Black Carbon, Air Quality and Climate

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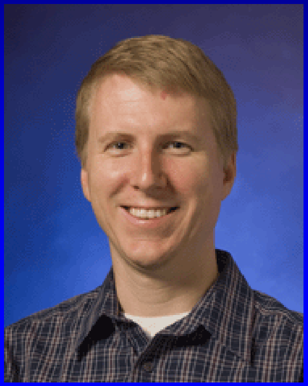

Peter Adams

Rawad Saleh

R. Subramanian

Black Carbon, Air Quality and Climate

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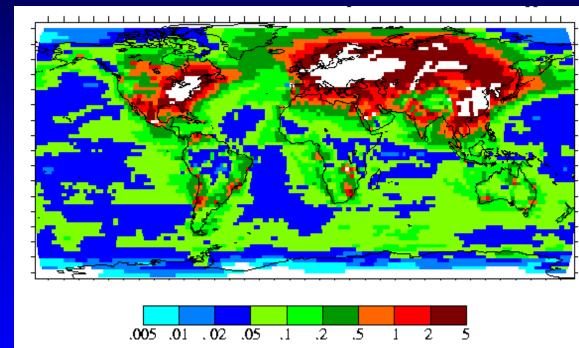


Sources of BC

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

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

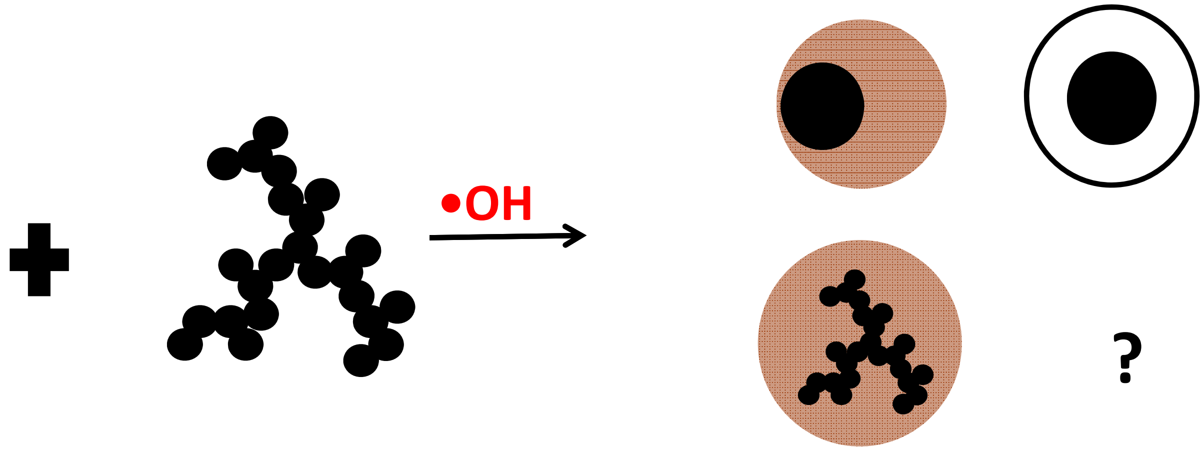
BC Emissions, Chemical Aging, and Optical Properties

