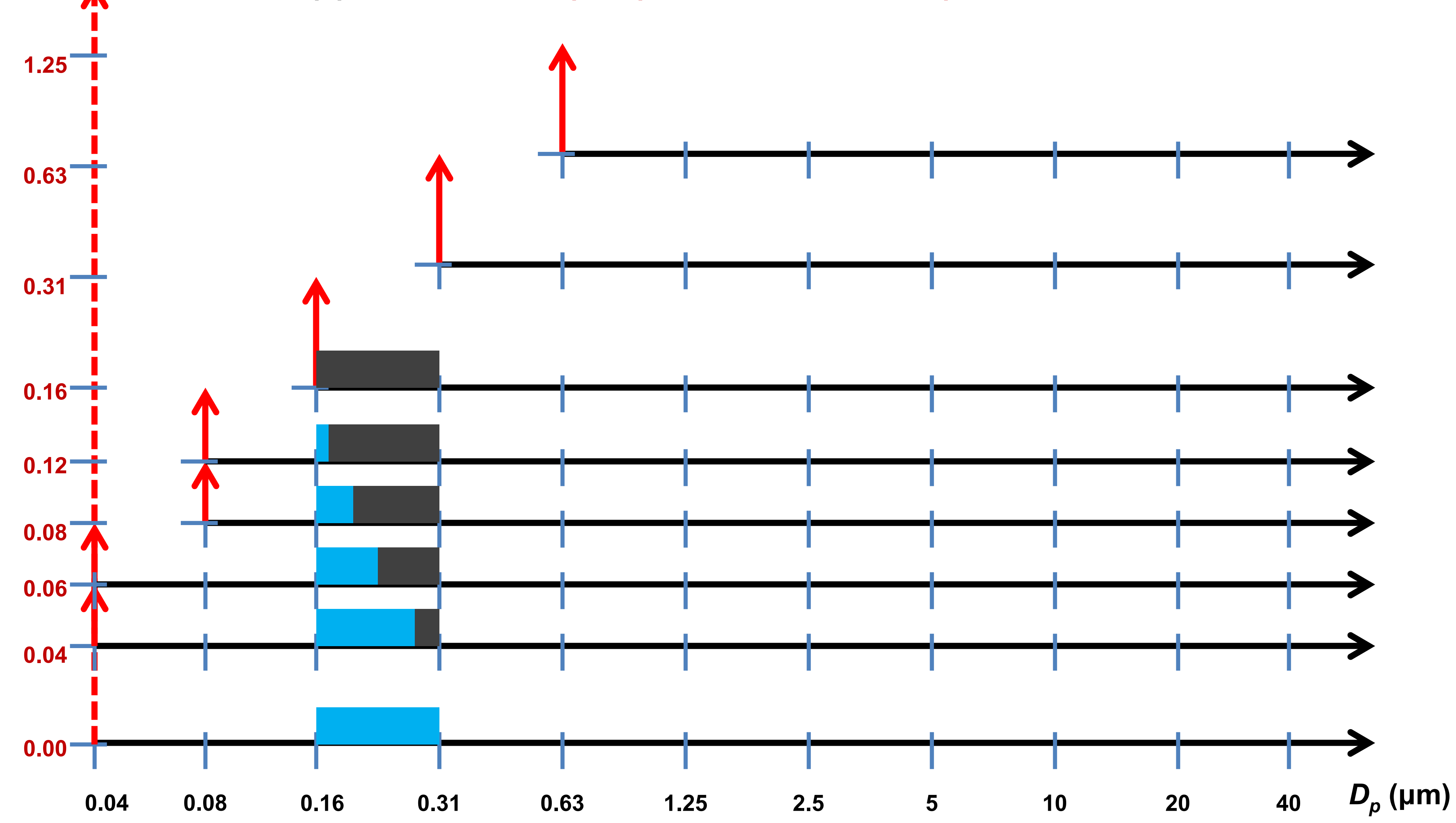
10 size sections : from 40 nm up to 40 μm

