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Black Carbon, Air Quality and Climate

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The Team

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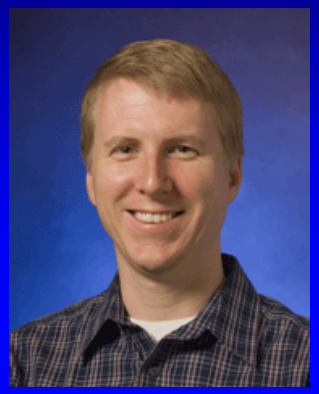

Peter Adams

Rawad Saleh

R. Subramanian

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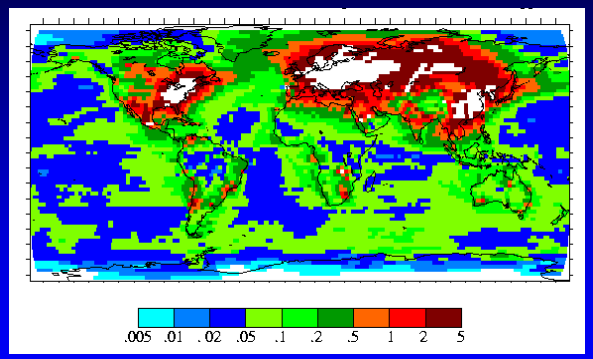
Black Carbon, Air Quality and Climate

Sources of BC

?



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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

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Project Overview

1. Laboratory Studies
   - Primary emissions characterization
   - Aging of primary emissions
2. Emission inventory development
   - Source-resolved inventories
   - Inventories for number
3. Model extension
   - Particle number source attribution
   - Mixing state and optical properties
4. Black carbon number concentrations
5. Regional scale simulations - Scenarios and controls

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BC Emissions, Chemical Aging, and Optical Properties

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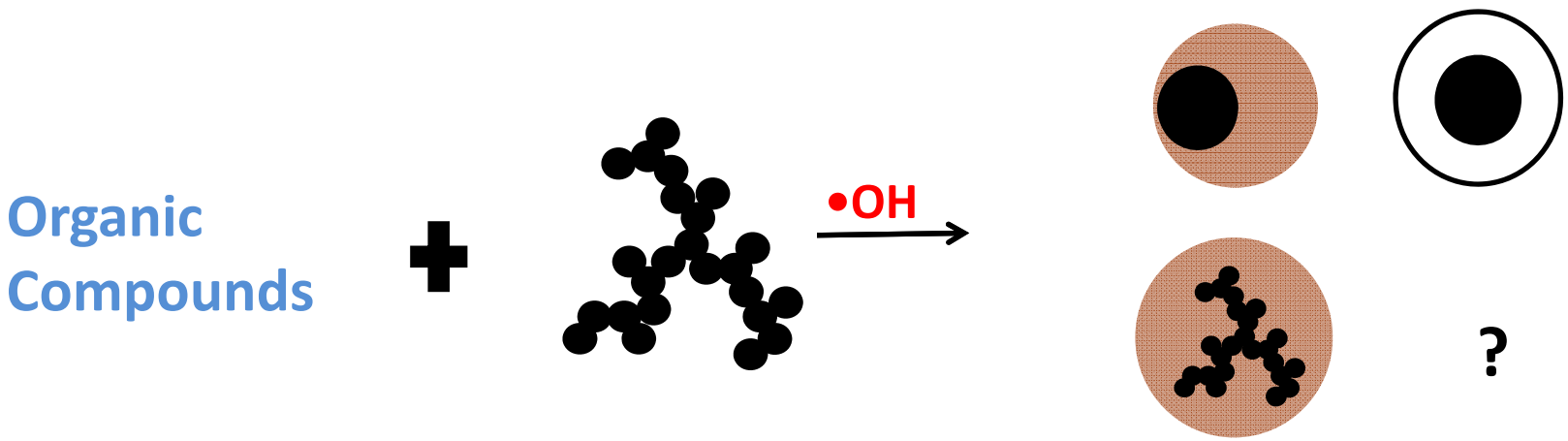
Combustion Emissions

R. Saleh

BC particles act as condensation sites for OA



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

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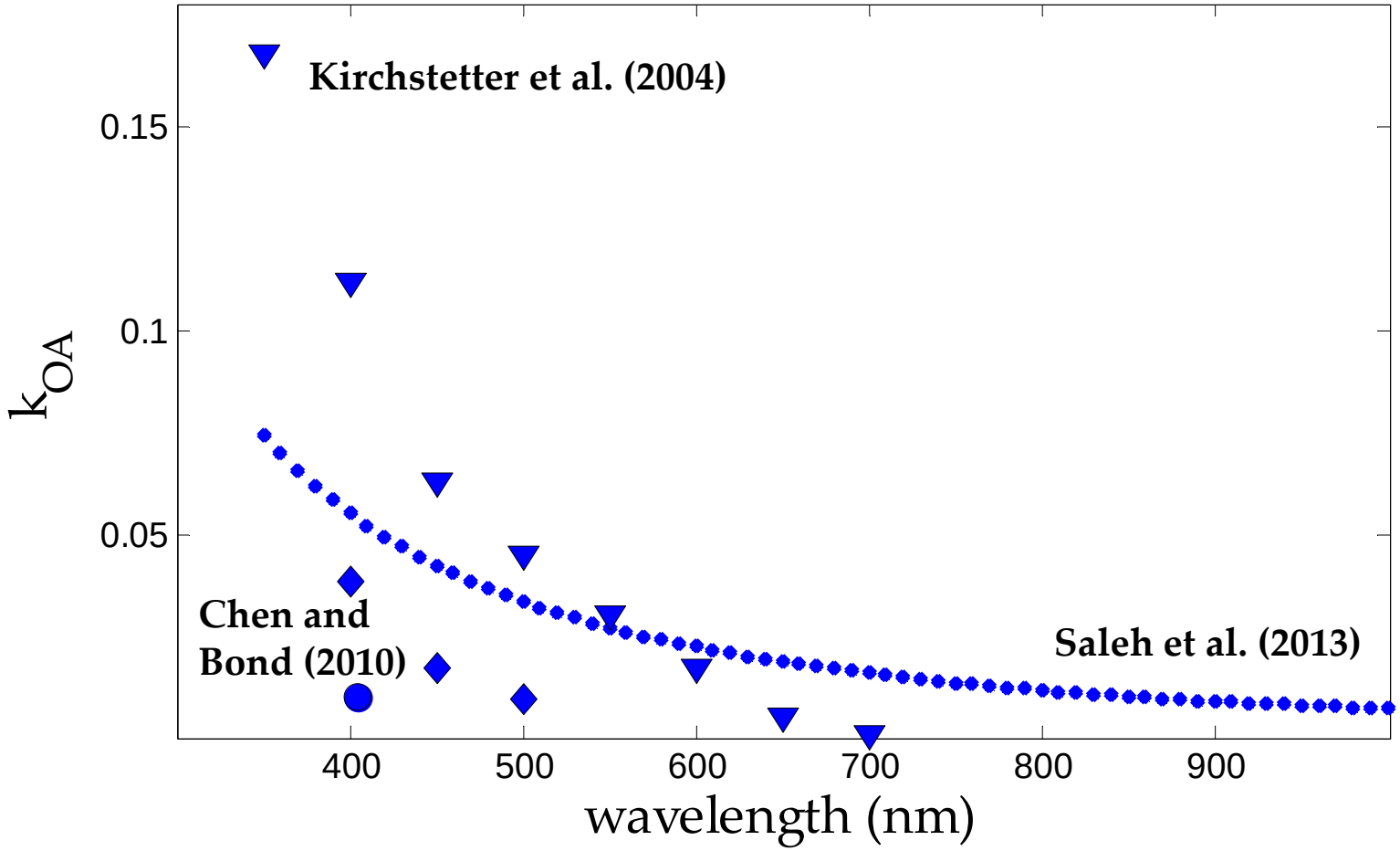
BrC in Biomass Burning: Chaos



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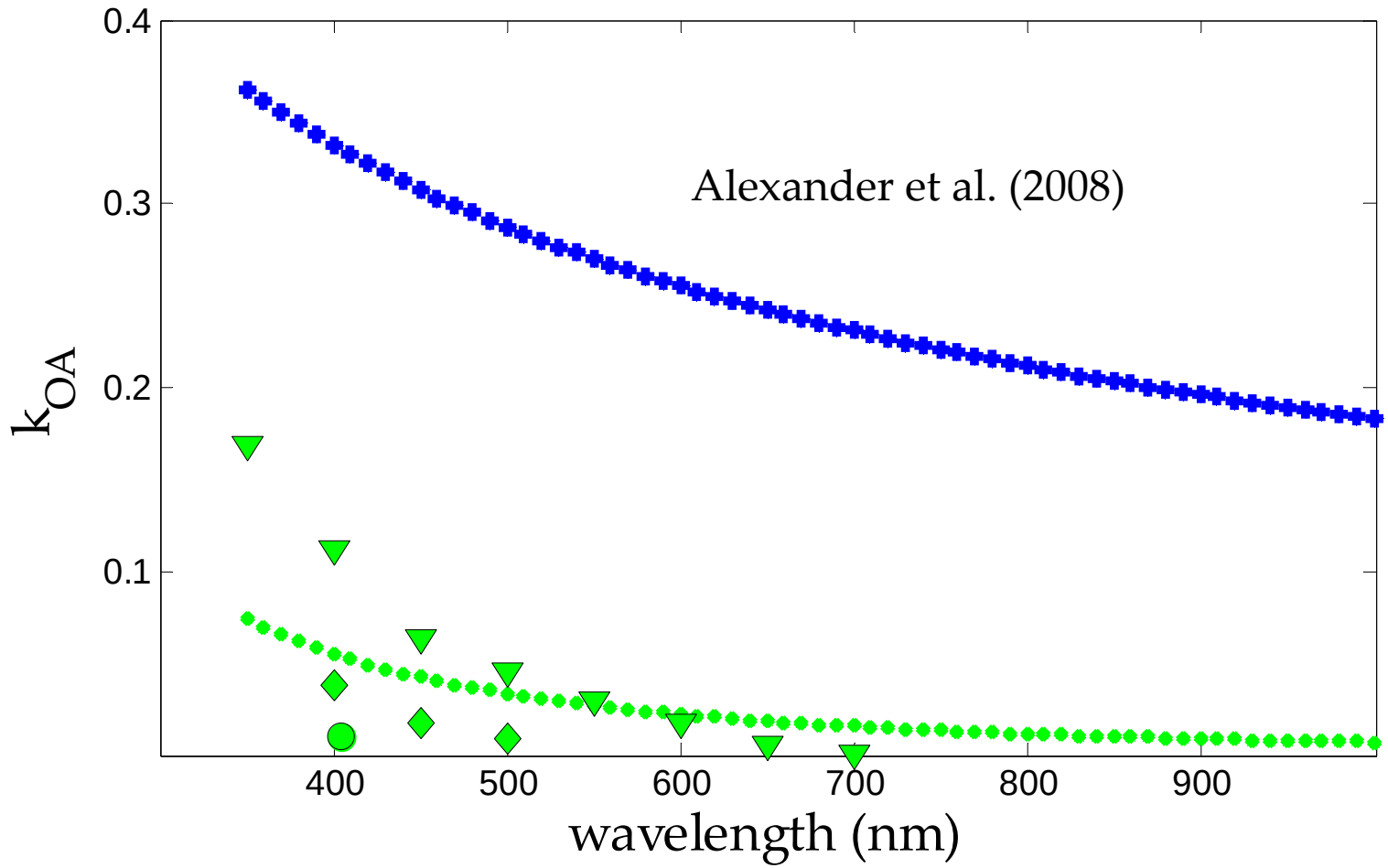
BrC in Biomass Burning: More Chaos



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

Aethalometer

OA/BC from biomass burning

SP2

PASS-3

HR-SP-AMS

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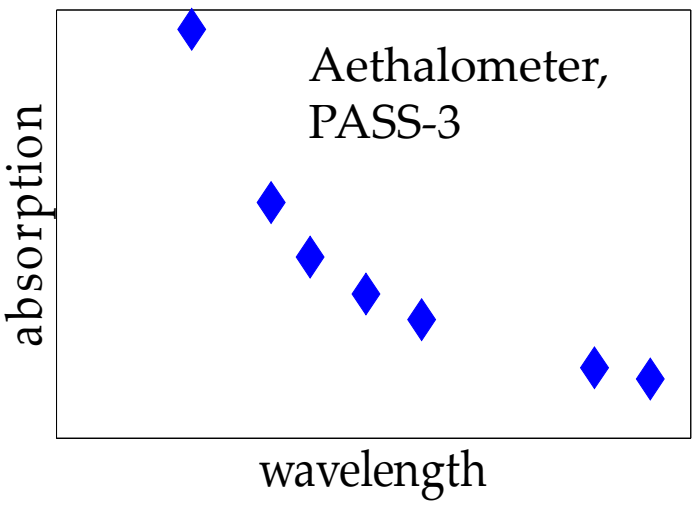
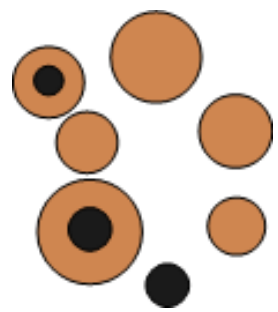
Estimation of OA Optical Properties

b_{abs} = f(size distributions, refractive index [BC], refractive index [OA])

Bond et al. [2005]

$$k_{OA}(\lambda) = k_{OA,550}\left(\frac{550}{\lambda}\right)^{w}$$





SMPS,
SP2

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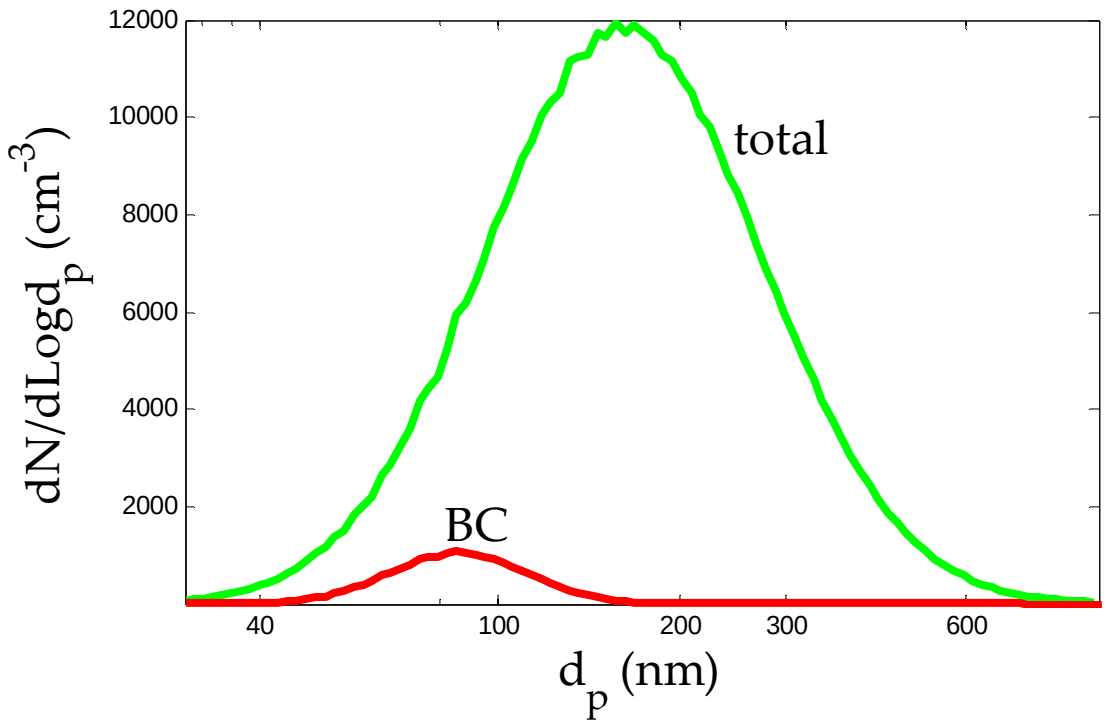
Morphology and Mixing State



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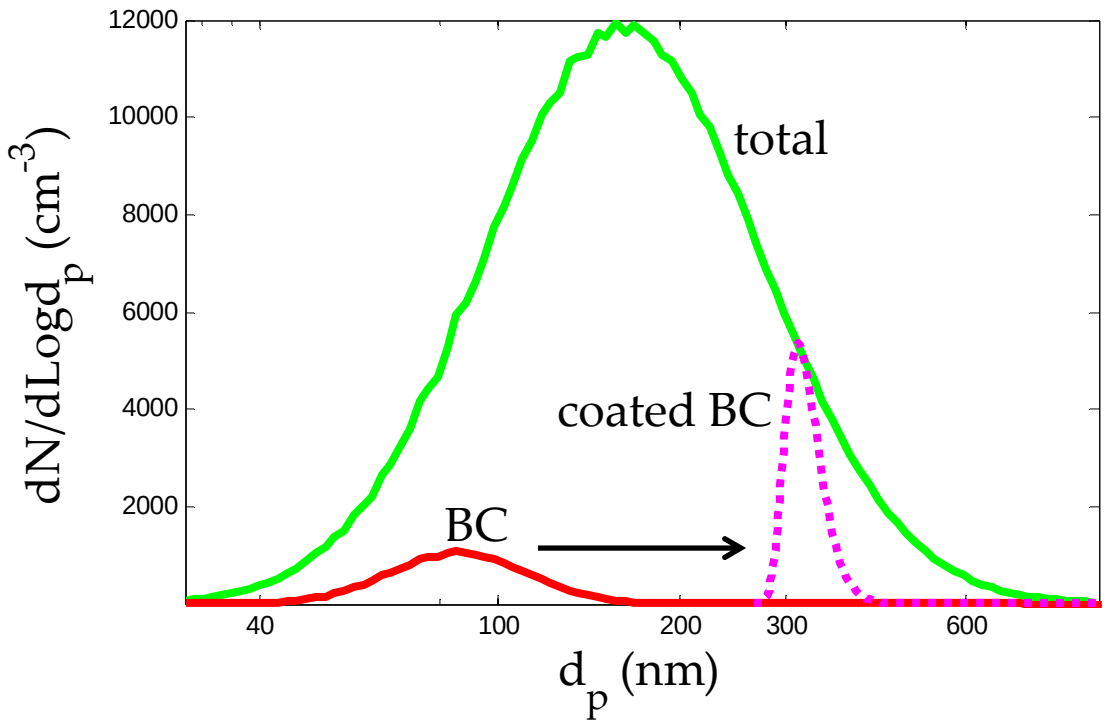
Morphology and Mixing State



- We simulate the condensation process of OA on BC.
- The growing distribution cannot go beyond the SMPS distribution.
- We can only constrain the maximum coating thickness.
- This maximizes the lensing effect, thus minimizes BrC absorption
- Conservative approach.