Combustion Emissions

R. Saleh

BC particles act as condensation sites for OA

Organic Compounds

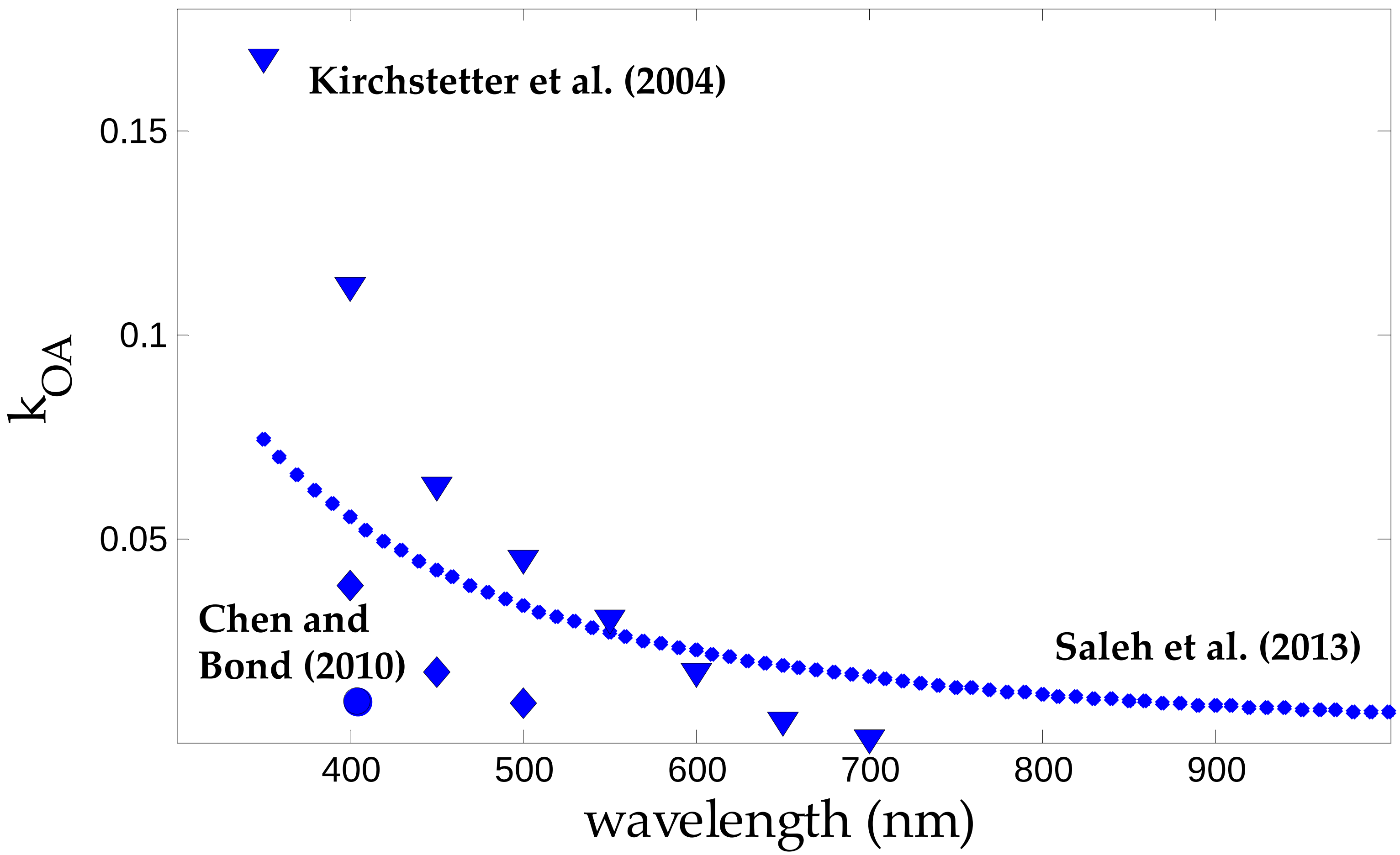


Black Carbon

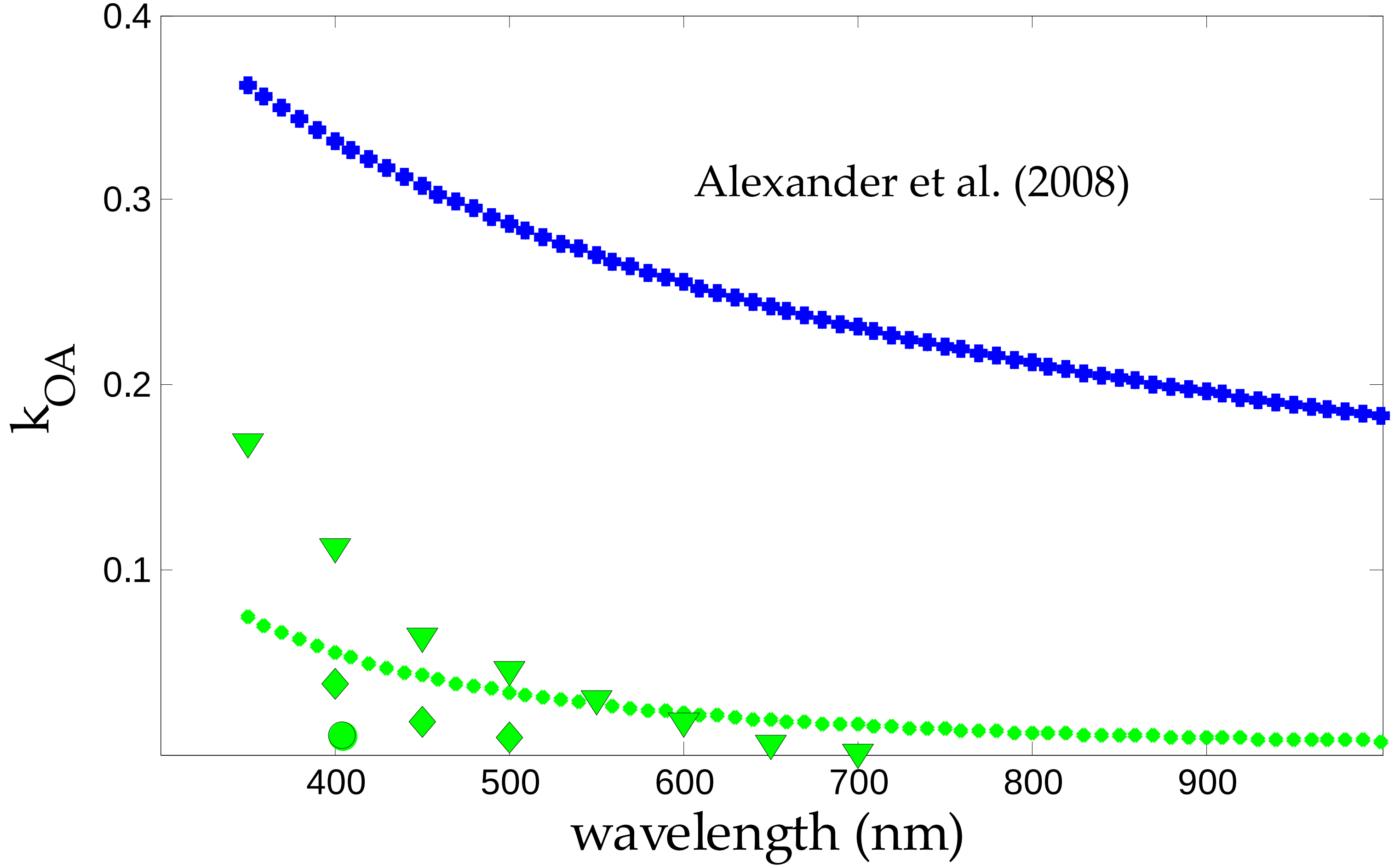
Core –Shell morphology

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

BrC in Biomass Burning: Chaos



BrC in Biomass Burning: More Chaos



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