Simulating BC Mixing State In PMCAMx



PMCAMx

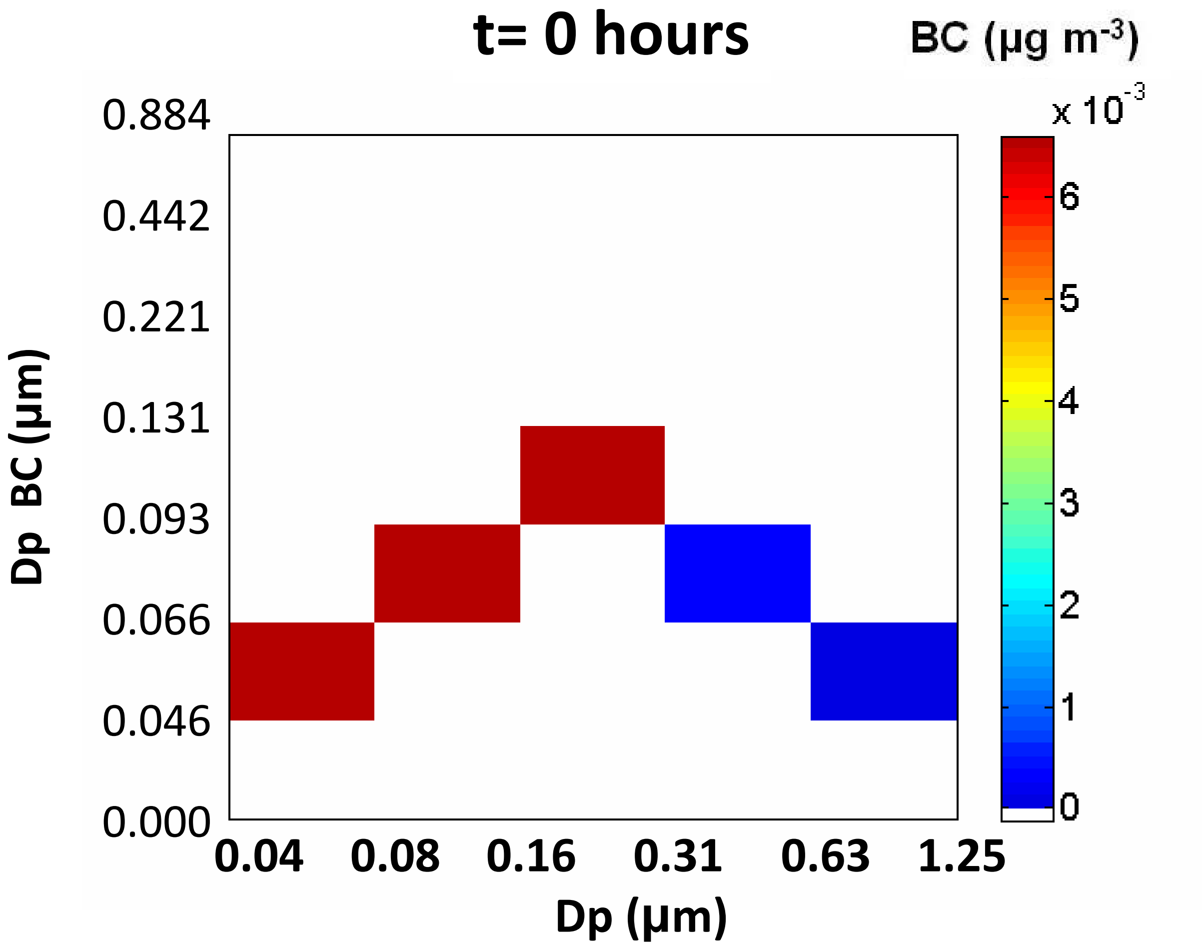
New approach: Multiple particle size/composition distributions



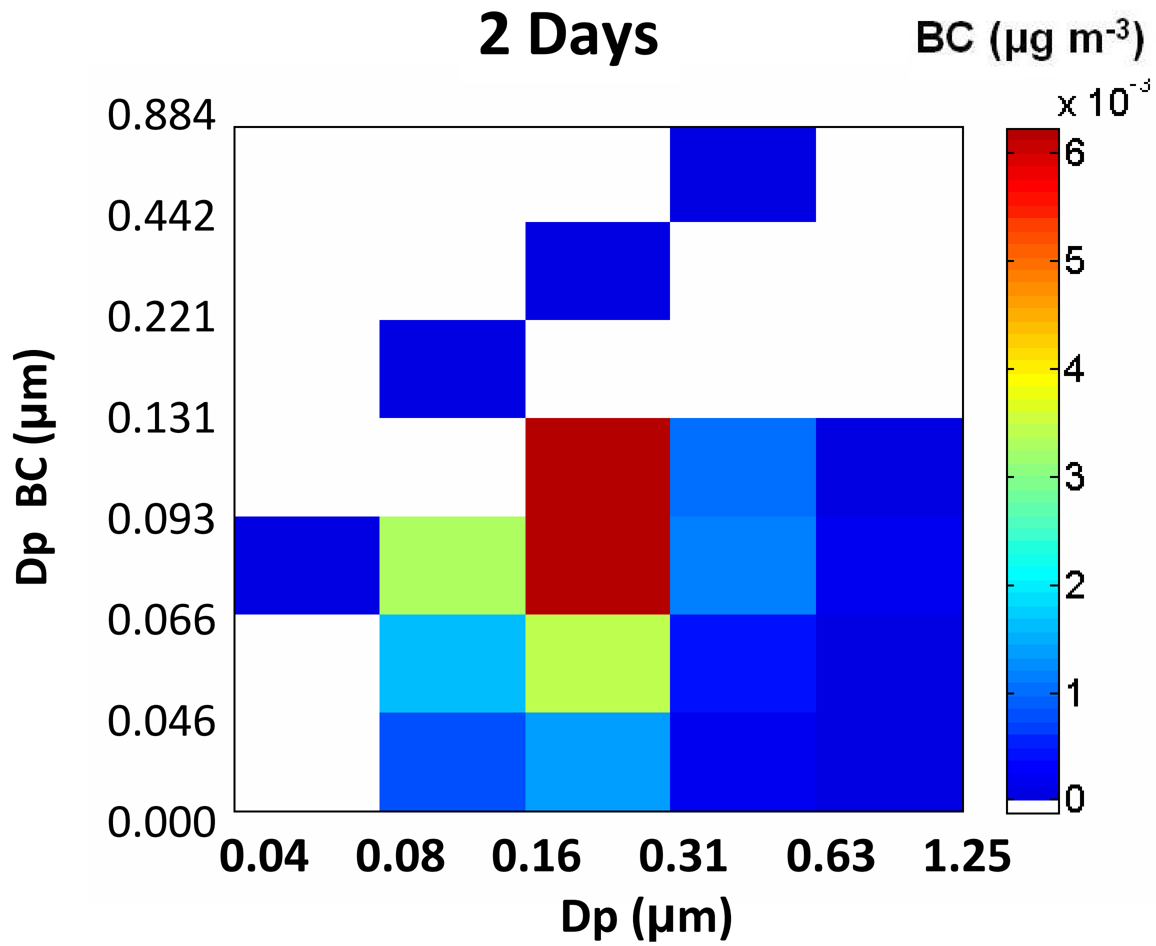
Based on the core diameter of BC (D_{BC})