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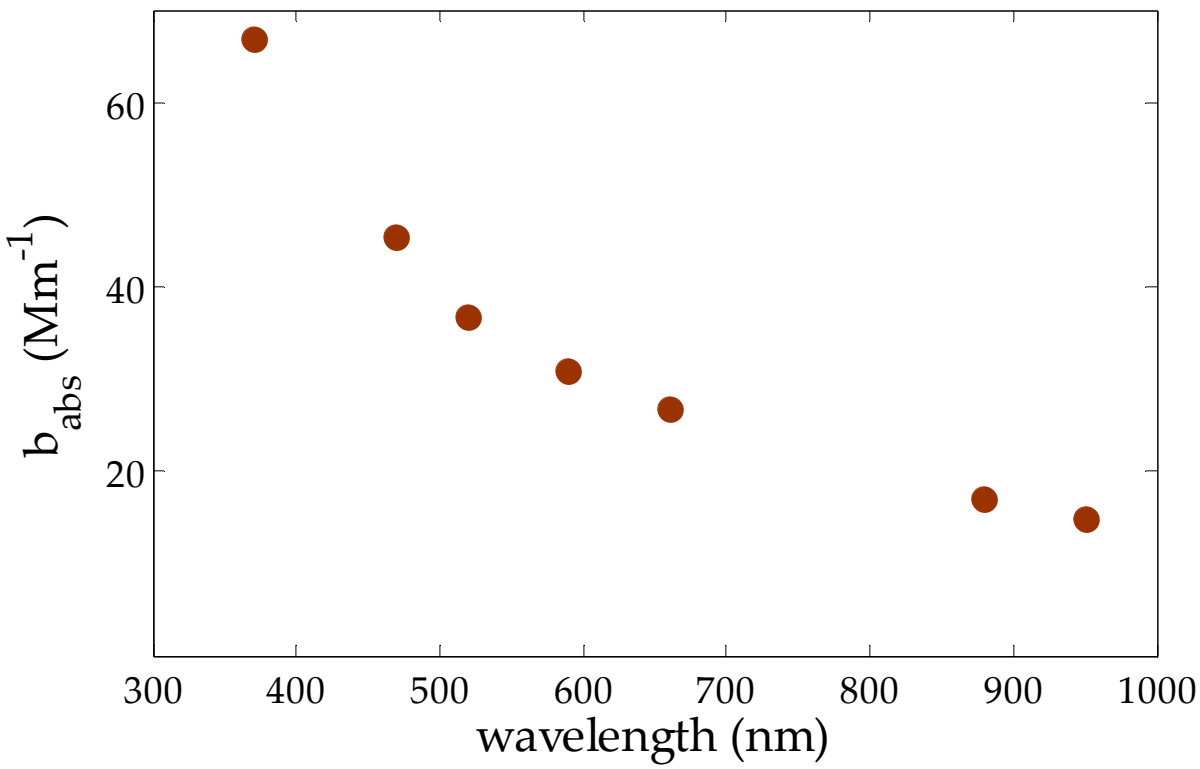
The Fit



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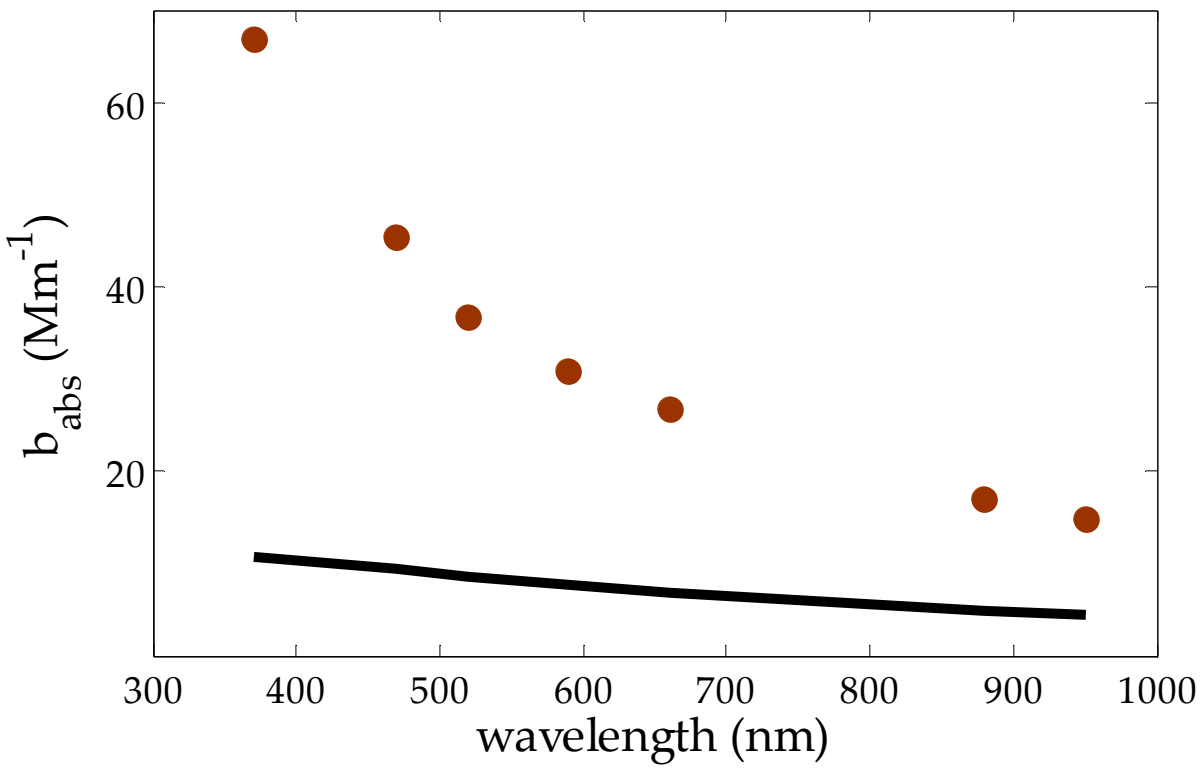
The Fit (Absorption due to BC only-Mie calculations)



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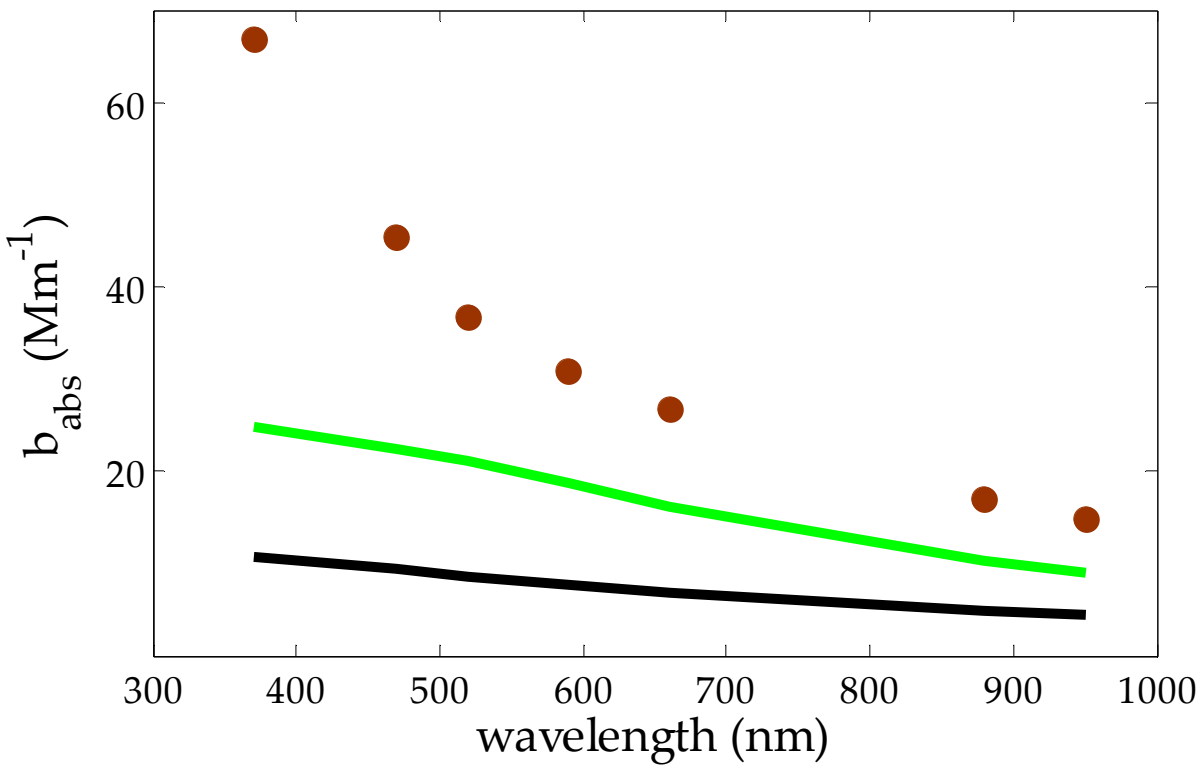
The Fit (Absorption due to BC+ Lensing)



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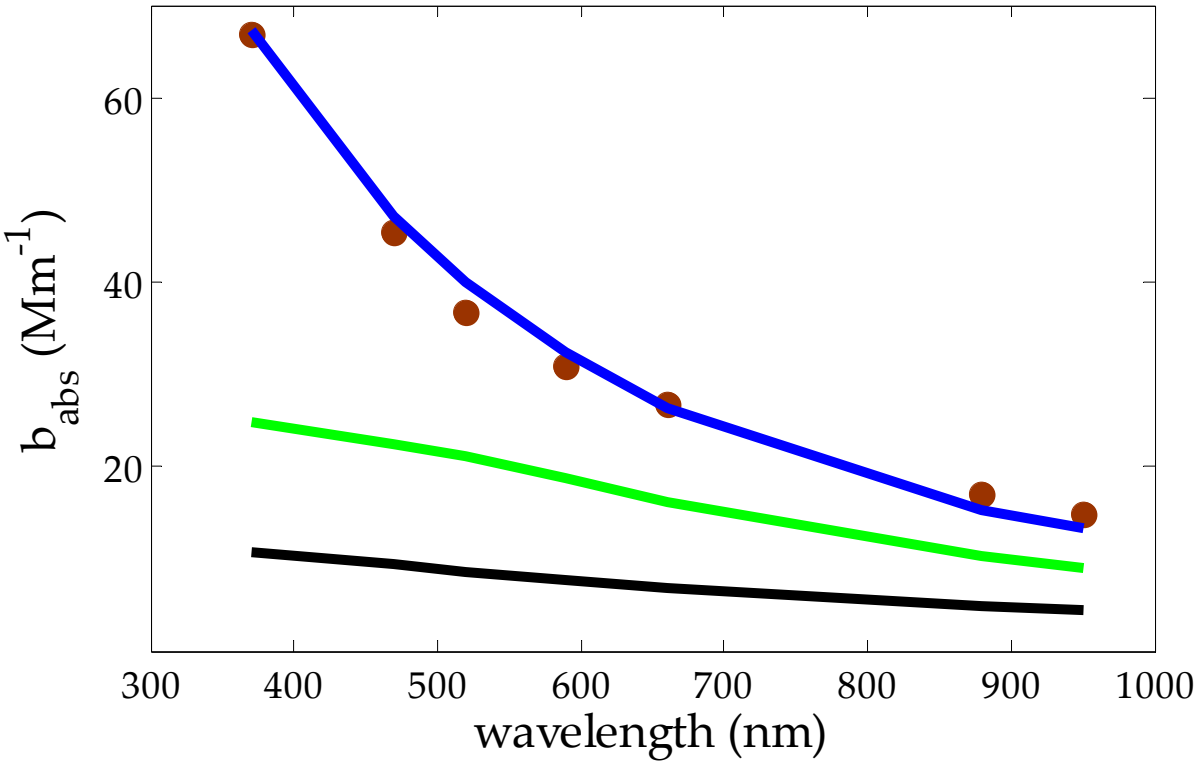
The Fit (Absorption due to BC+ Lensing+BrC)



The best fit, from which we obtain the absorptivity of OA.

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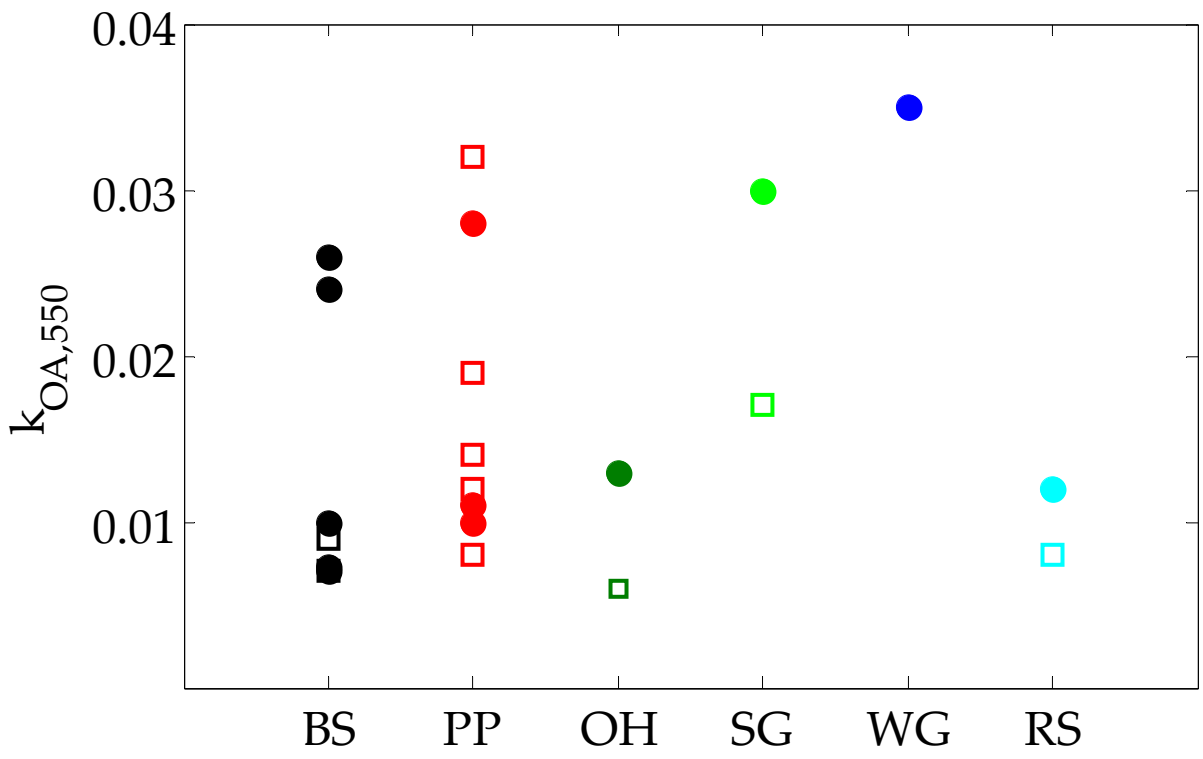
Chaos Returns !



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.