Aethalometer

OA/BC from biomass burning

SP2

PASS-3

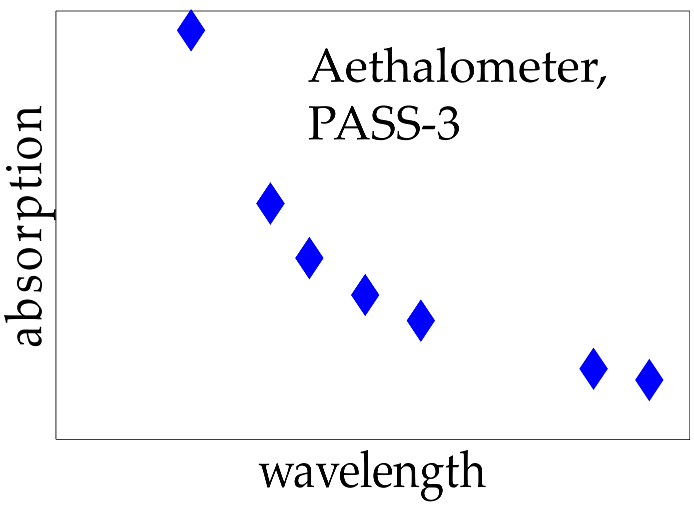
HR-SP-AMS

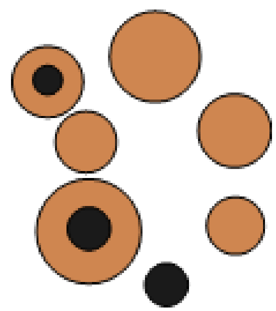
Estimation of OA Optical Properties

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

Bond et al. [2005]

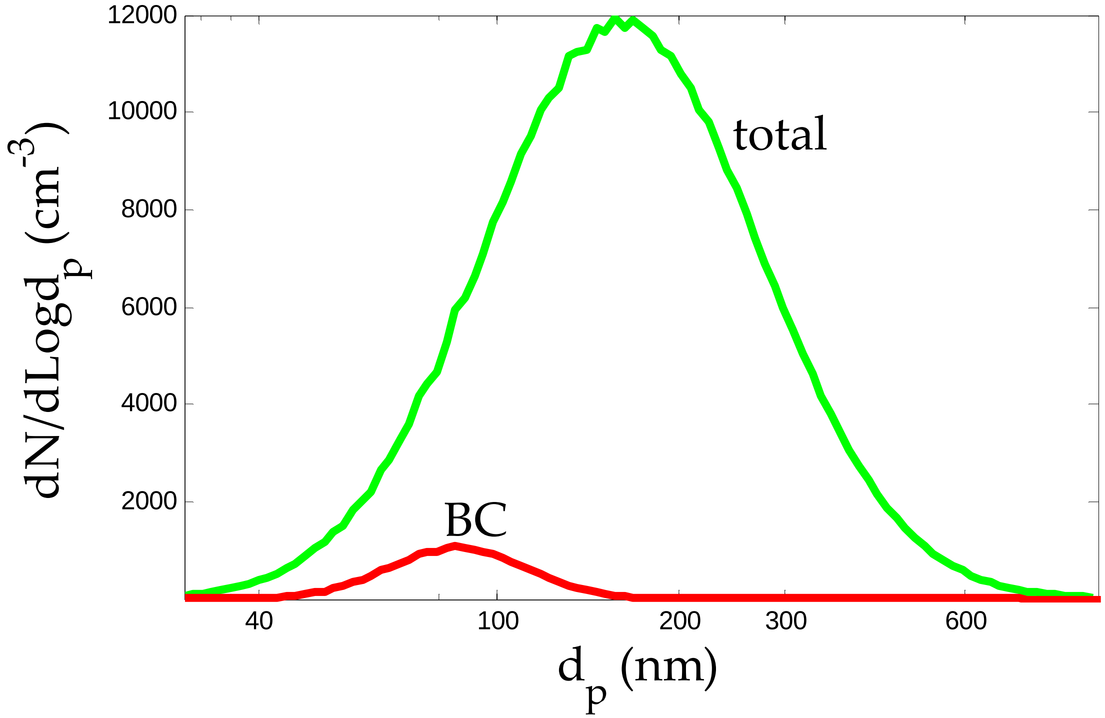
$$k_{OA}(\lambda) = k_{OA,550}\left(\frac{550}{\lambda}\right)^{w}$$