Variable % of BC mass within particles of a given section !

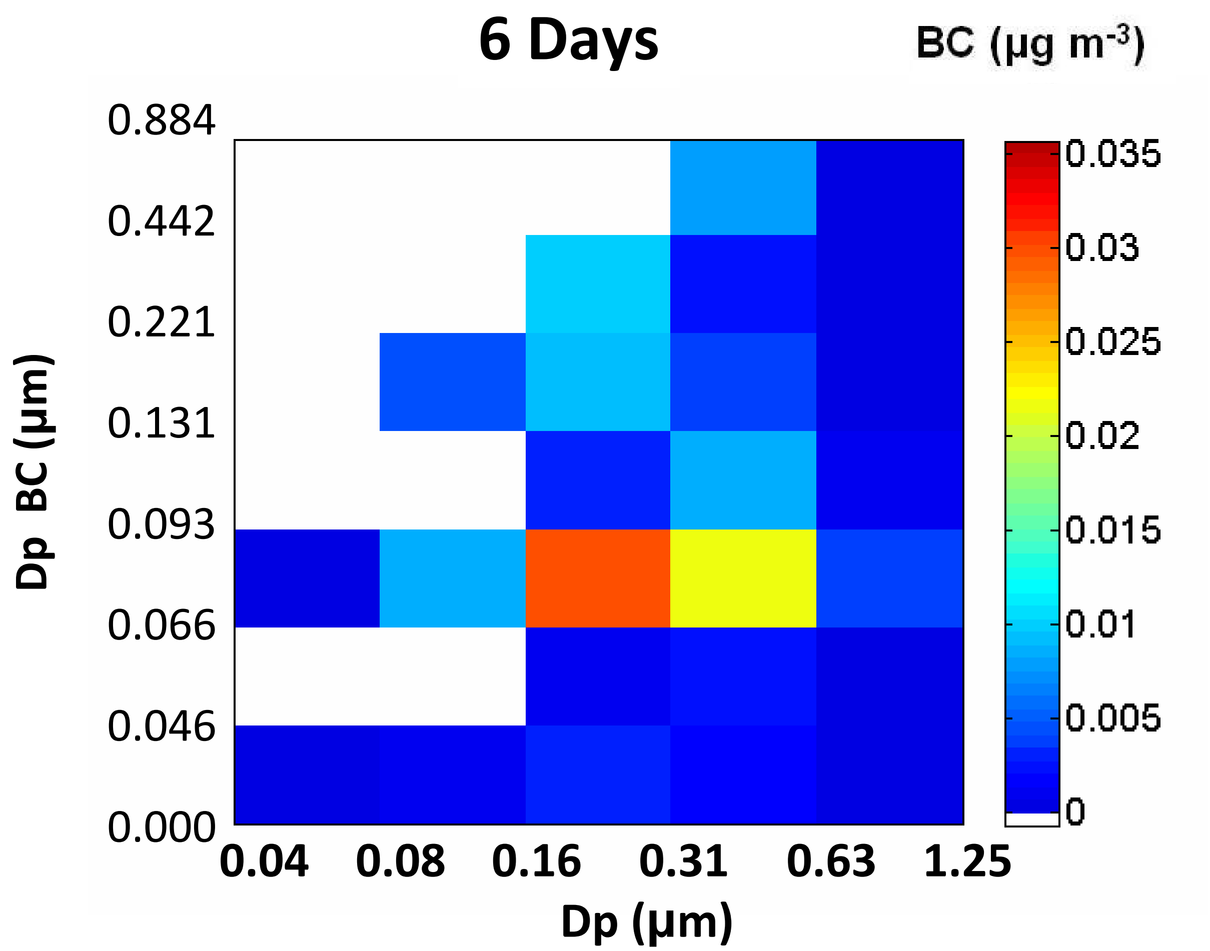
Predicted BC Distribution



Predicted BC Distribution