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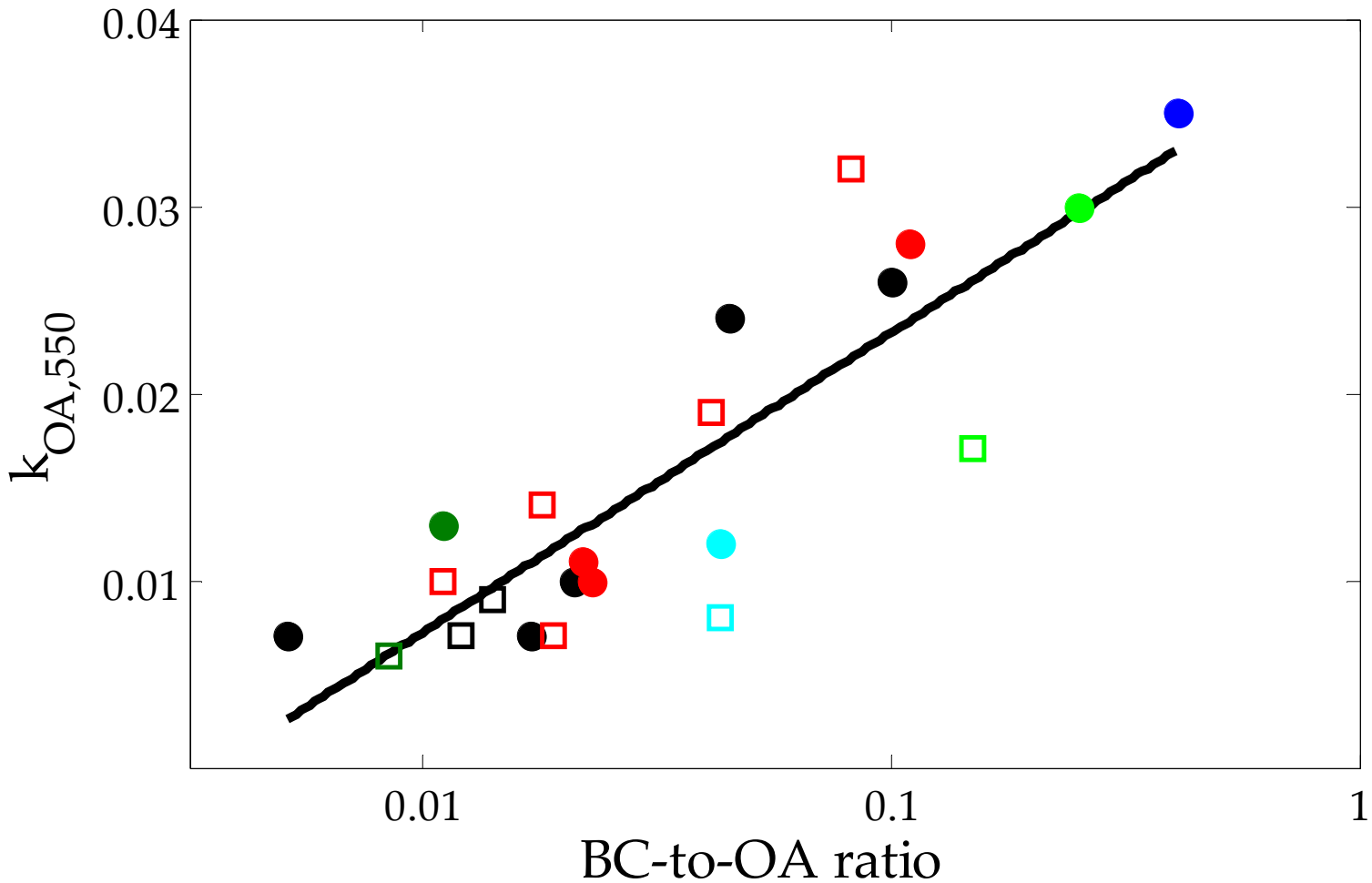
Some Order



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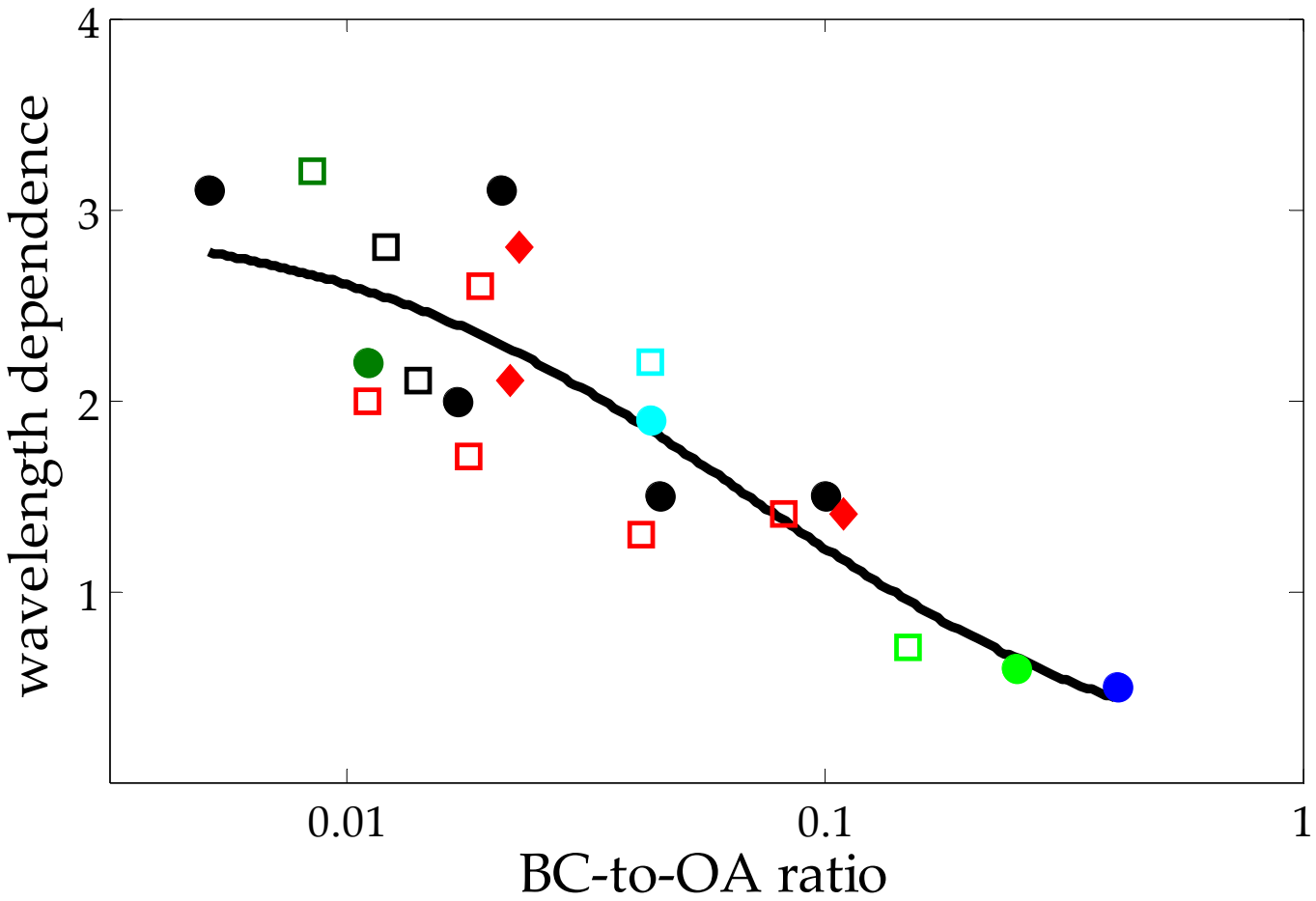
Some Order



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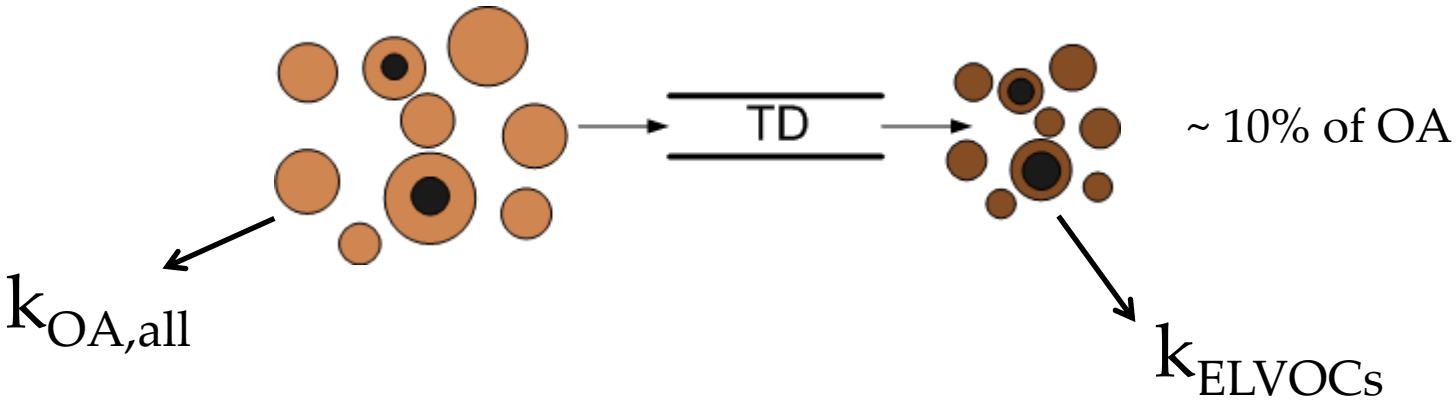
BrC and OA Volatility



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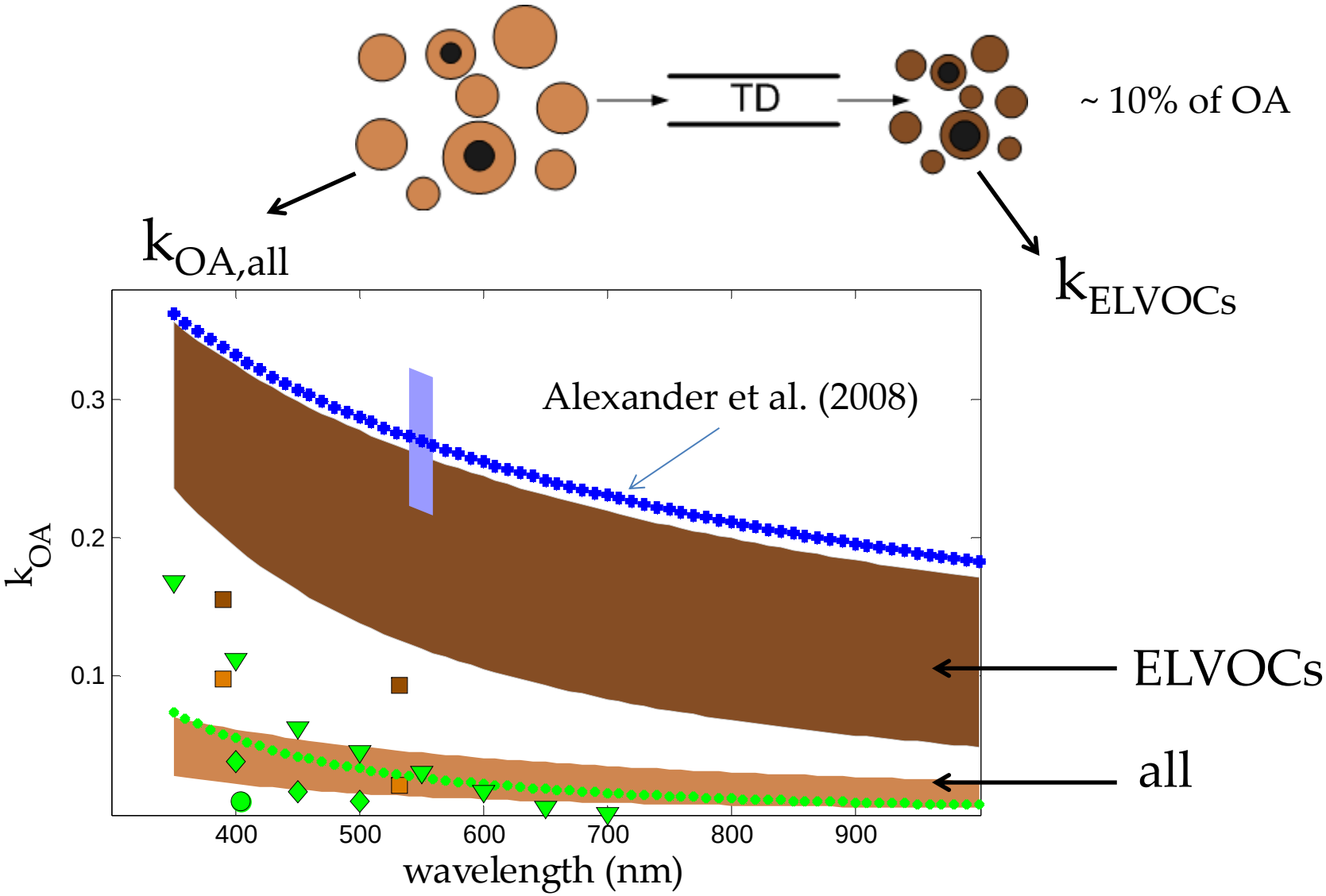
BrC and OA Volatility



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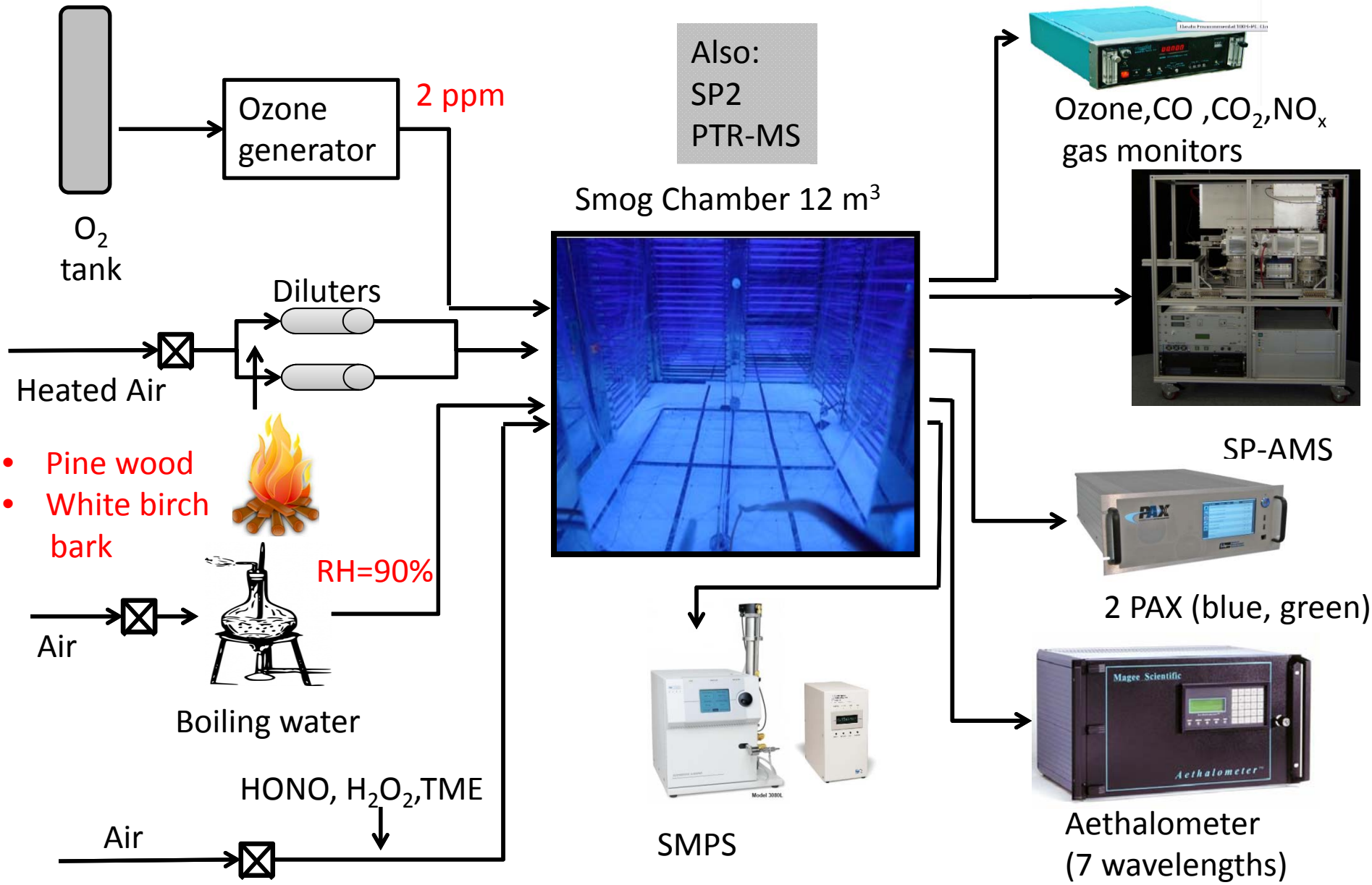
CMU Smog Chamber