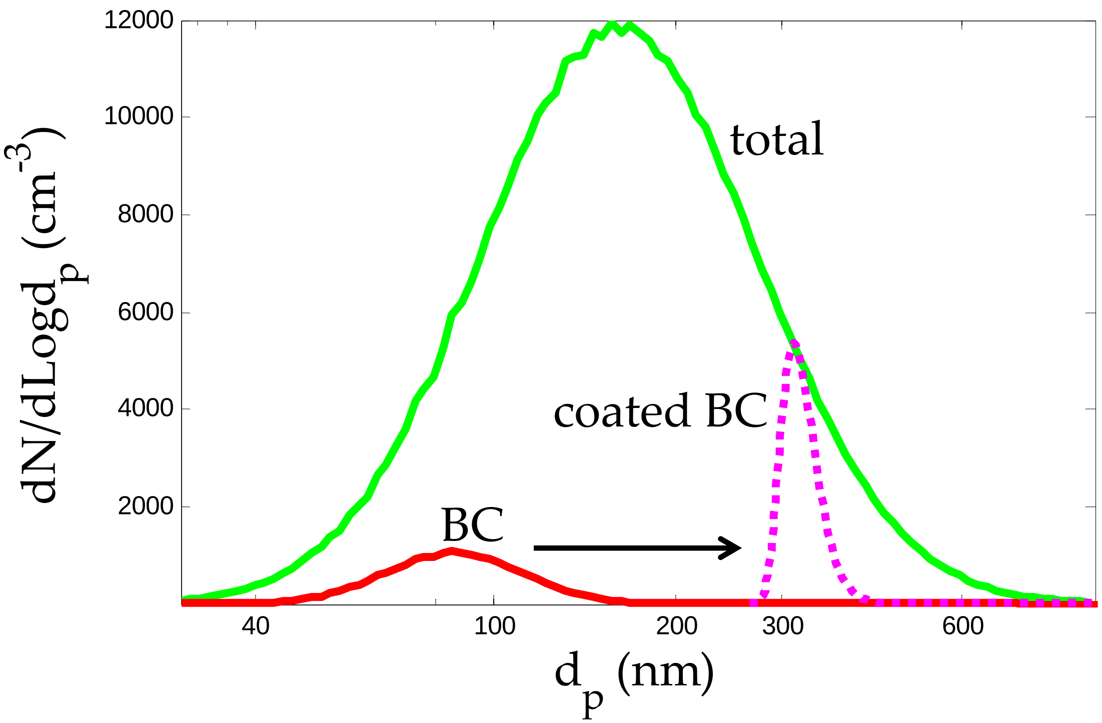


SMPS,
SP2

Morphology and Mixing State

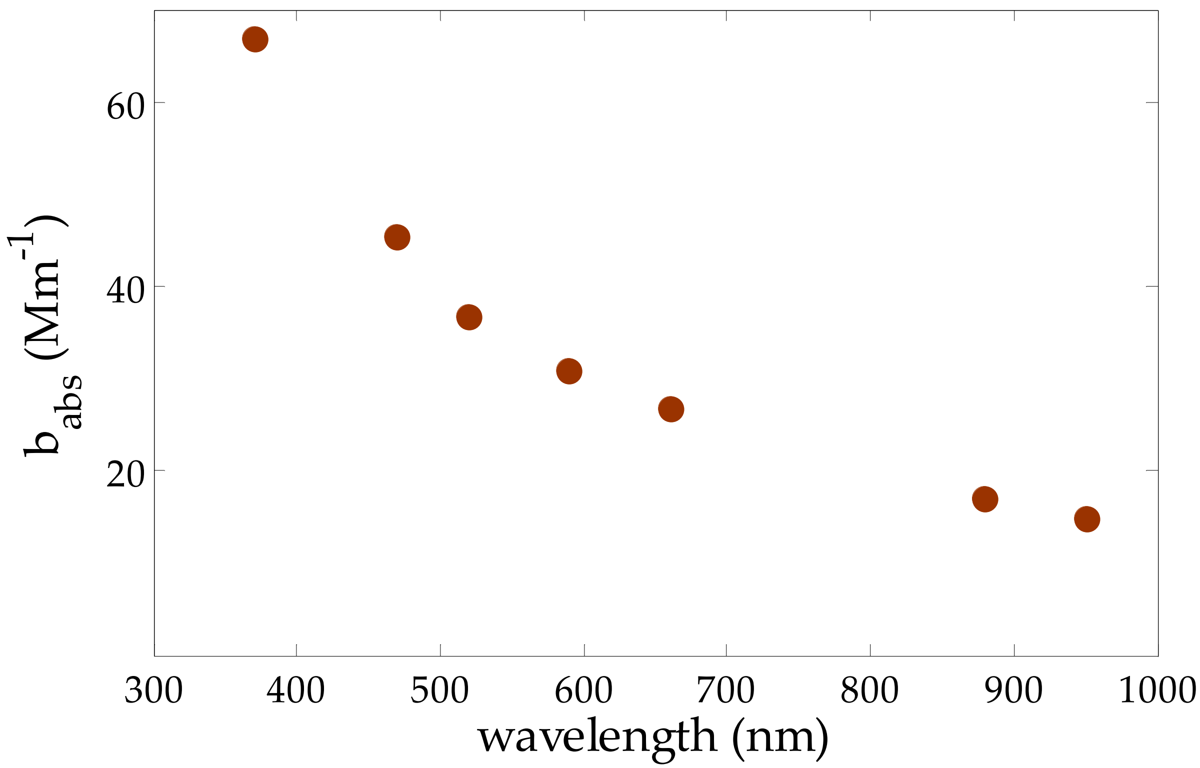


Morphology and Mixing State



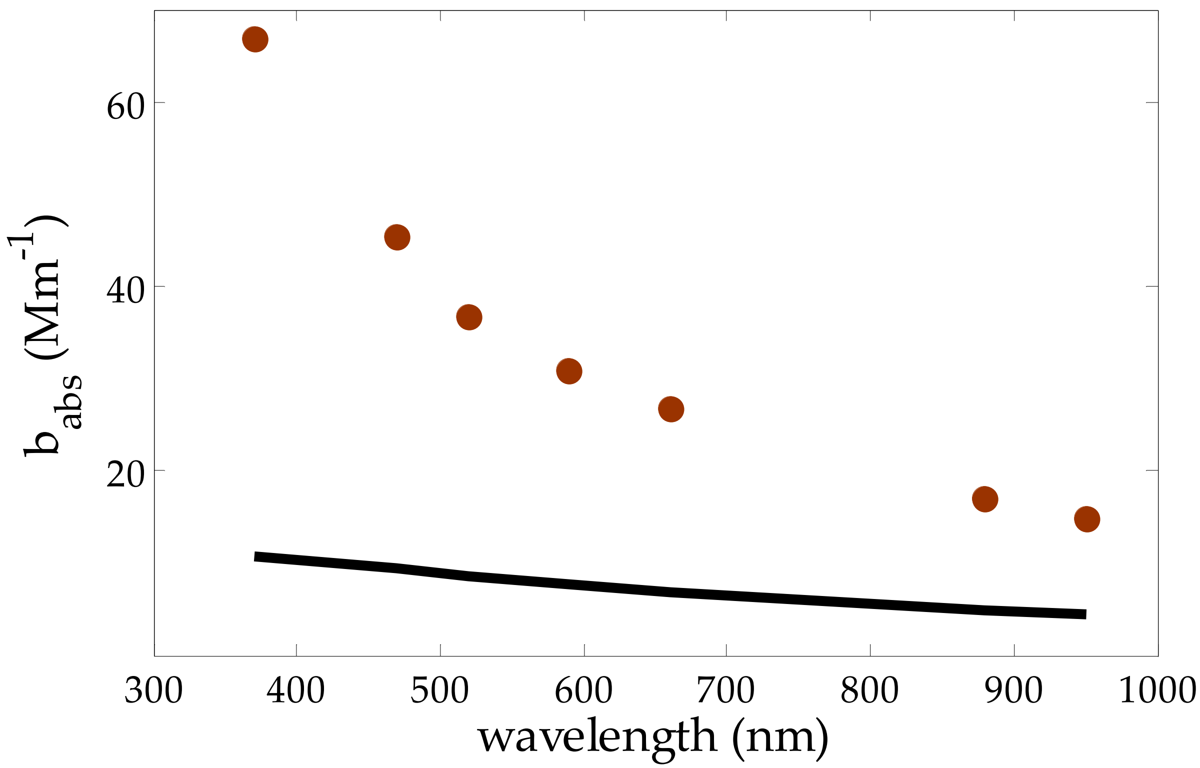
- We simulate the condensation process of OA on BC.
- The growing distribution cannot go beyond the SMPS distribution.
- We can only constrain the maximum coating thickness.
- This maximizes the lensing effect, thus minimizes BrC absorption
- Conservative approach.