Predicted BC distribution

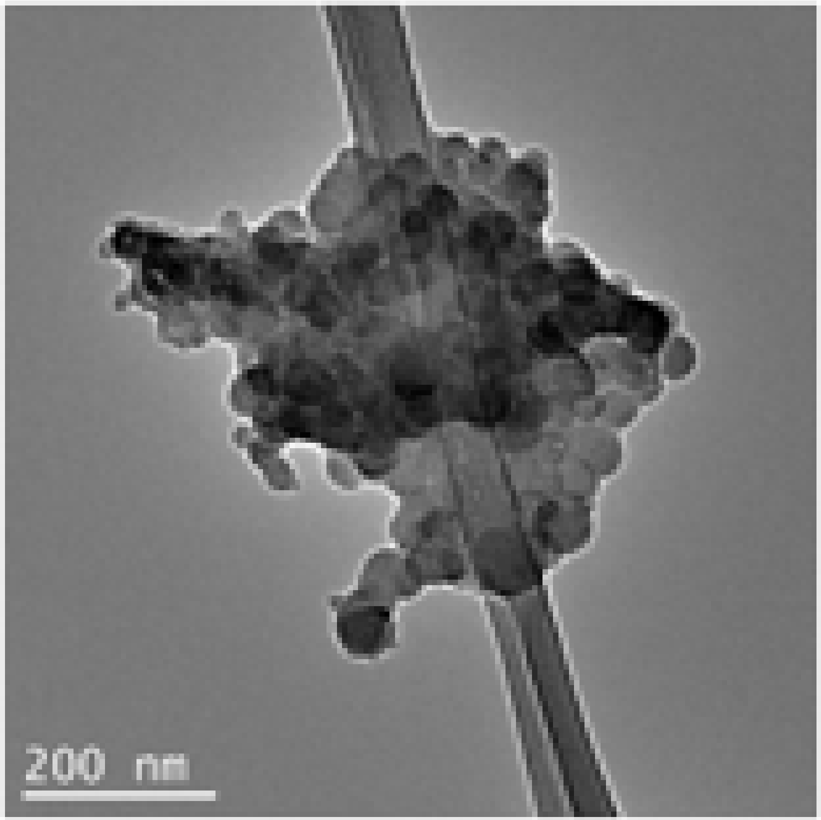


BC particle shape

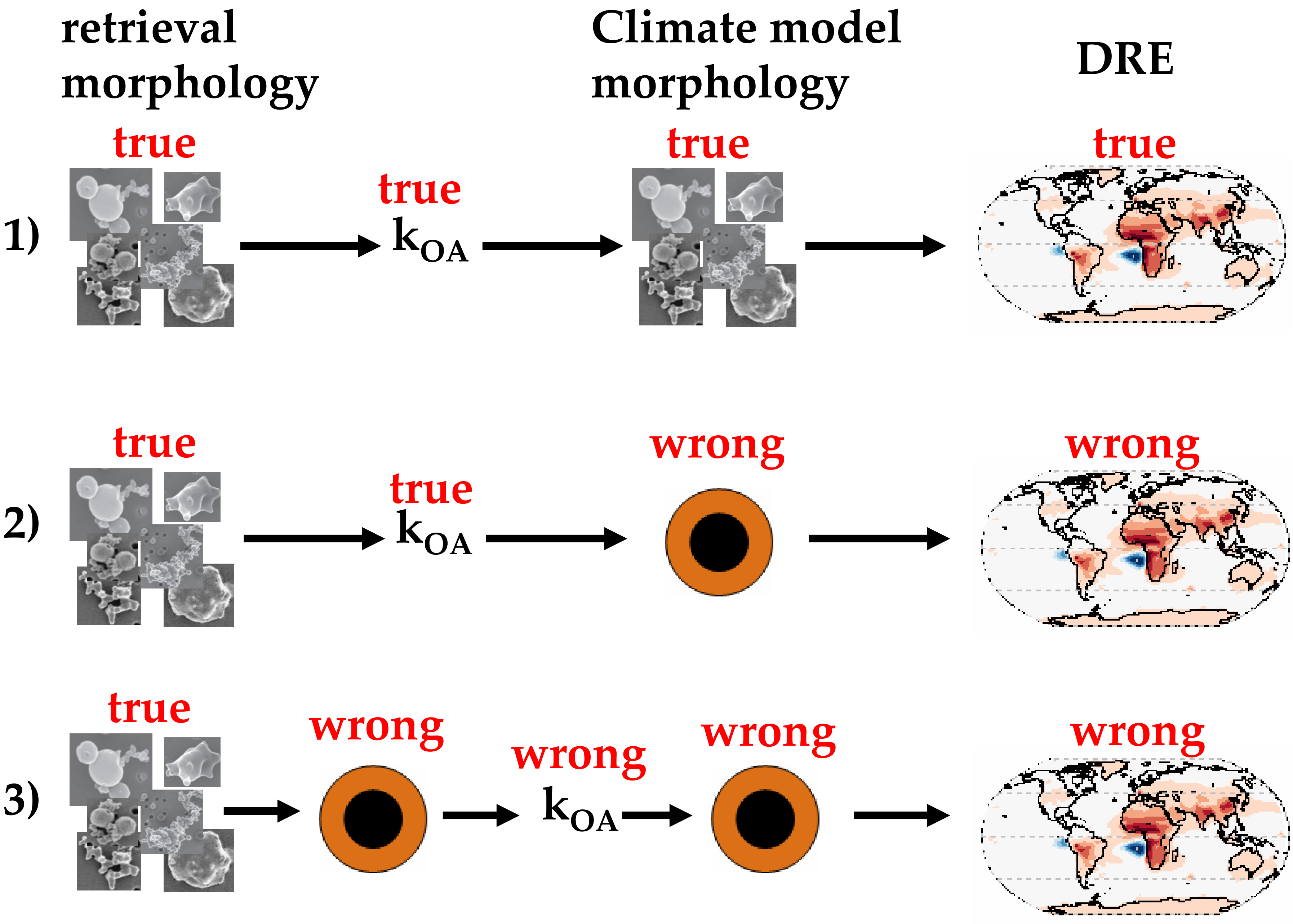
Do two wrongs make a right?