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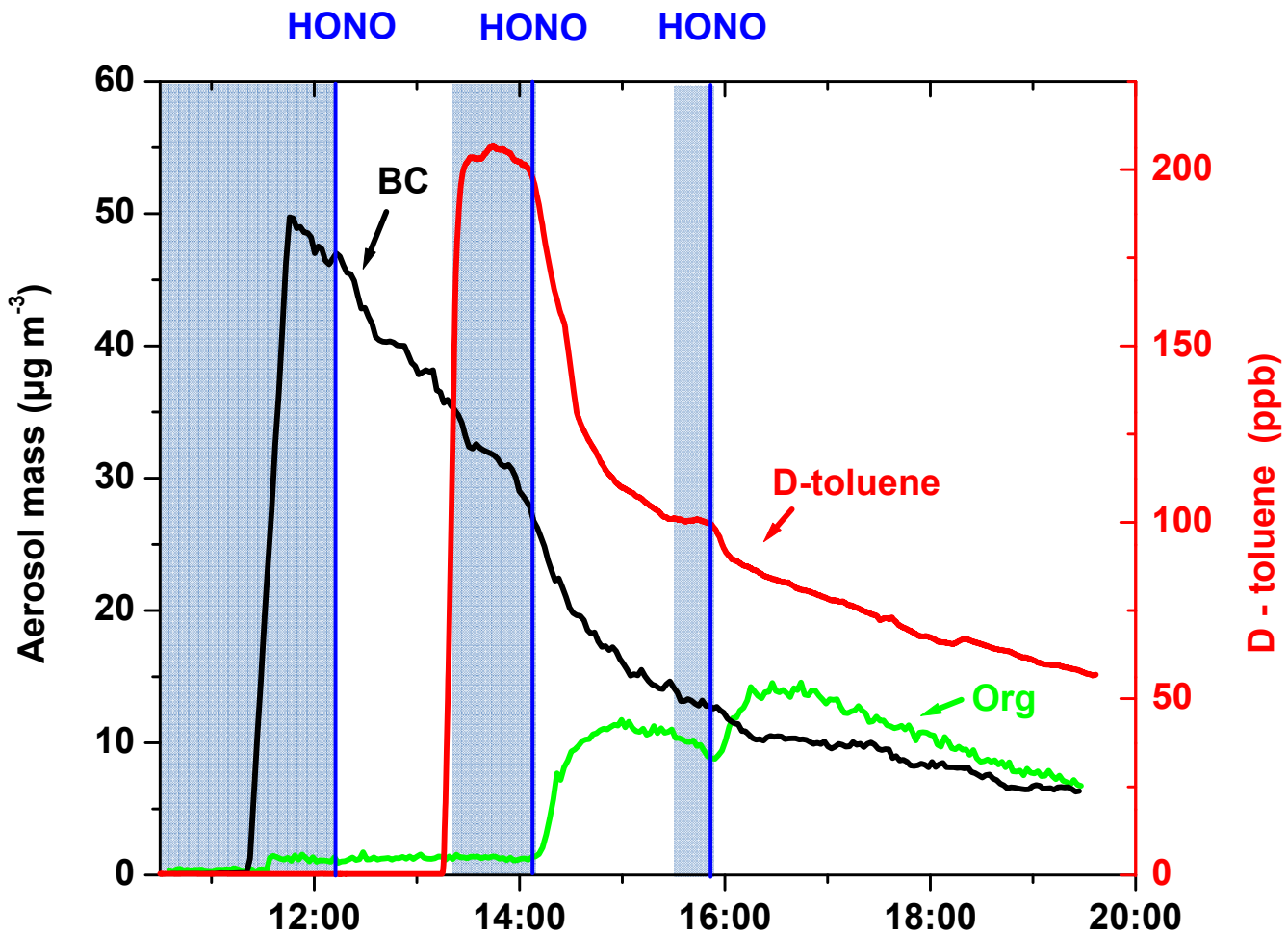
Coating of BC with D-toluene SOA

(fuel: White birch bark)



CE_{BC}= 0.4

CE_{org}=1

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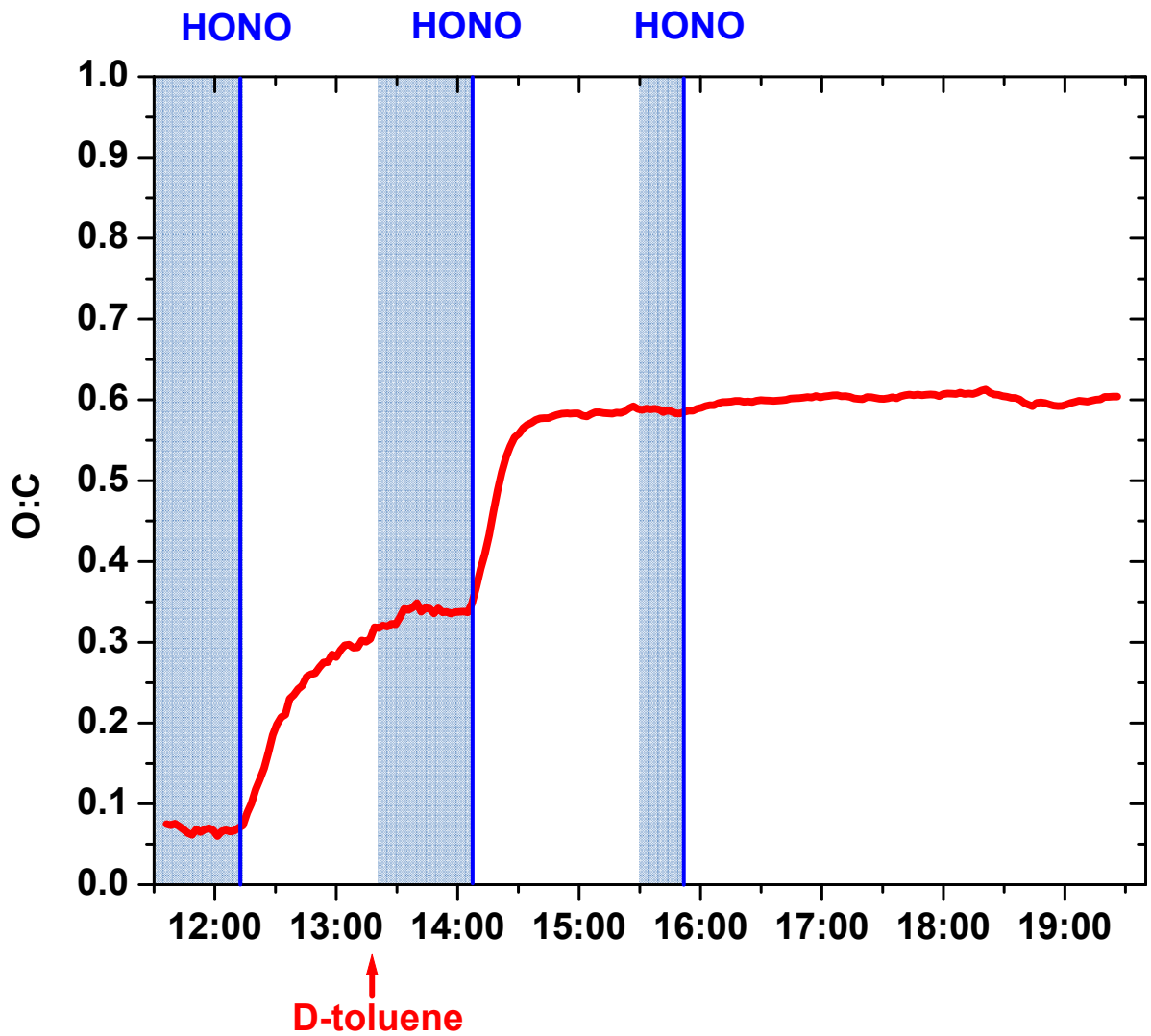
O/C during D-toluene SOA formation

(fuel: White birch bark)



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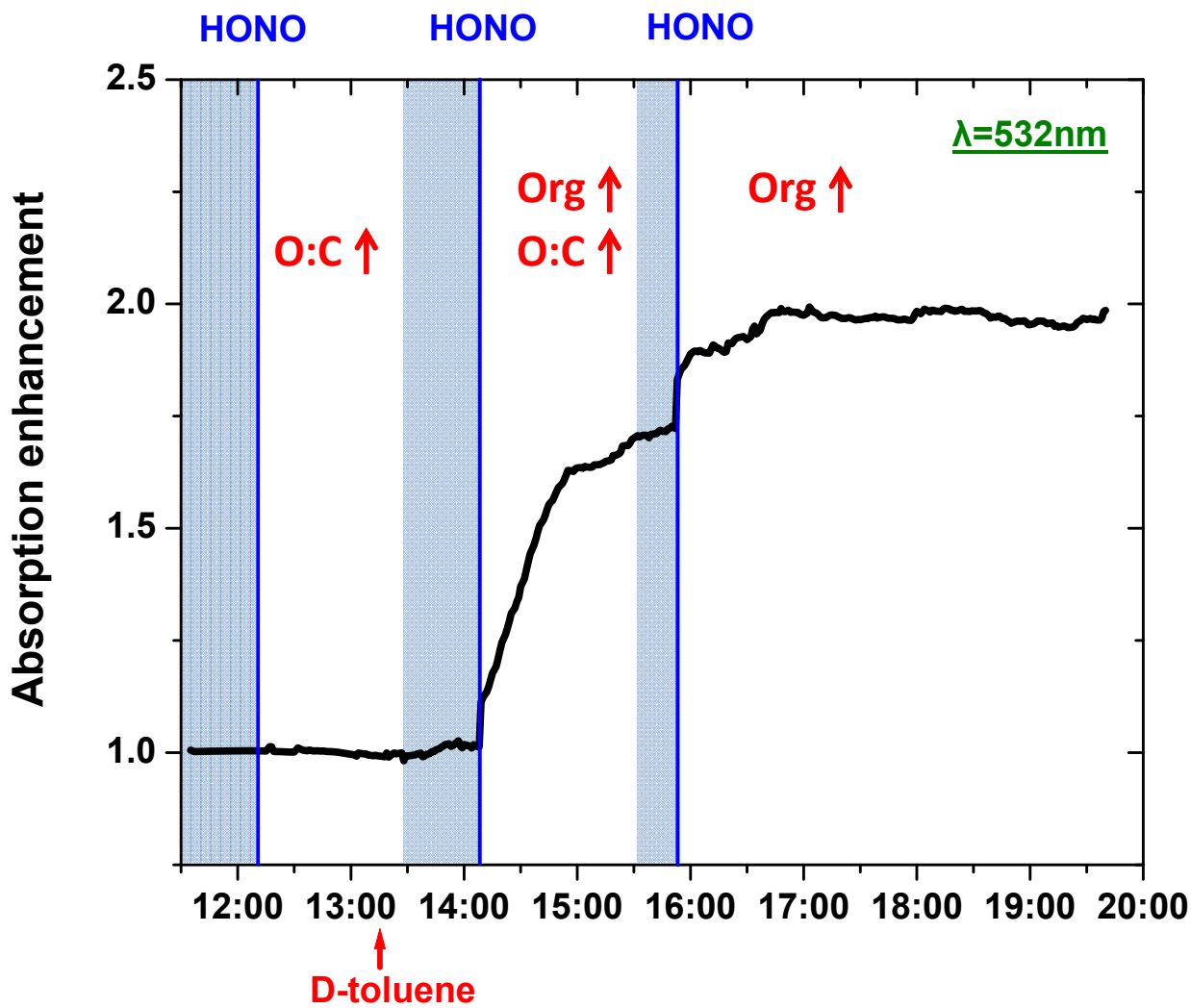
Absorption during D-toluene SOA formation

(fuel: White birch bark)



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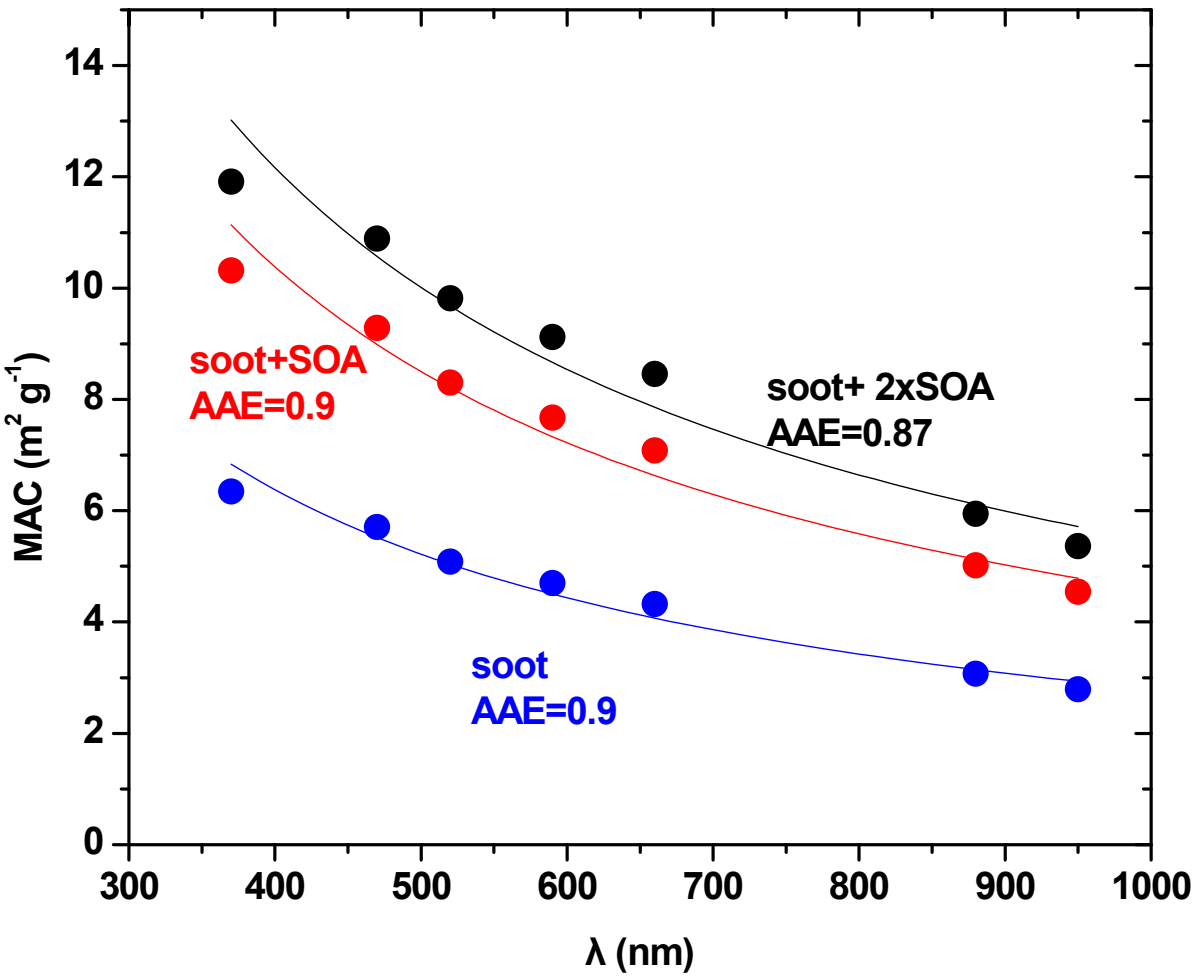
Absorption Angstrom exponent during D-toluene SOA formation

(fuel: White birch bark)



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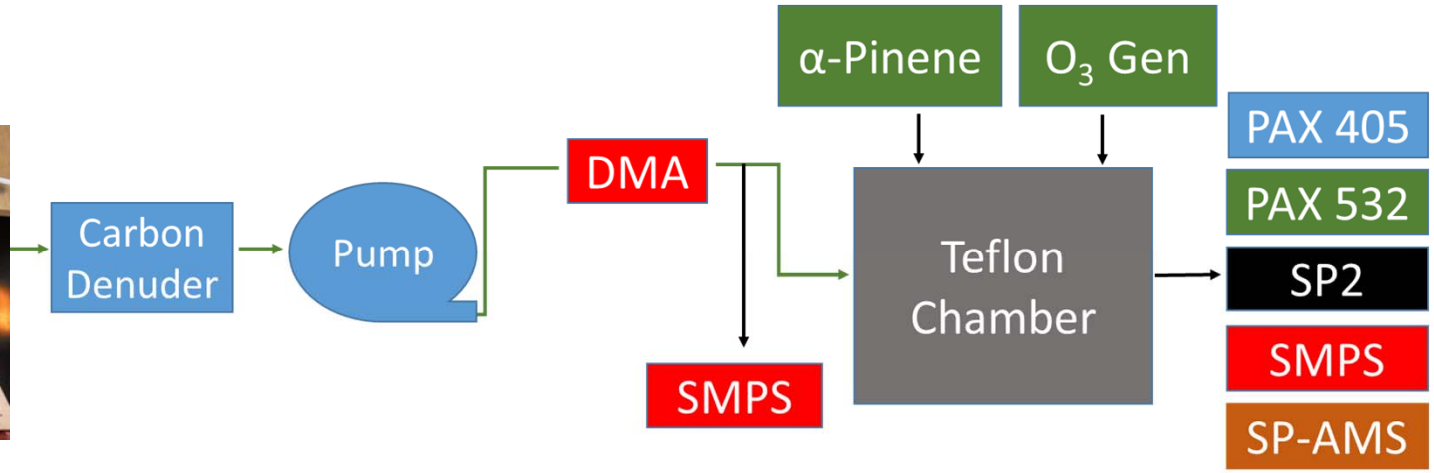
ıg of Monodisperse Cookstove Soot

R. Subramanian



- Absorption enhancement of mono-disperse aged BC particles.
- Three nascent BC core diameters (100, 130, 150 nm mass equivalent diameters).
- Soot was coated with α-pinene SOA in stages till a shell/core diameter ratio of ~2.5
- SP2 for BC mass; SP-AMS for organic aerosol mass; PAXs for light absorption/scattering.