The Fit

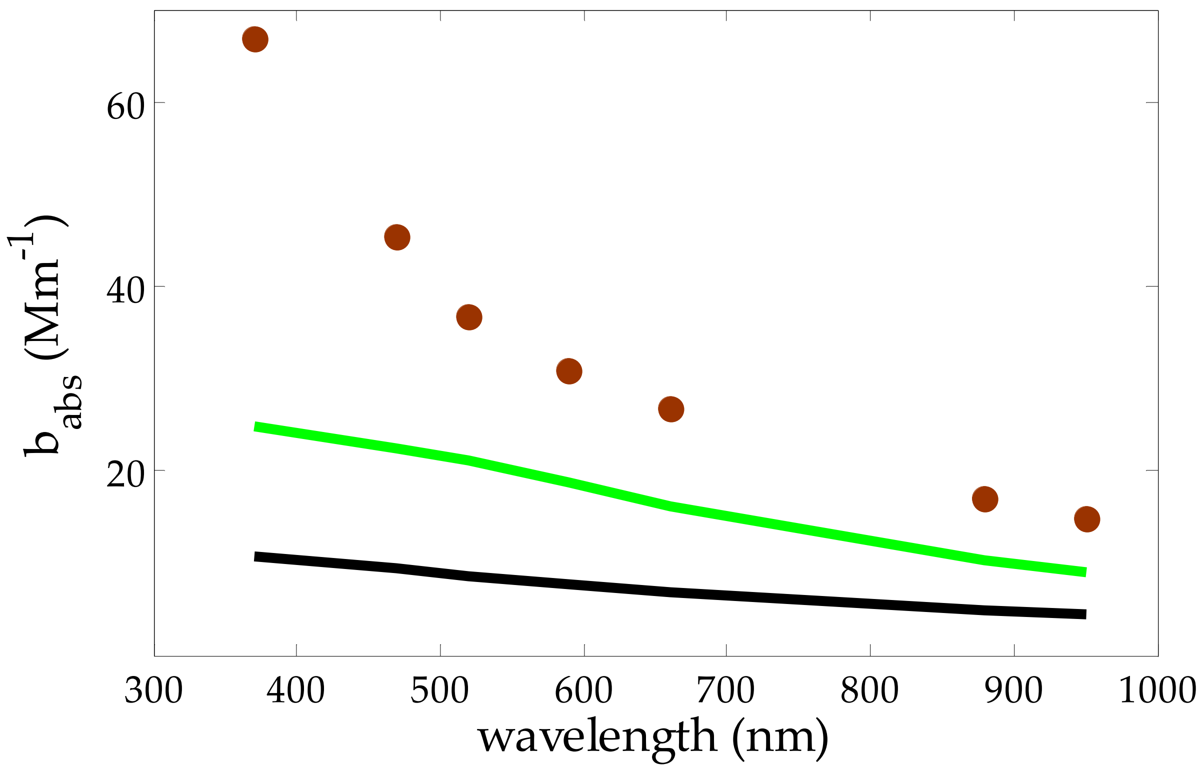


60
40
20
300
400
500
600
700
800
900
1000
wavelength (nm)
b_{abs} (Mm^{-1})

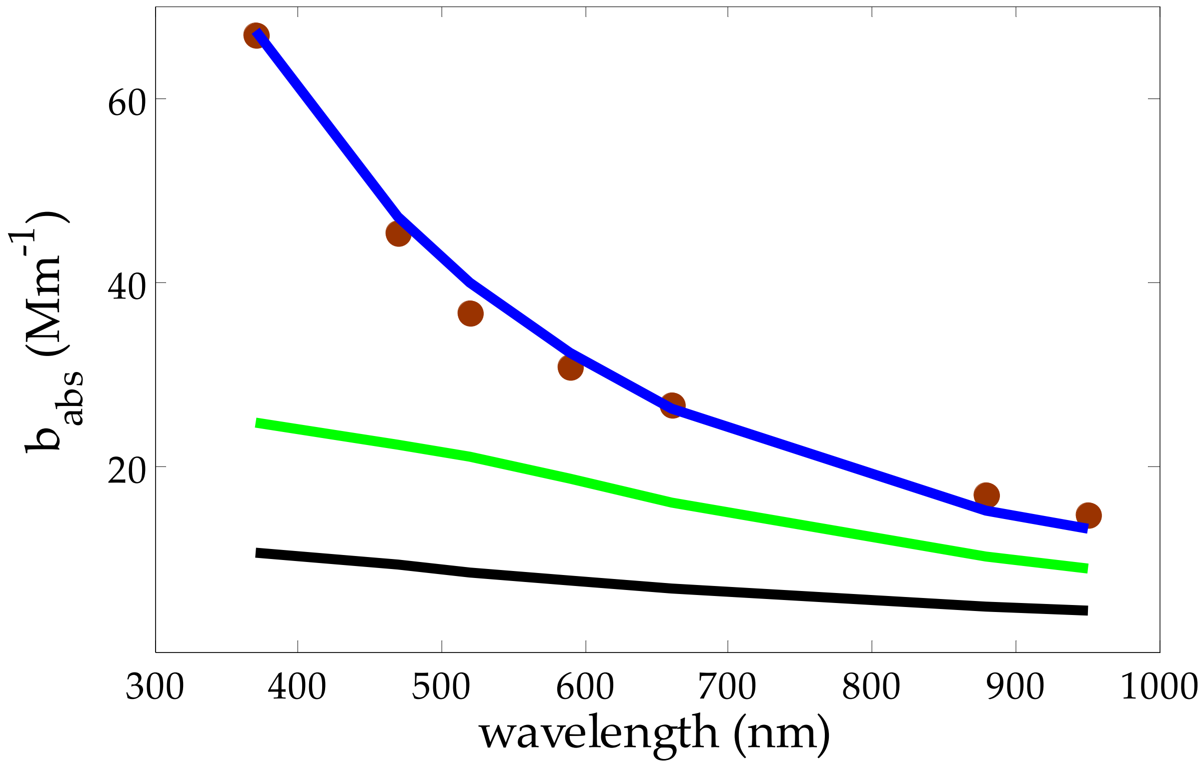
The Fit (Absorption due to BC only-Mie calculations)