TEM photos of coated particles

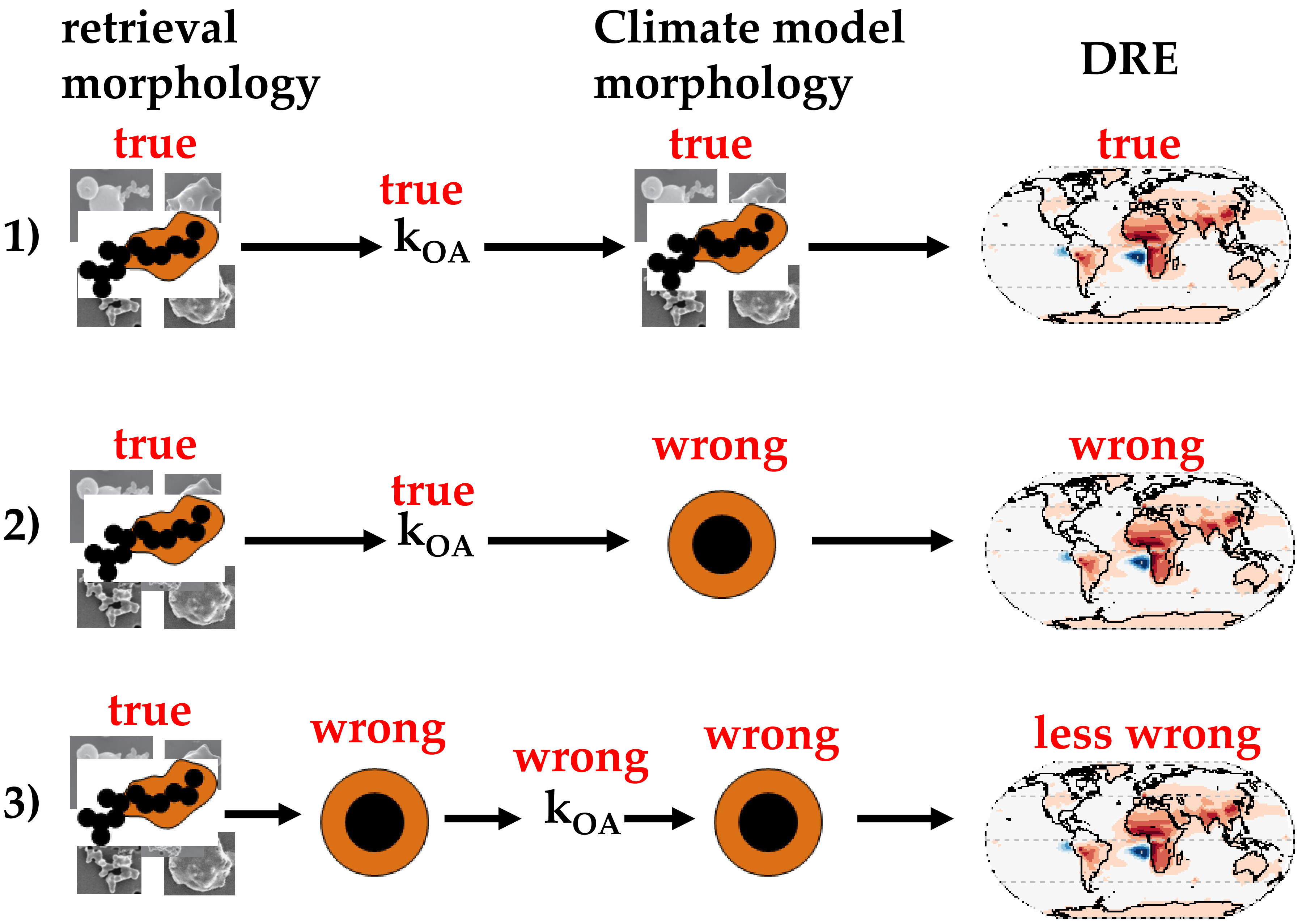
Typical BC particle after coating with secondary organic compounds.