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Black Carbon Emissions

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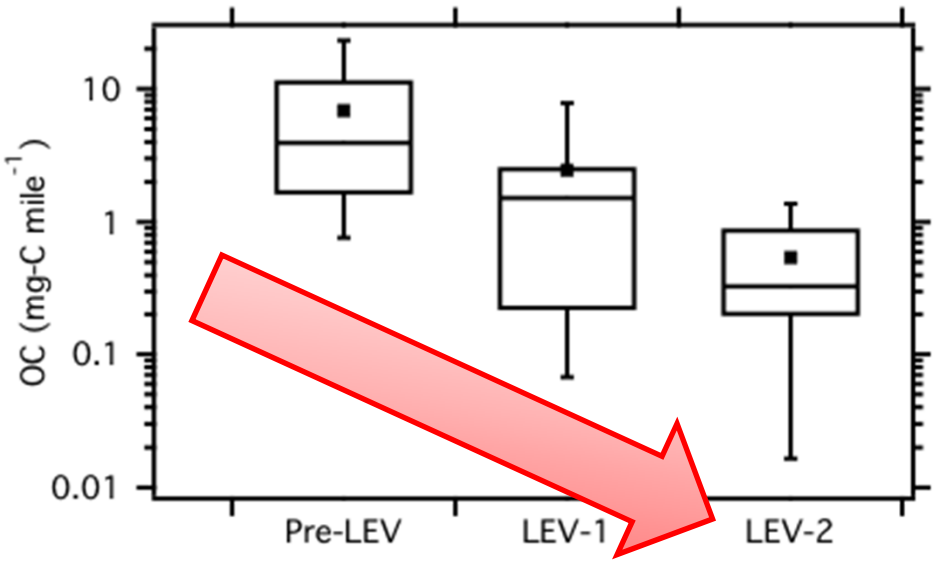
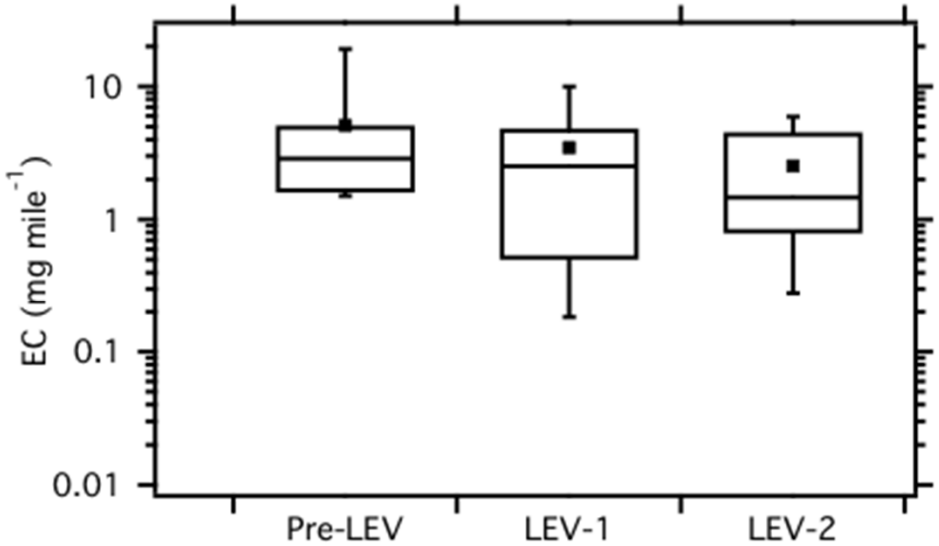
Organic/Elemental Carbon Emissions



OC emissions decrease with newer vehicles.



EC much less sensitive to more stringent standards.

Pre-LEV made before 1994
LEV-1 1994-2003
LEV-2 2004 and later

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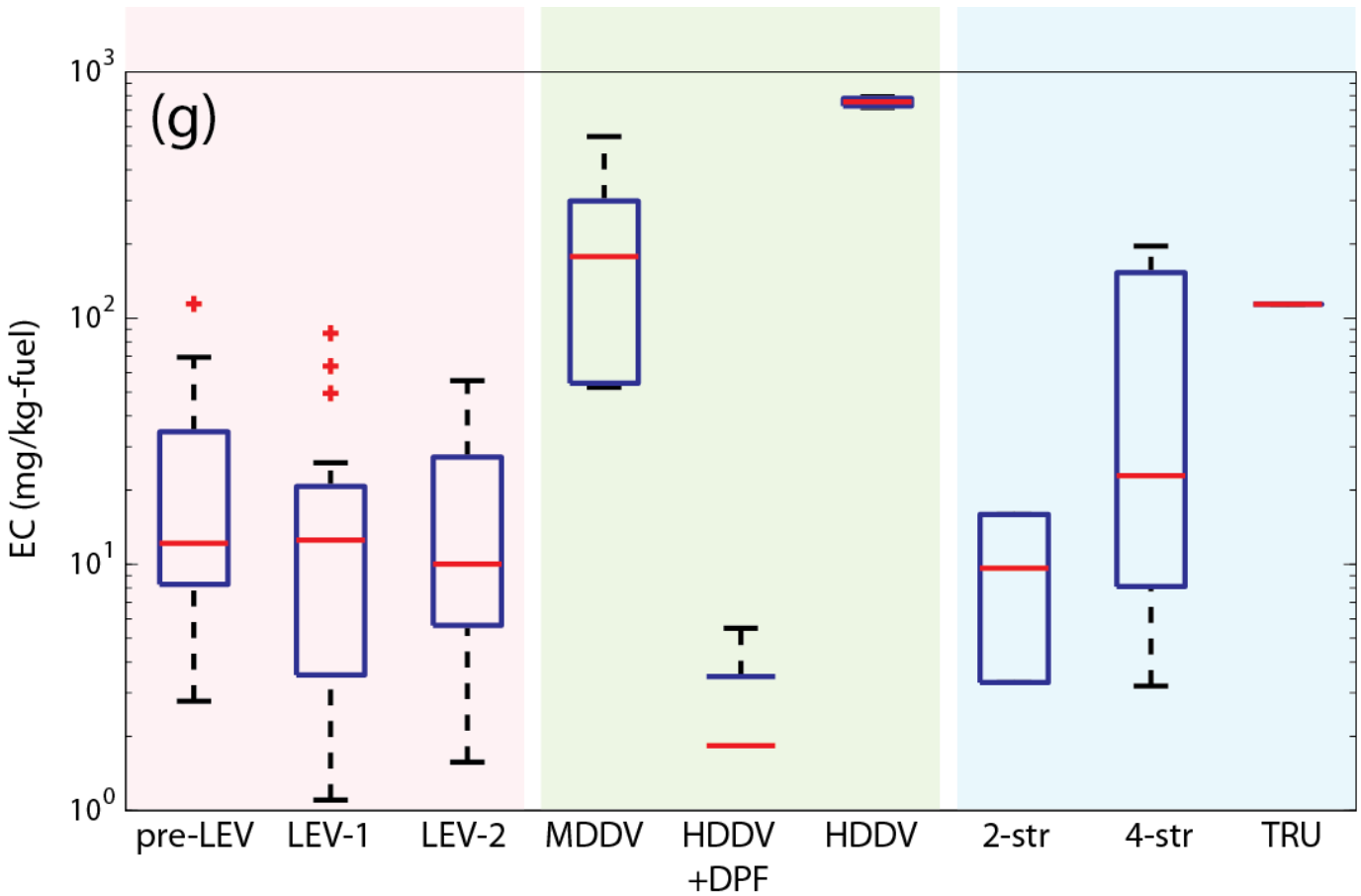
EC Emissions



May et al. (Atmos. Environ, 2014)

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BC and Aerosol Number Concentrations

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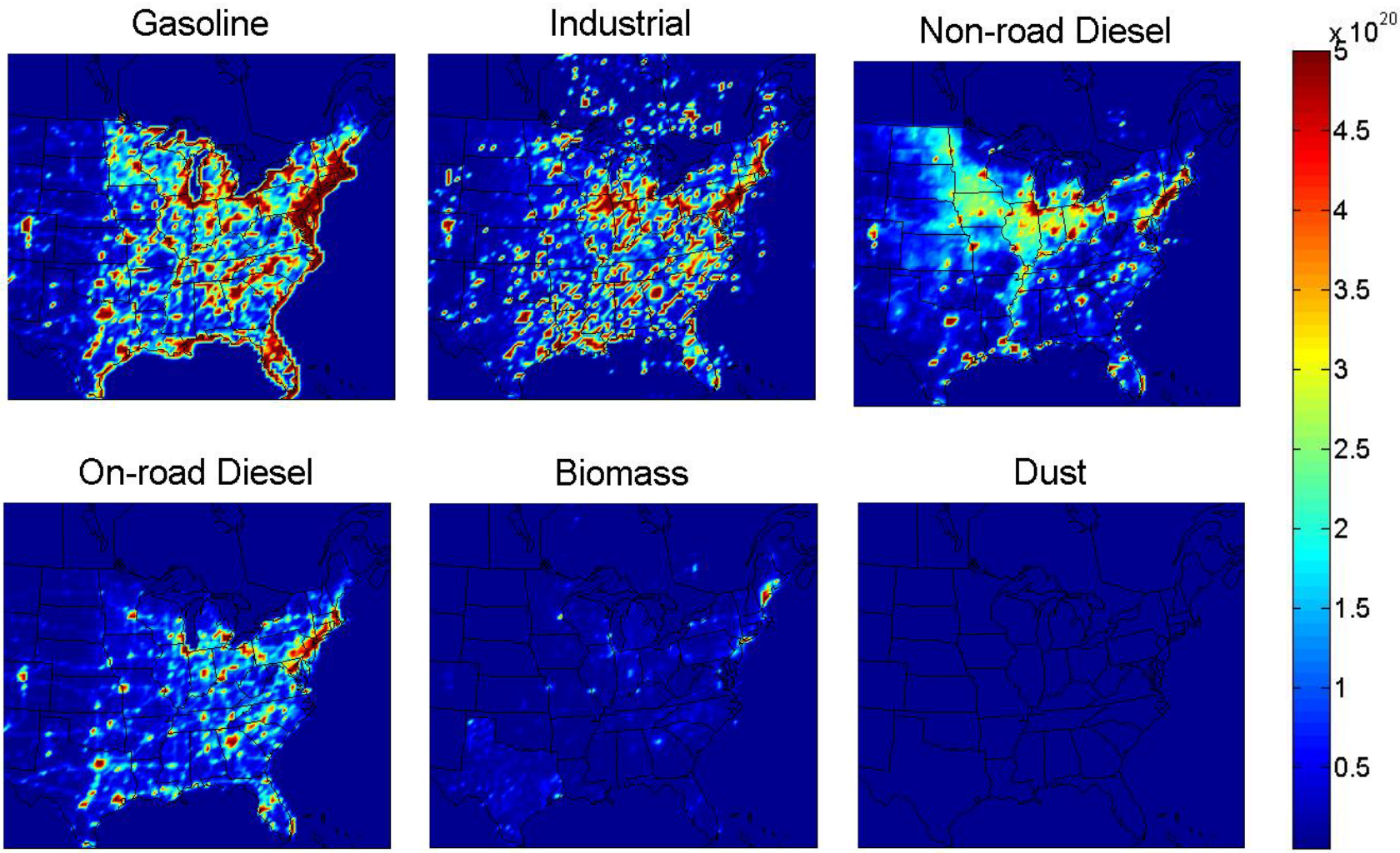
Source-Resolved Total Number Emissions (particles d^{-1} km^{-2})



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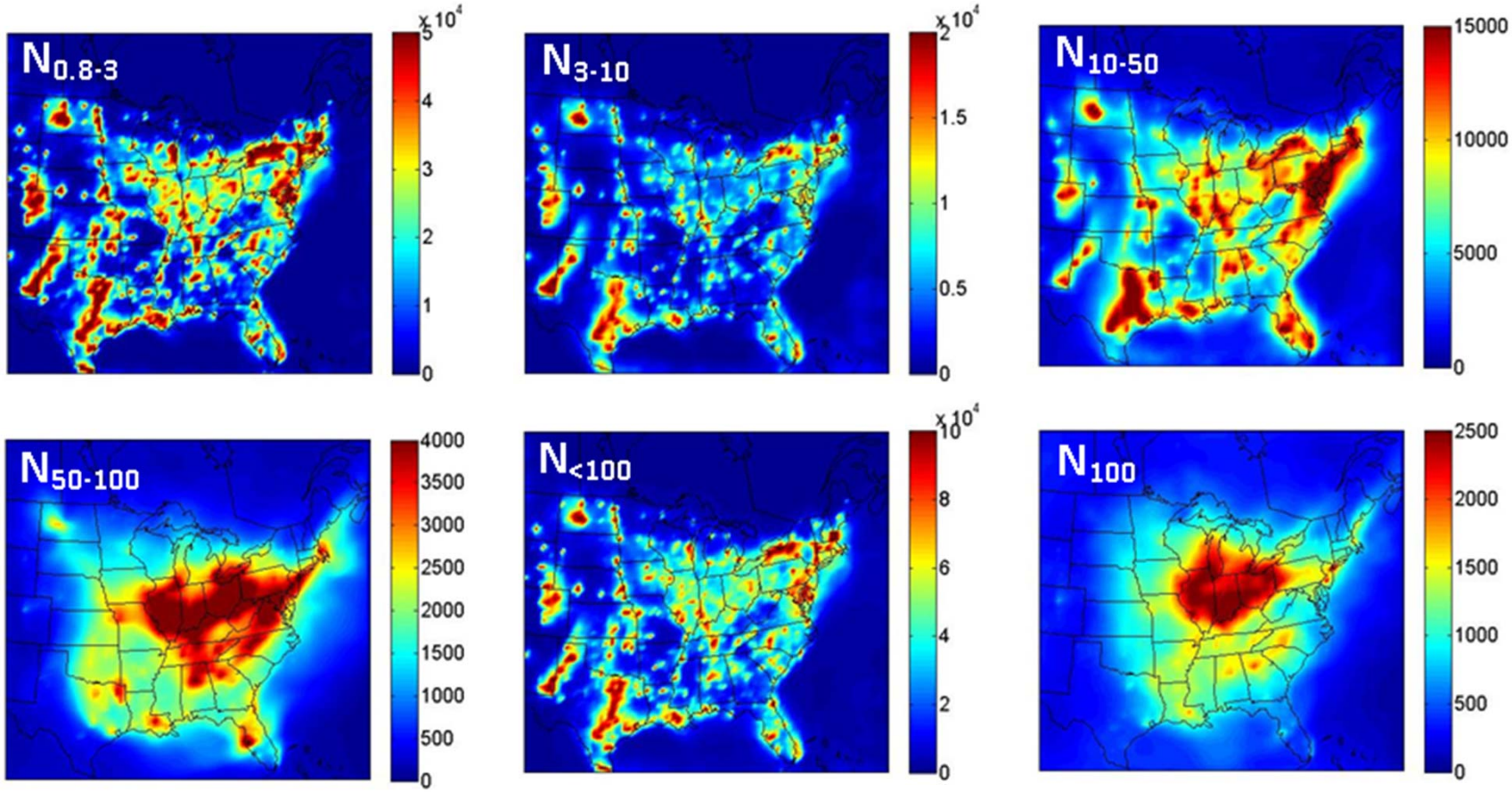
PMCAMx-UF base number concentration (particles cm^{-3})