The Fit (Absorption due to BC+ Lensing)

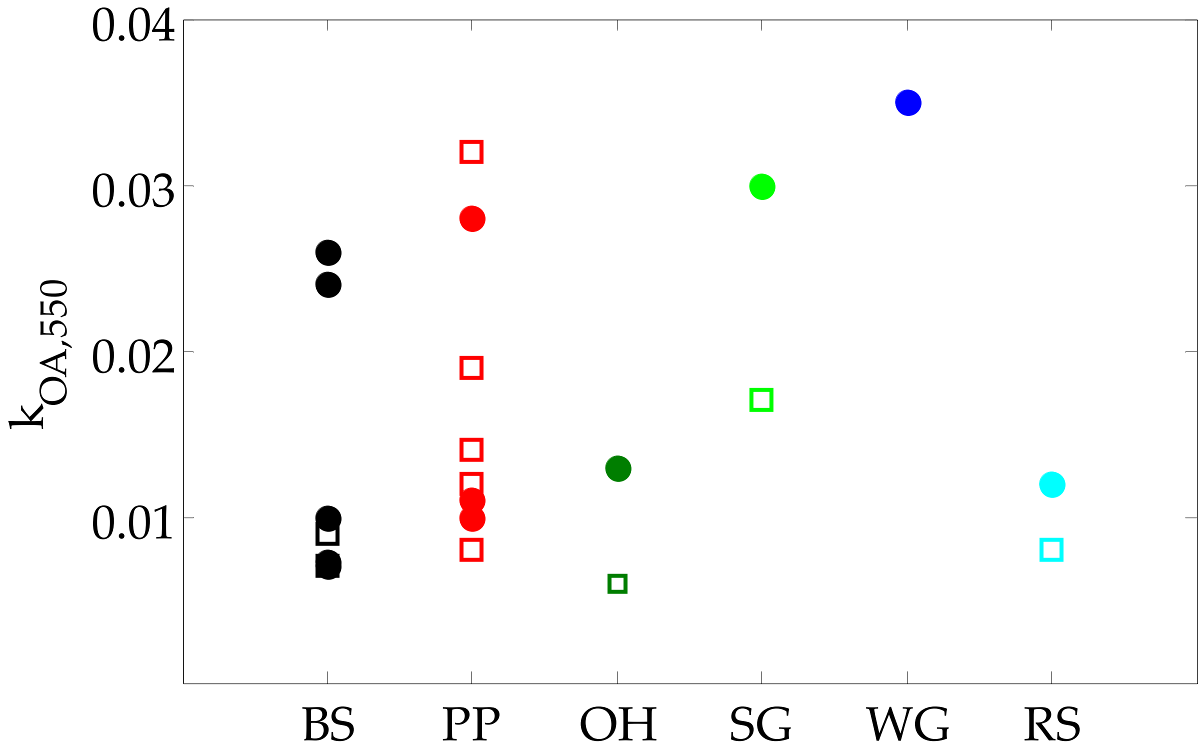


The Fit (Absorption due to BC+ Lensing+BrC)



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

Chaos Returns !