Saliba et al., in prep.



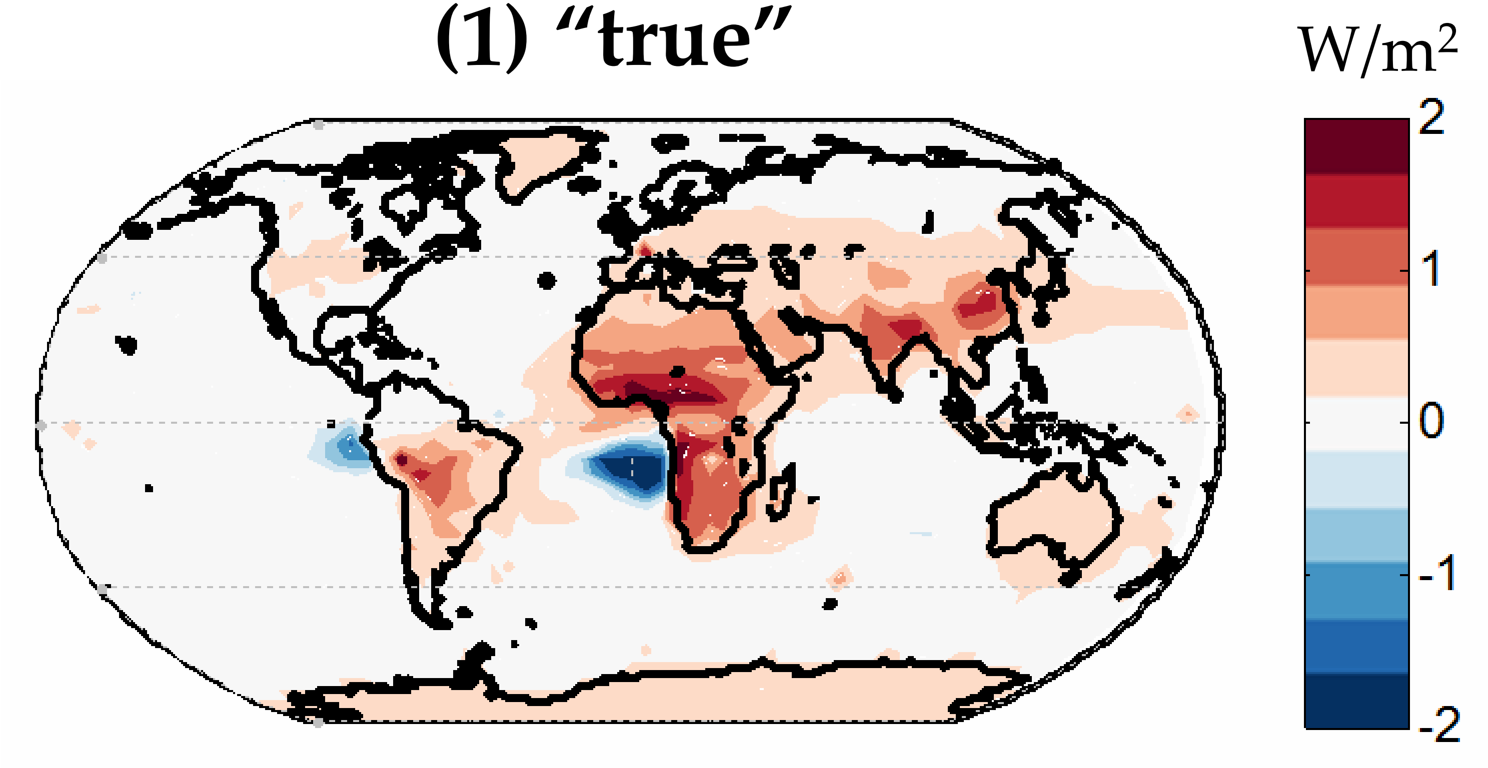
retrieval morphology
Climate model morphology
DRE
1)
true
true
k_{OA}
true
true
2)
true
true
k_{OA}
wrong
wrong
3)
true
wrong
wrong
k_{OA}
wrong
wrong