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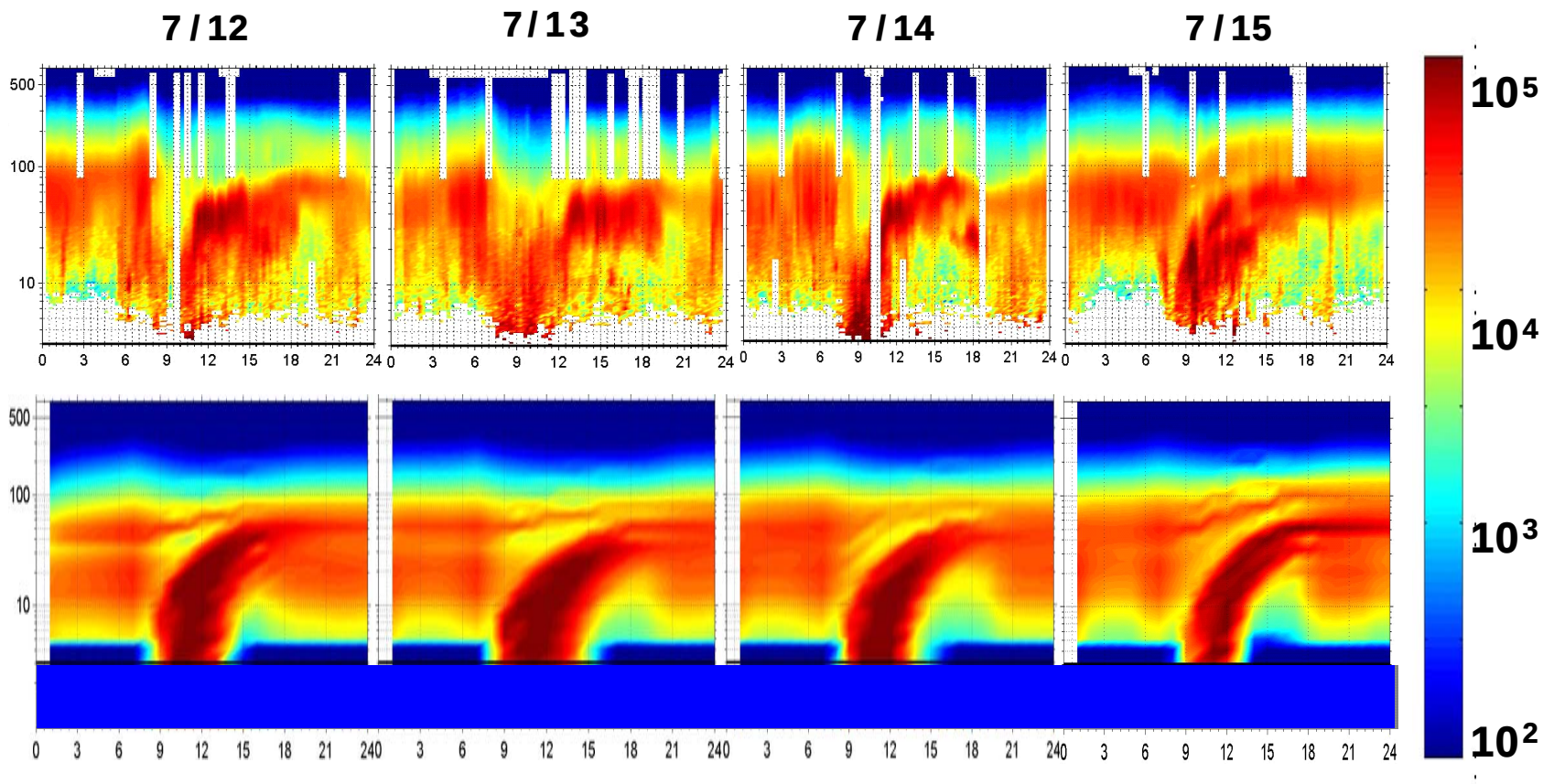
PMCAMx-UF Evaluation (Pittsburgh)



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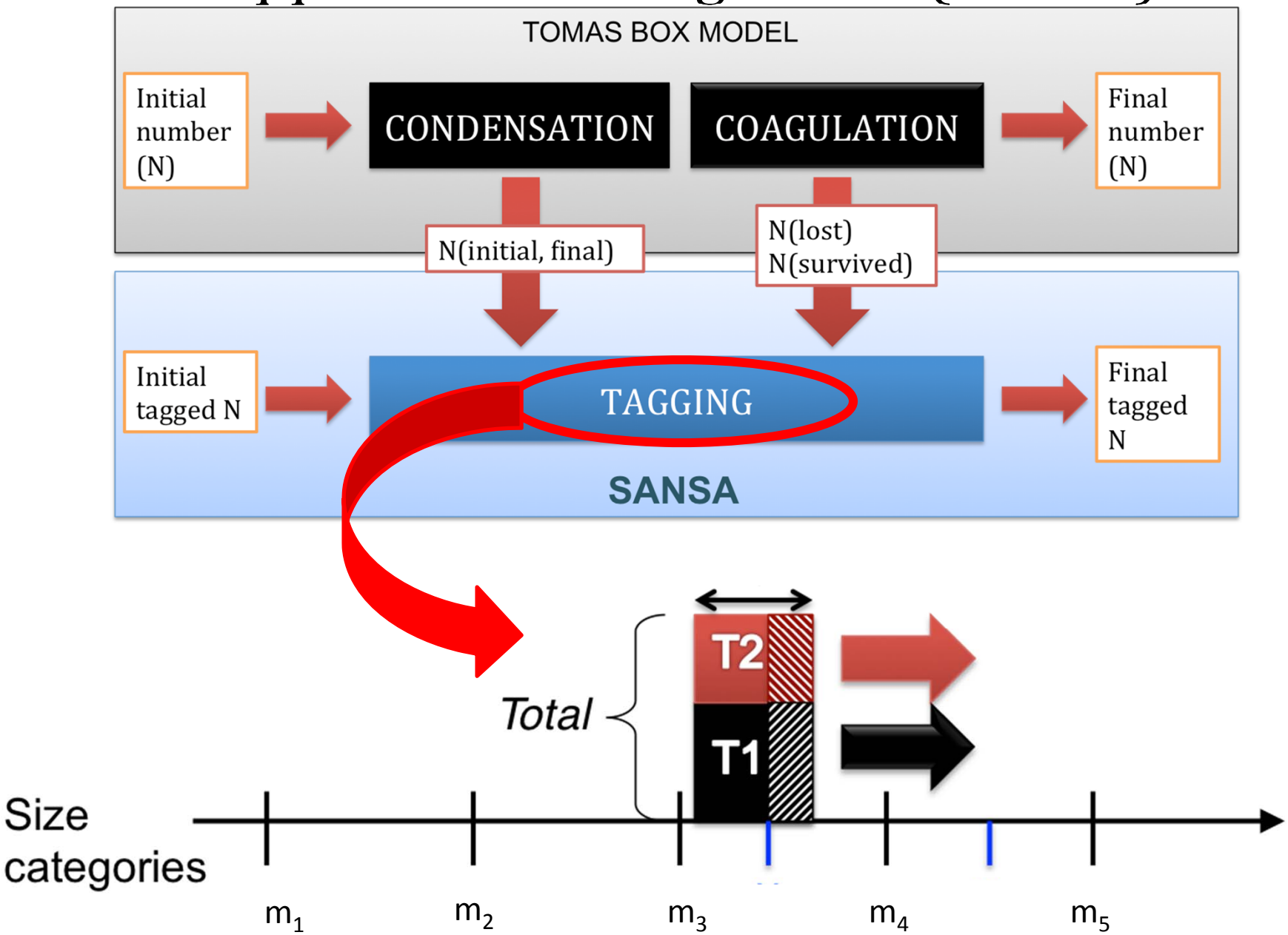
Size-resolved Aerosol Number Source Apportionment algorithm (SANSA)



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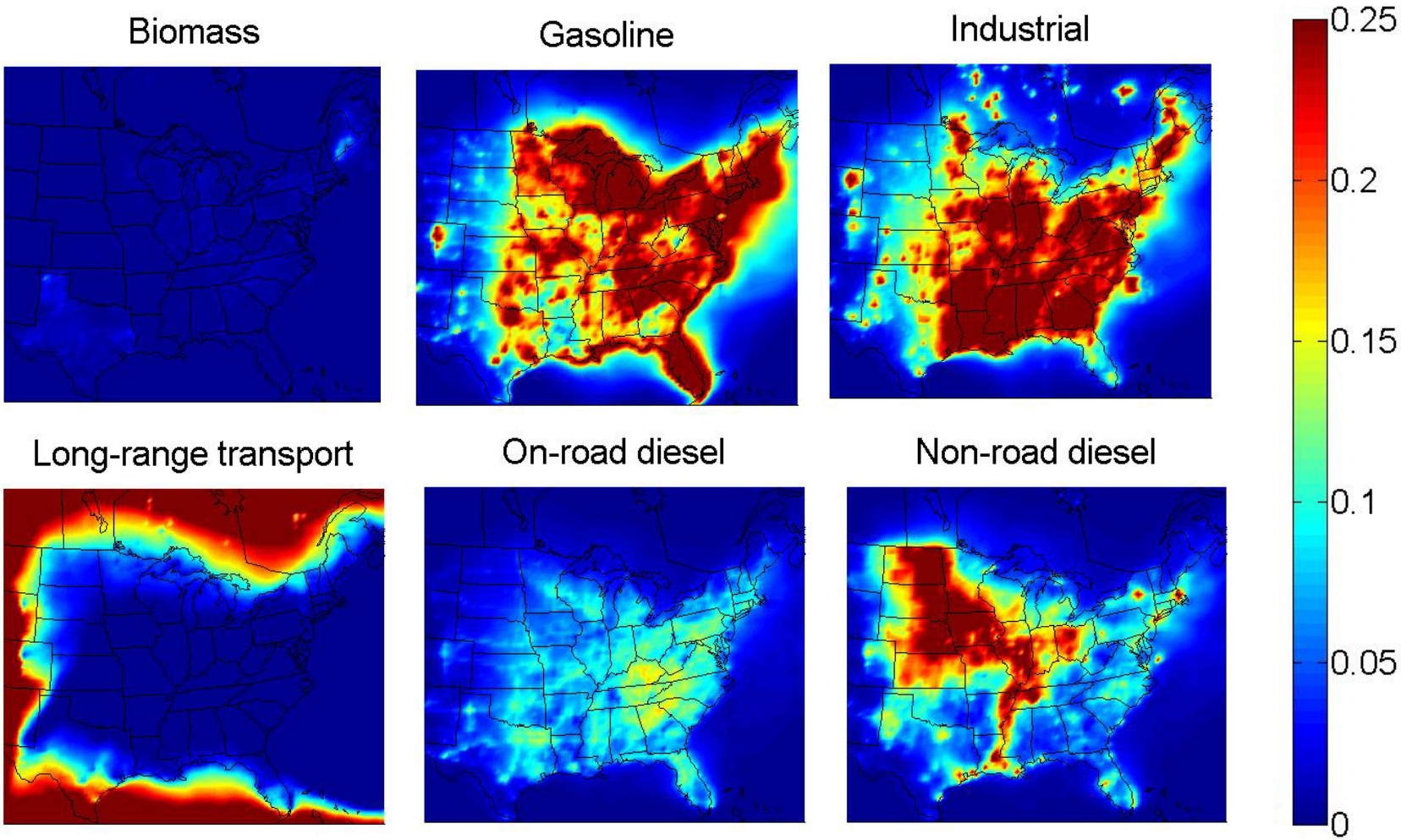
Total primary particle number fractional source contributions



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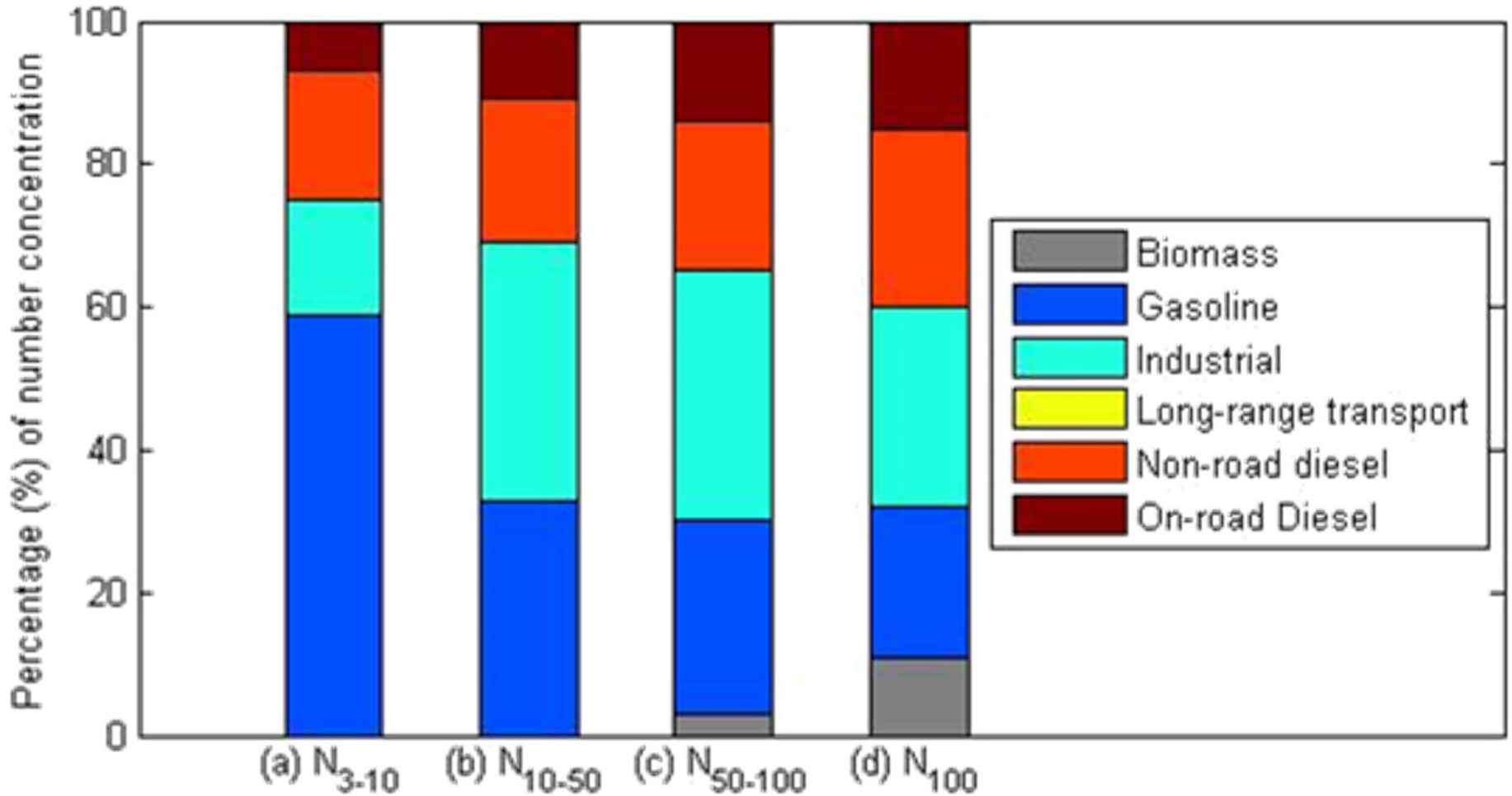
Primary particle number source apportionment in Pittsburgh



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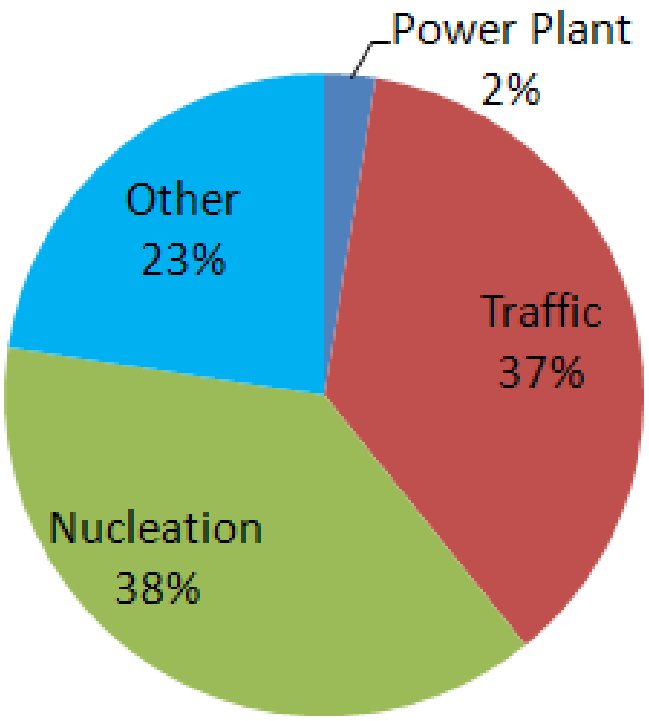
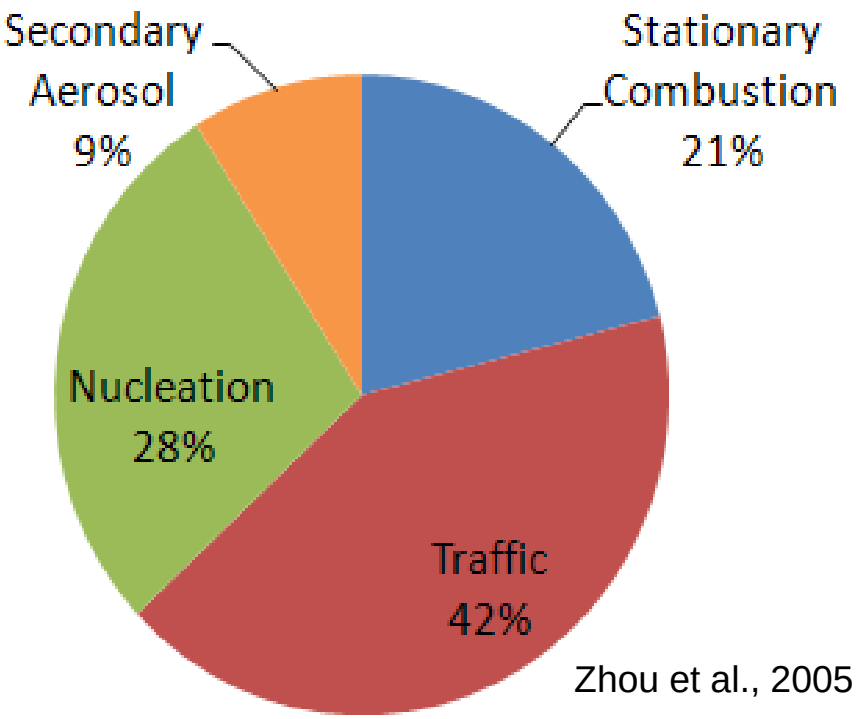
Sources of Measureable (>3 nm) Particle Number in Pittsburgh