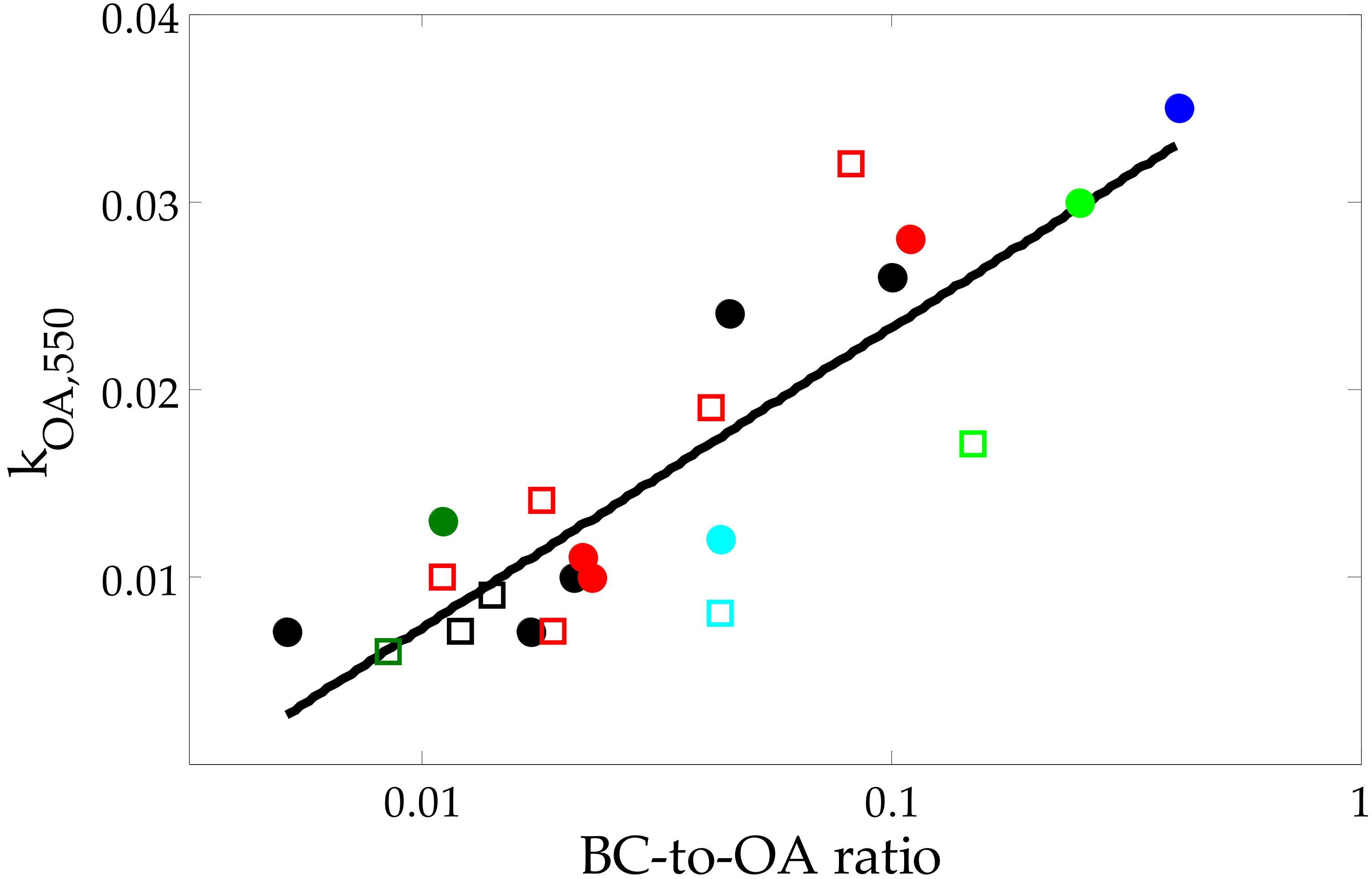


FLAME IV

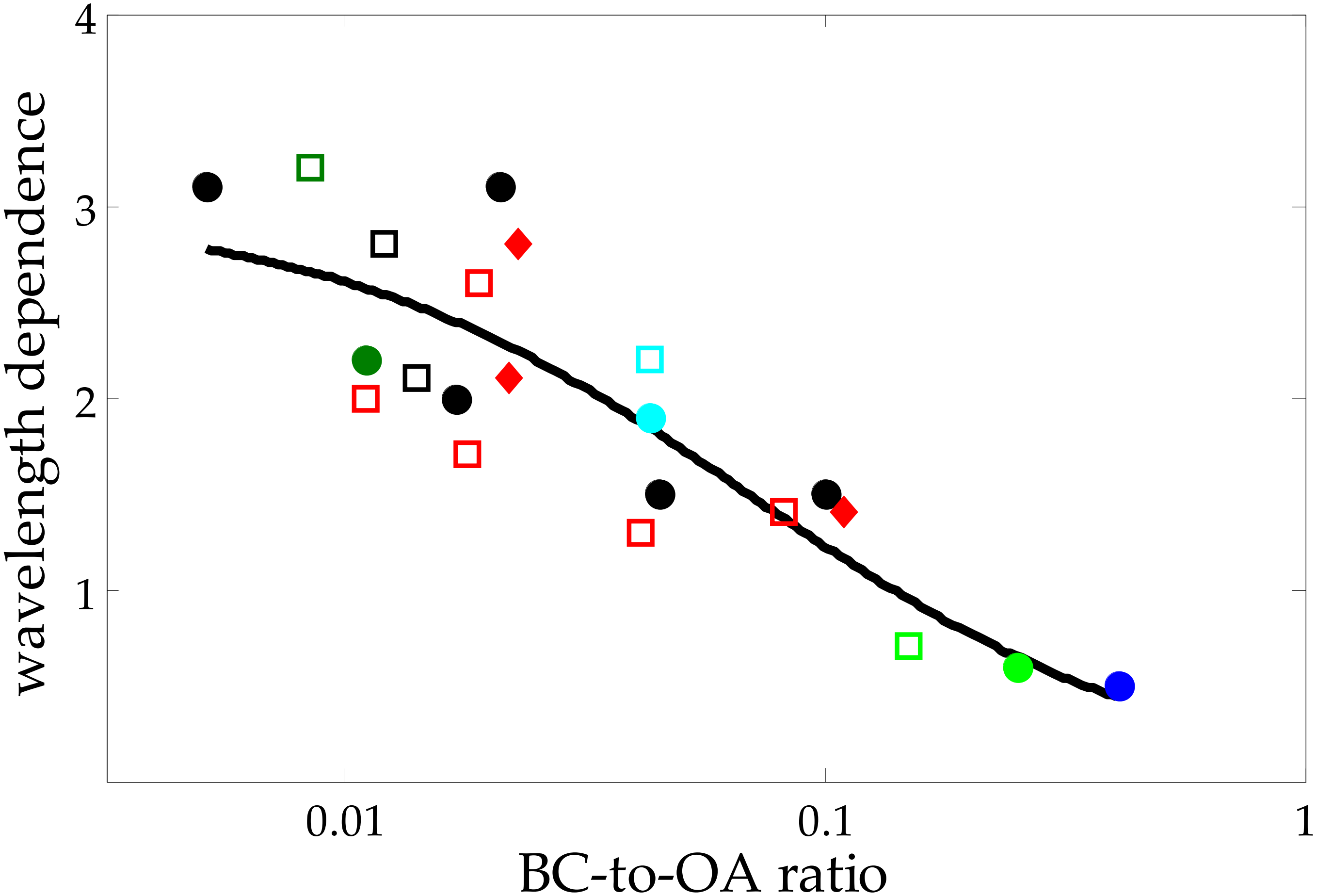
BS: black spruce
PP: ponderosa pine
OH: organic hay
SG: saw grass
WG: wire grass
RS: rice straw
Closed symbols: fresh
Open symbols: aged