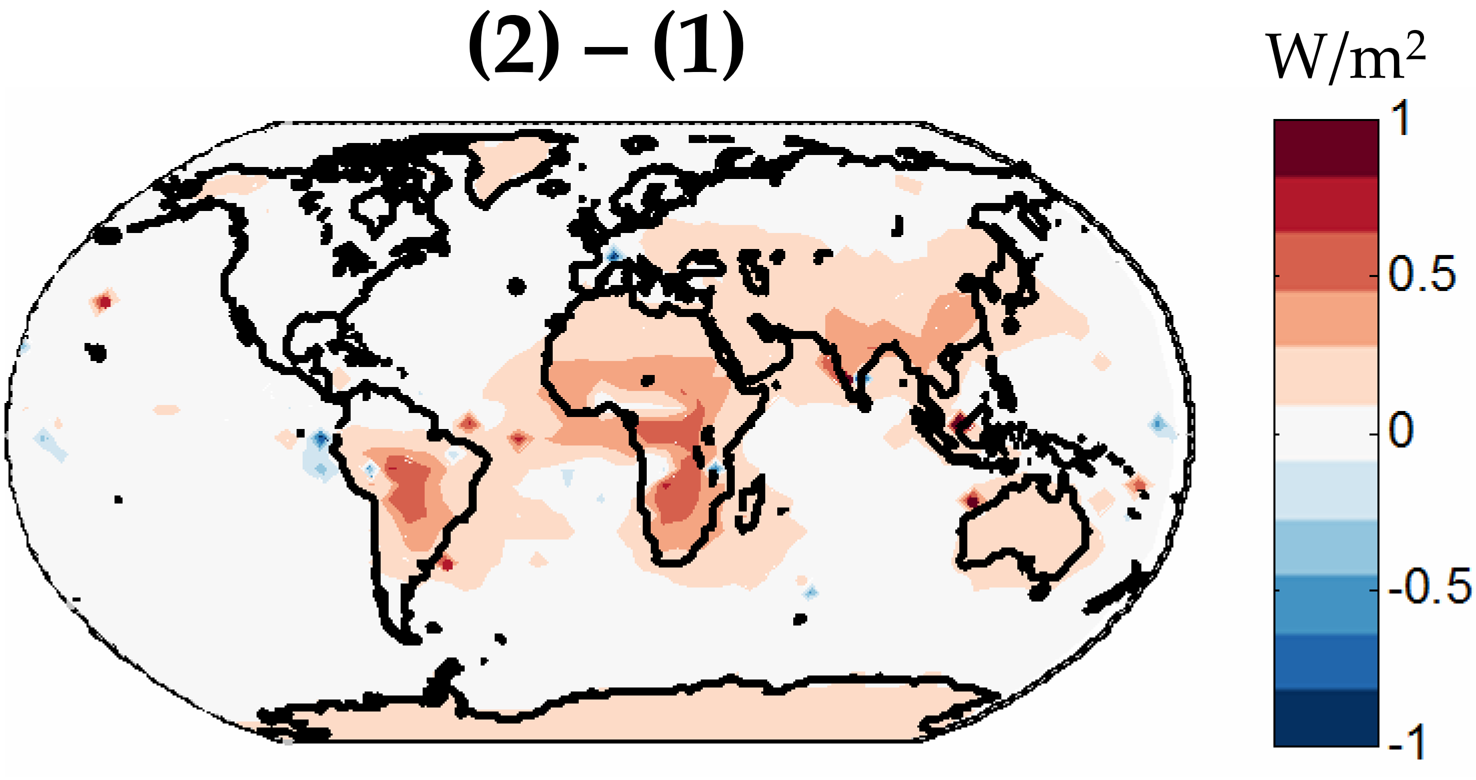
retrieval morphology
Climate model morphology
DRE
1)
true
true
k_{OA}
true
true
2)
true
true
k_{OA}
wrong
wrong
3)
true
wrong
wrong
k_{OA}
wrong
less wrong

GEOS-Chem modeling:
DRE of biomass-burning BC+OA

(1) "true"