PMCAMx



Predicted: 29,000 cm^{-3}

Calculated from Measurements



Zhou et al., 2005

"Measured": 26,000 cm^{-3}

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

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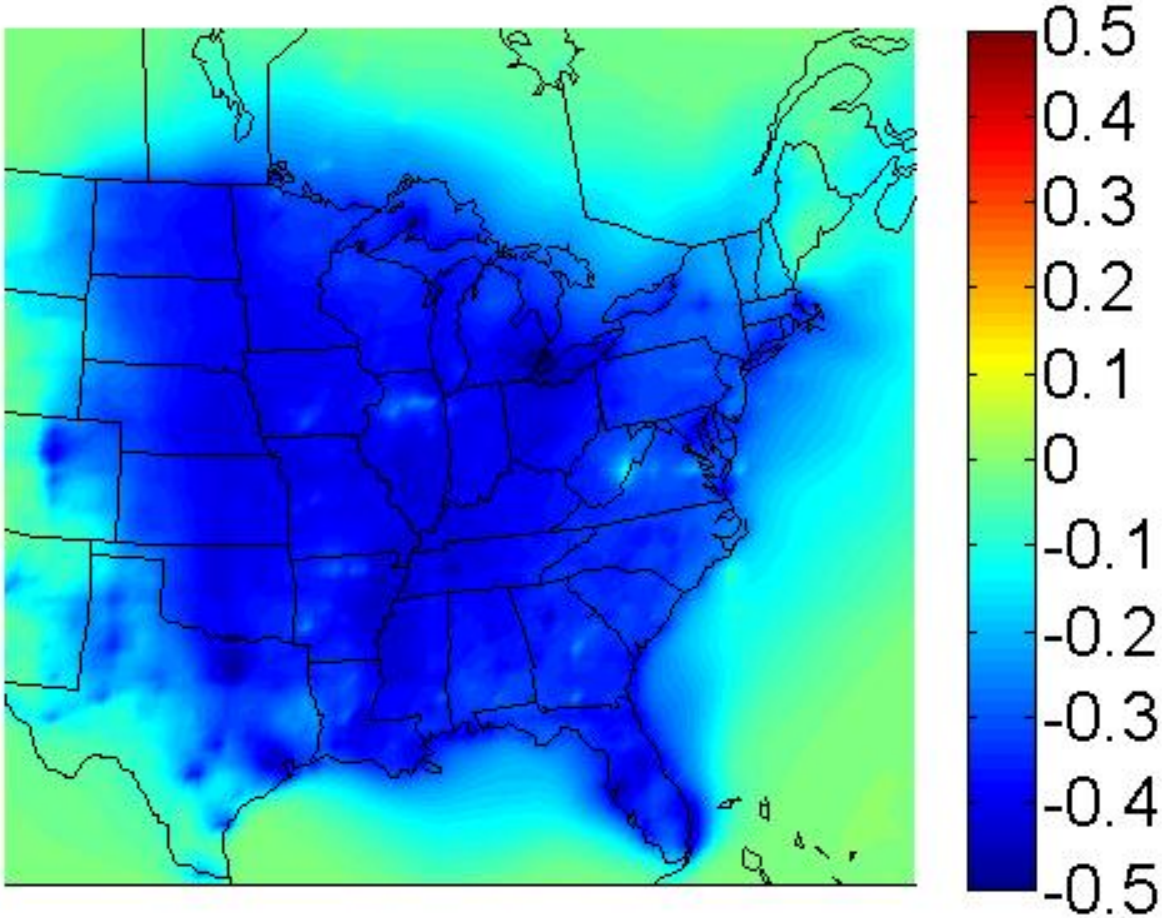
Fractional Change of EC



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

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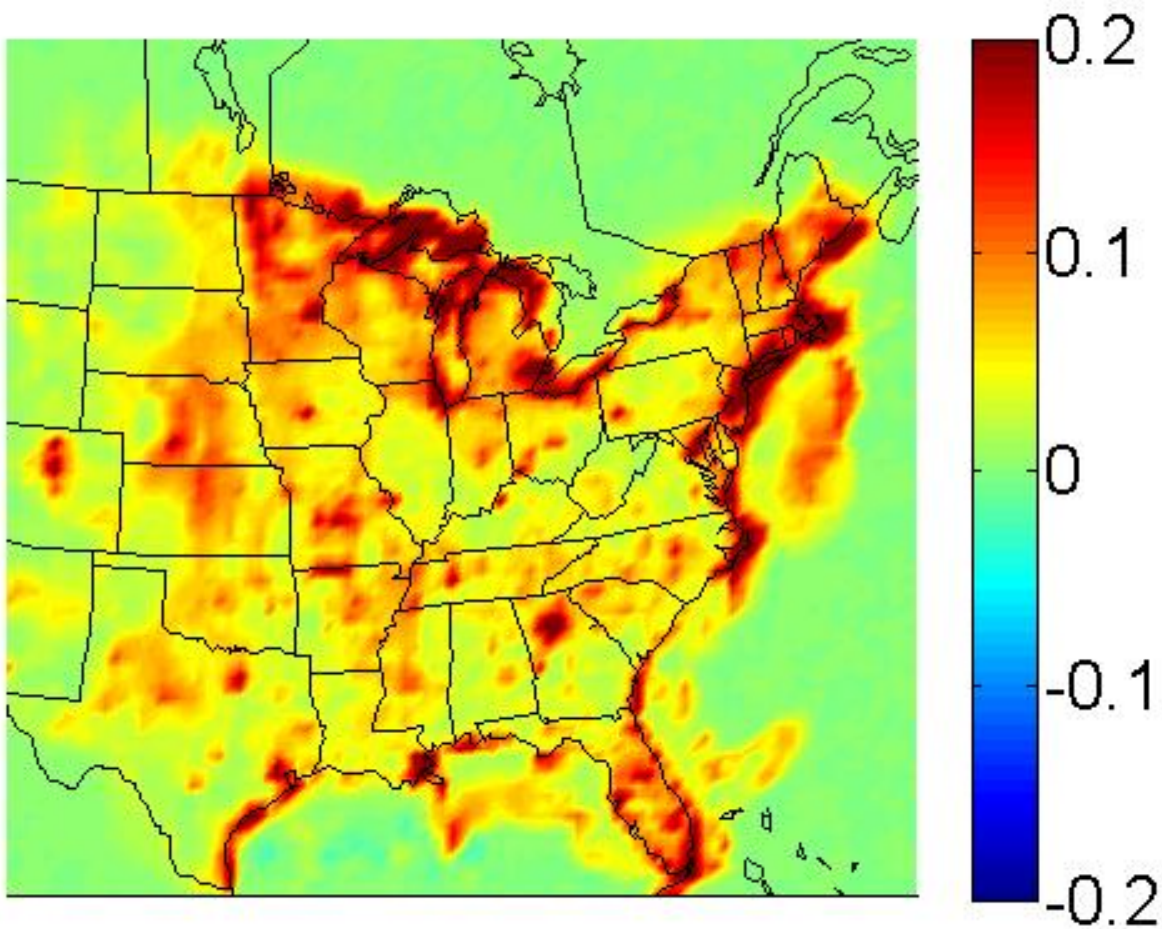
Fractional Changes of $N_{0.8\text{-}3}$



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

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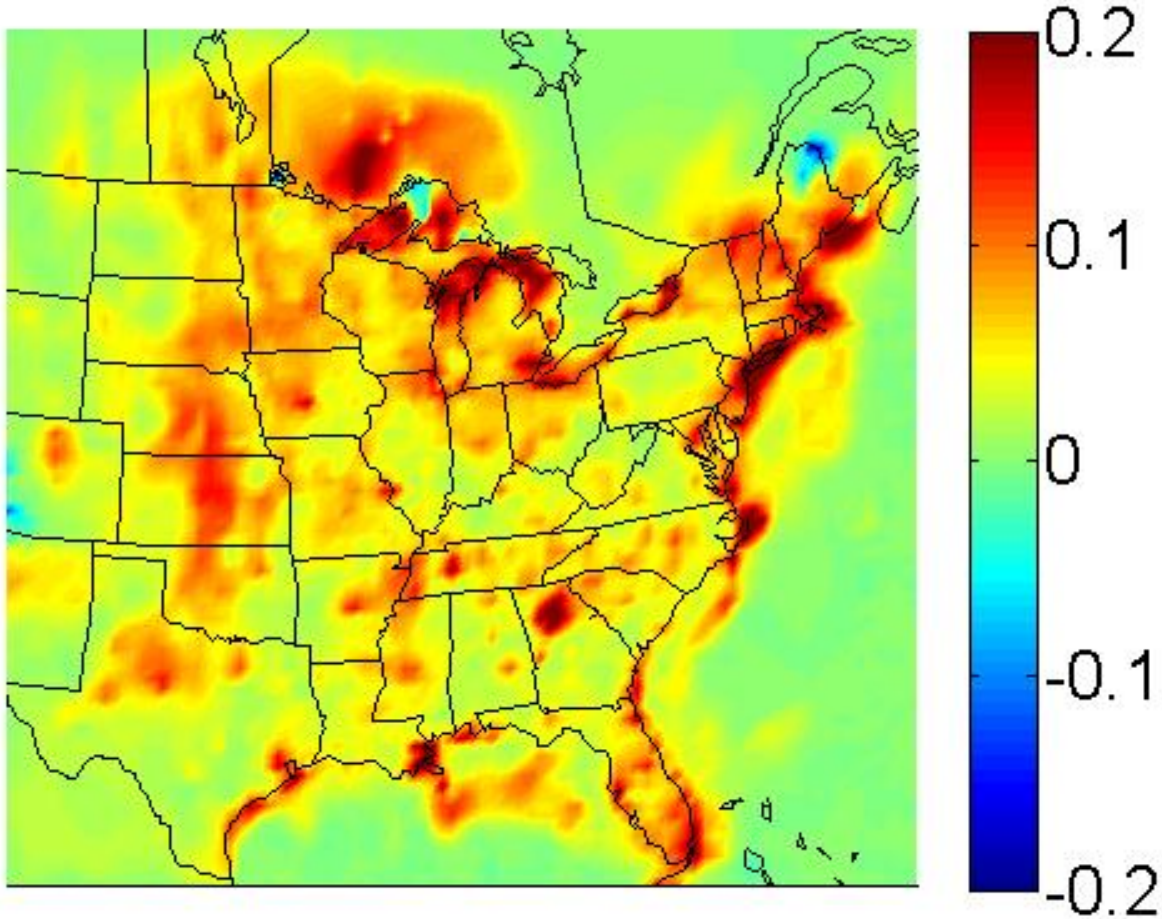
Fractional Changes of $N_{3\text{-}10}$



These increases also suggest that nucleation may increase and nucleated particles grow into this size range.

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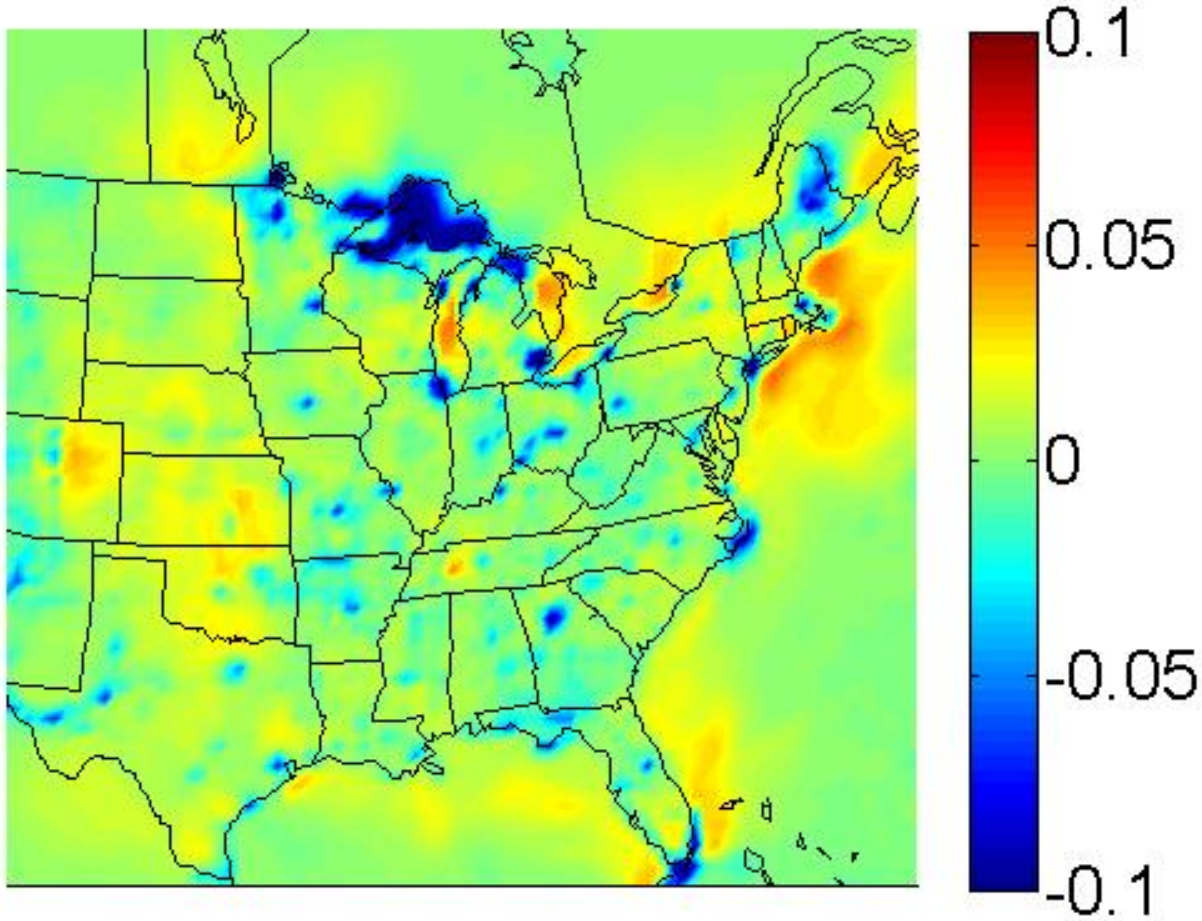
Fractional Changes of $N_{10\text{-}50}$



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

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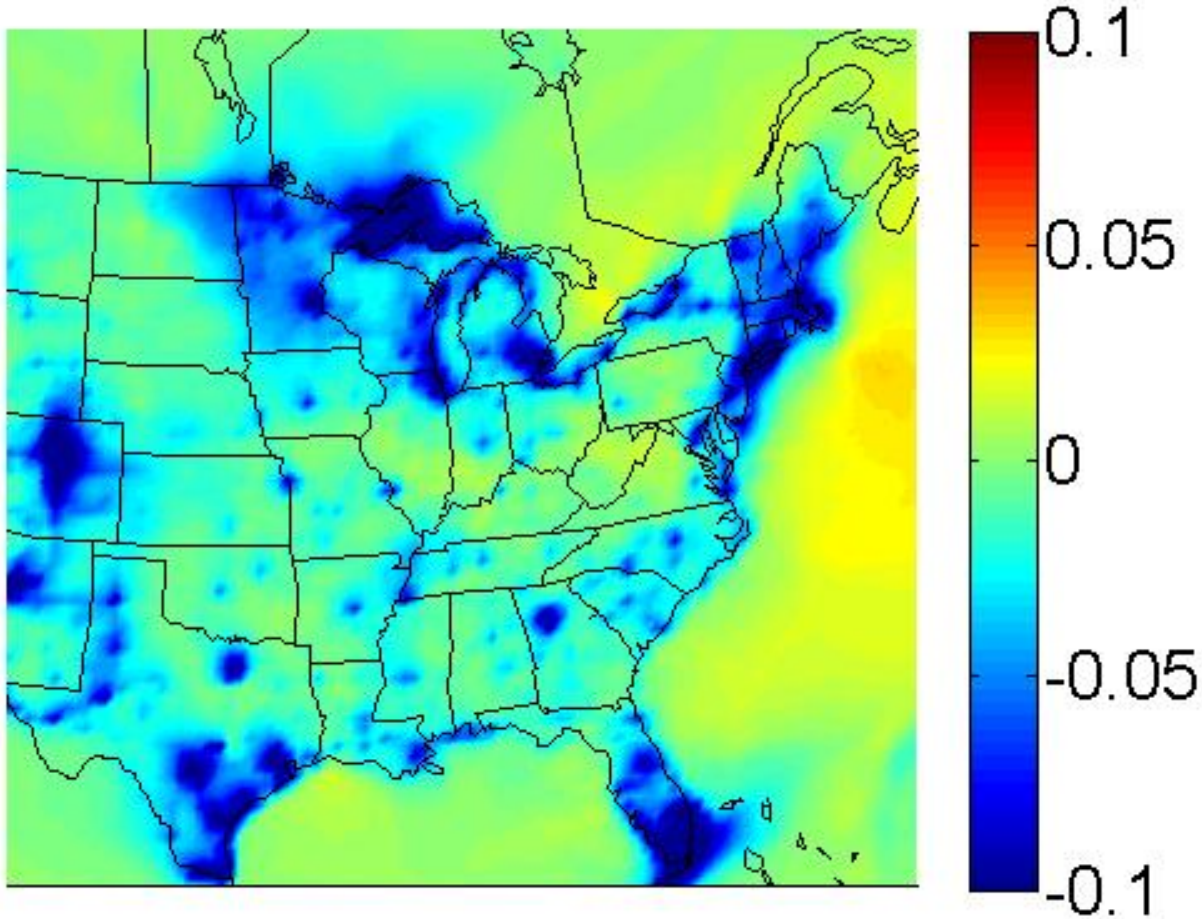
Fractional changes of $N_{50\text{-}100}$