GEOS-Chem Modeling
DRE of biomass-burning BC+OA

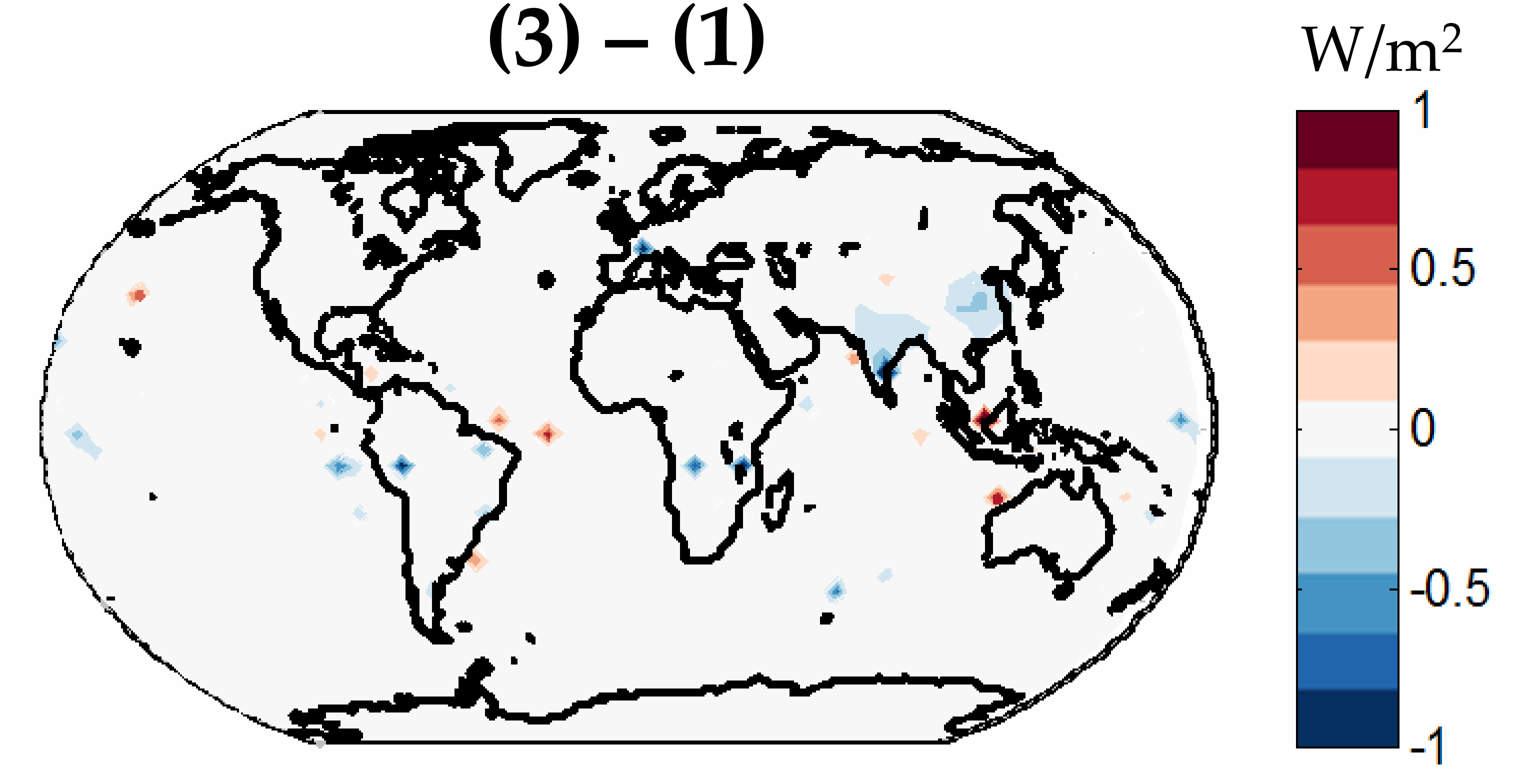


global average = +0.11 W/m^2

relative error = 40%

GEOS-Chem modeling:
DRE of biomass-burning BC+OA

(3) – (1)



global average = -0.01 W/m²

relative error = 3%

Conclusions

- Condensation and chemical aging of biogenic and anthropogenic SOA on BC was reproduced within experimental error by core-shell Mie models.
  - No effect of O:C during aging of SOA
- Brown carbon in emissions from biomass burning is associated mostly with organic compounds of extremely low volatility
  - Effect can be parameterized as a function of BC/OA
  - Quite sensitive to burn conditions
- Estimated radiative forcing of 0.1-0.2 W m^{-2} due to biomass burning BrC.
  - Net effect of biomass burning is still cooling.
- This effect was not observed in diesel emissions

Conclusions

- Diesel sources responsible for approximately 25% of particle number emissions in the Eastern US during summer
  - 30% of emissions of N_{100}
- Reduction of these emissions leads to increases of nucleation rates
  - Increases of very small particles predicted
  - The N50 and N100 concentrations decrease more than expected
  - This reduction in CCN could result in warming
- Development of a computationally efficient multi-distribution model to better simulate the mixing state of BC in regional models

Acknowledgments

- Graduate students/postdocs

  Laura Posner, Christos Fountoukis, Antonis Tassoglou.