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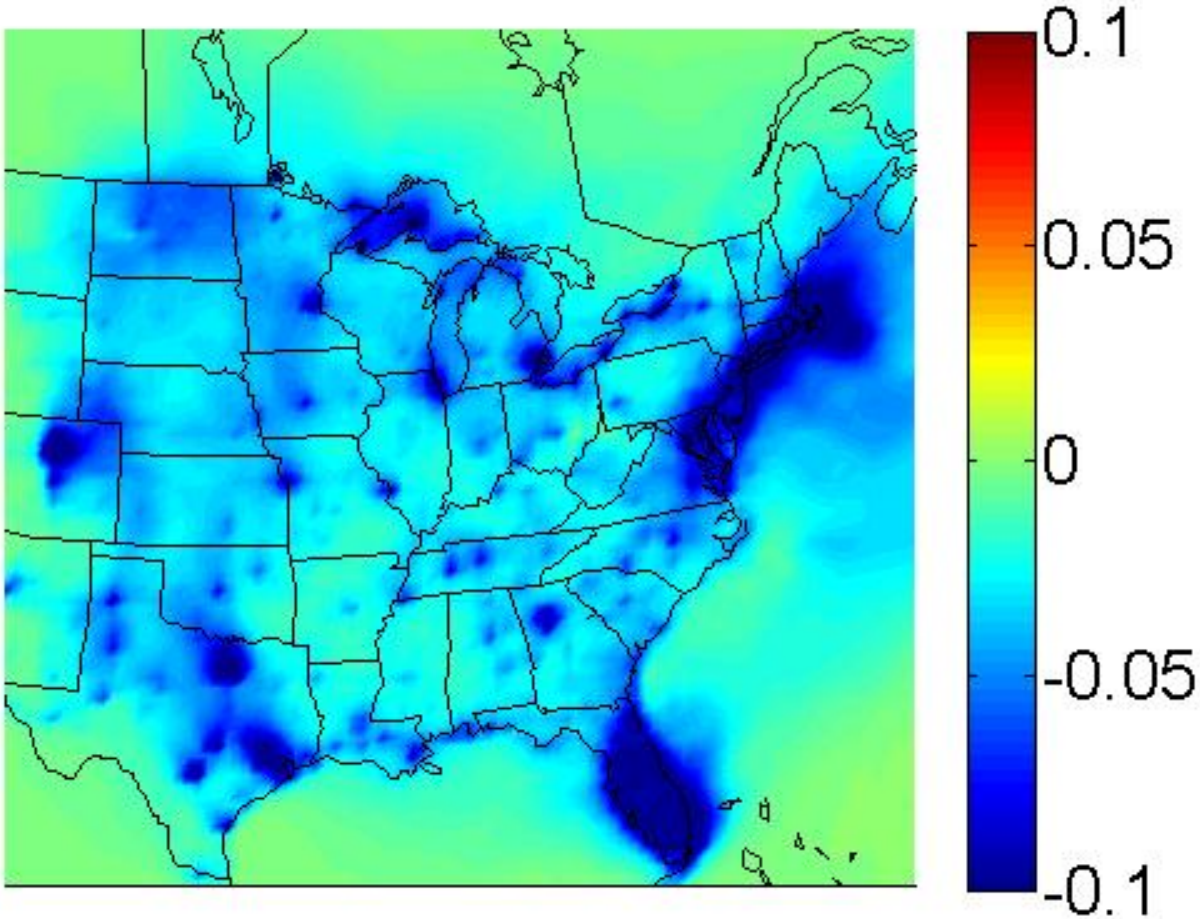
Fractional changes of N_{100}



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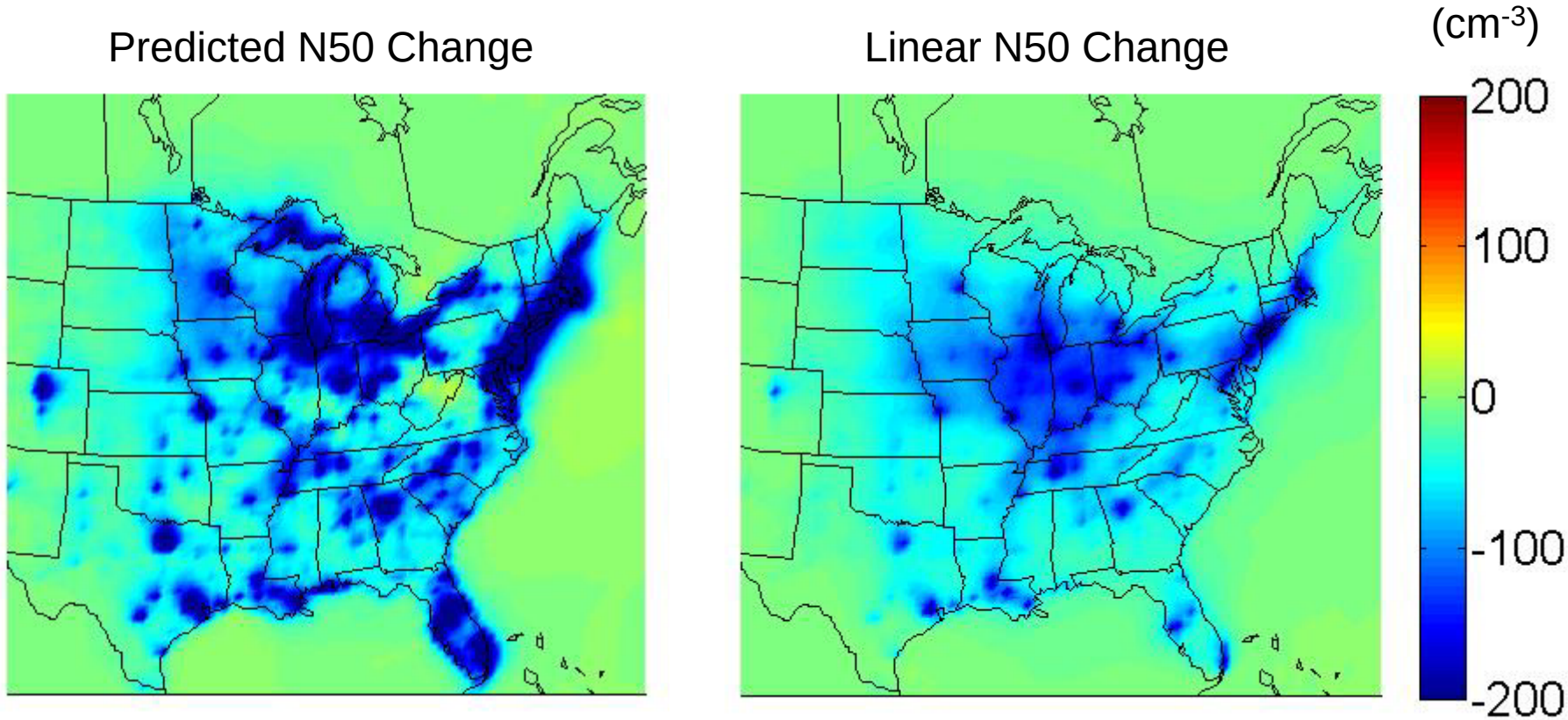
Non-linear Response of CCN to Diesel PM Controls



50-100% higher reduction in N50 and N100 than the linear response

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Improving Regional Scale BC Models

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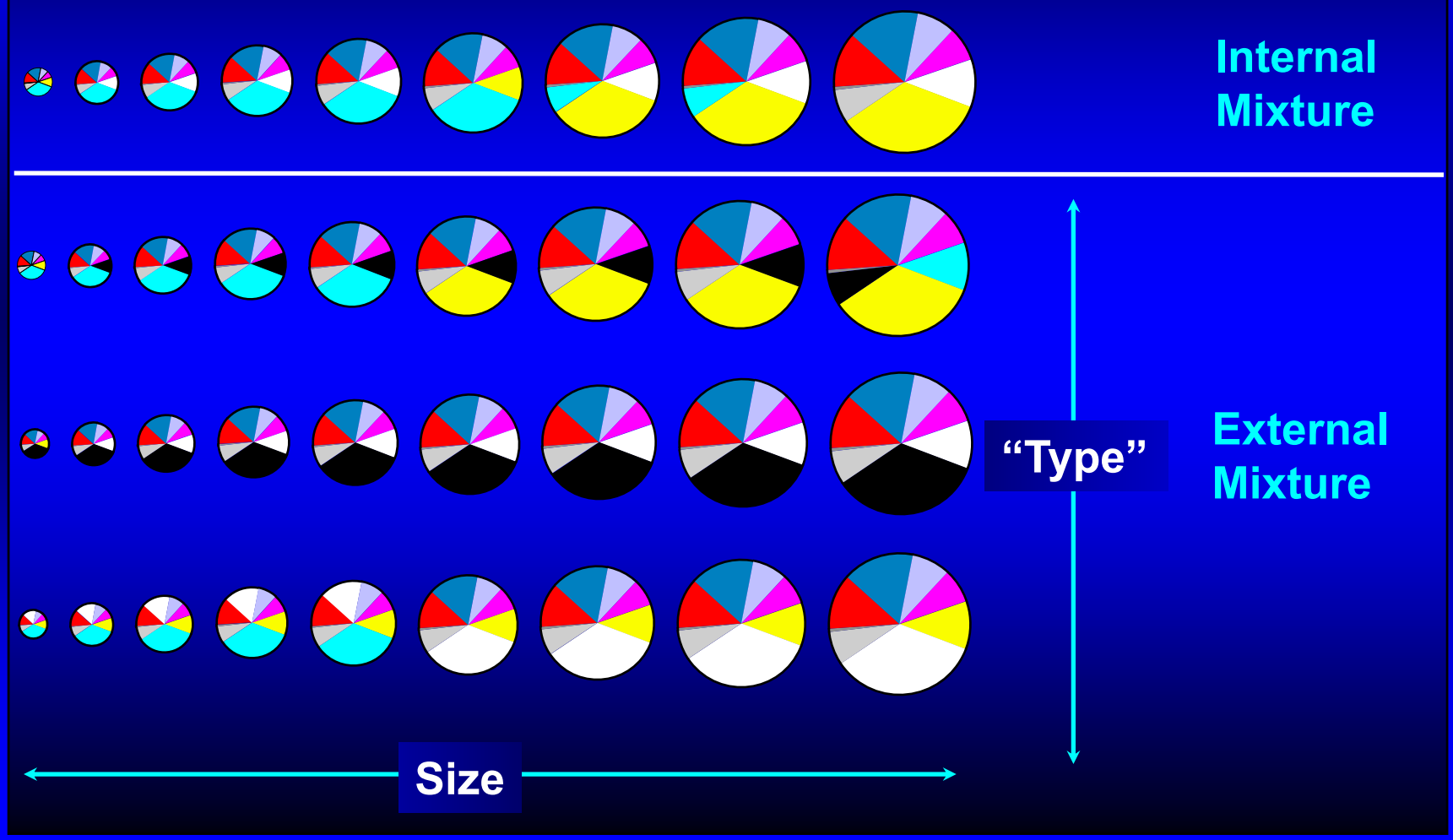
Relaxing the Internal Mixture Assumption



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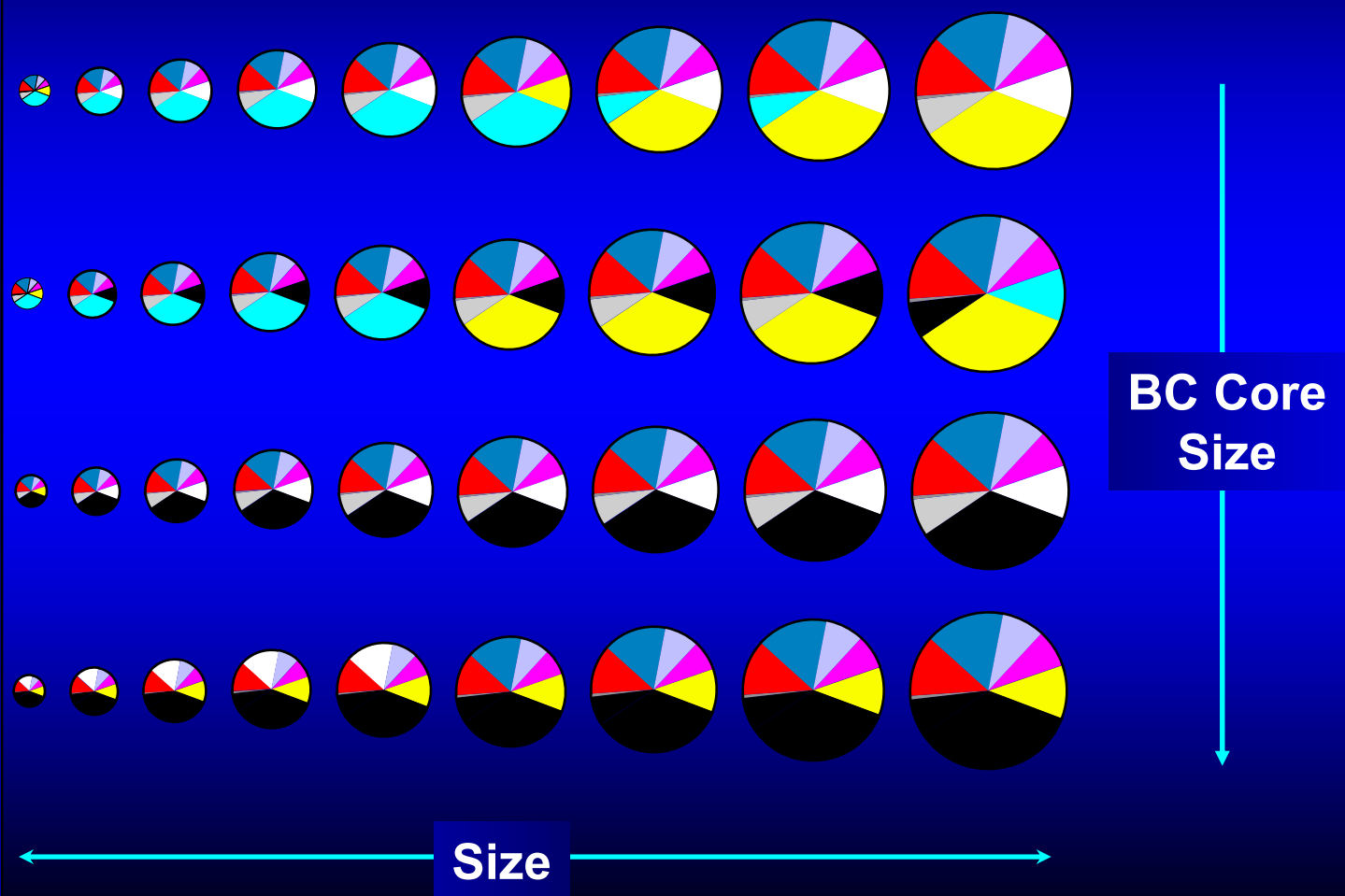
Simulating BC Mixing State In PMCAMx



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Conclusions

- 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
- This effect was not observed in diesel emissions
- 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
- Estimated radiative forcing of 0.1-0.2 W m^{-2} due to biomass burning BrC.
  - Net effect of biomass burning is still cooling.

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Conclusions

- New particle number source apportionment algorithm (SANSA) for TOMAS (used in PMCAMx, GISS-II' and GEOS-CHEM)
- 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

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  Laura Posner, Christos Fountoukis, Antonis Tassoglou.

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