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

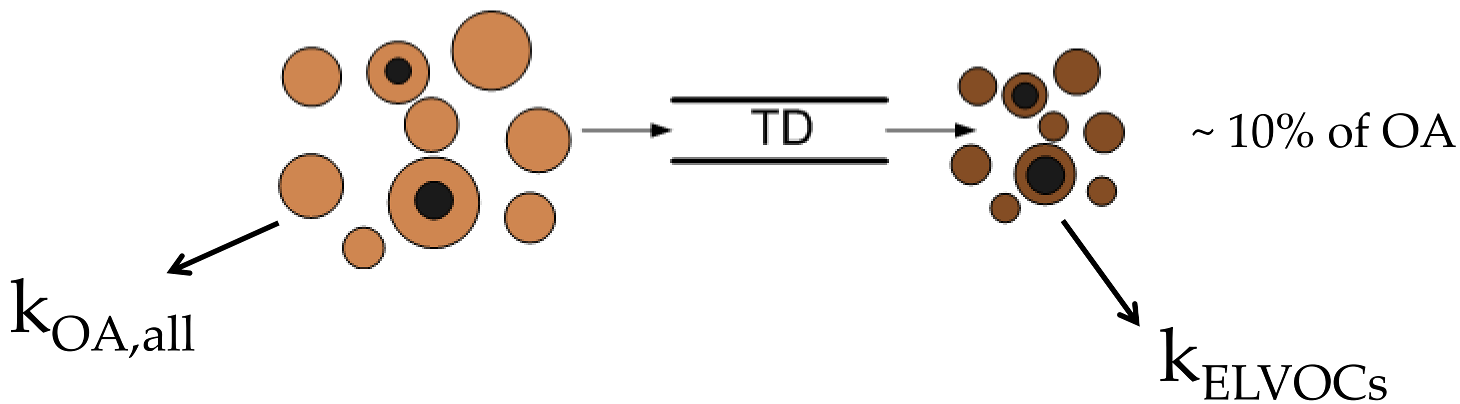
Some Order



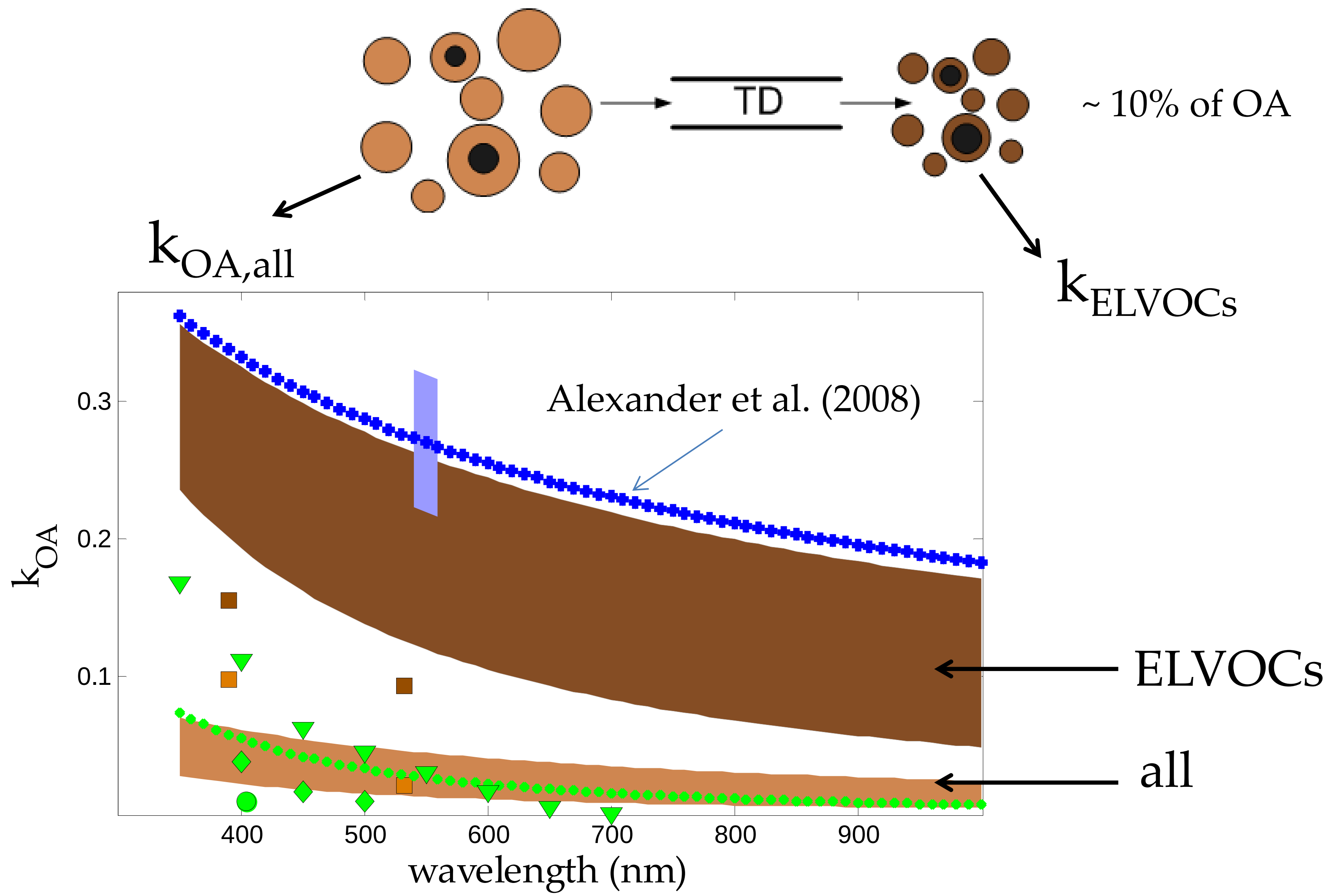
Some Order



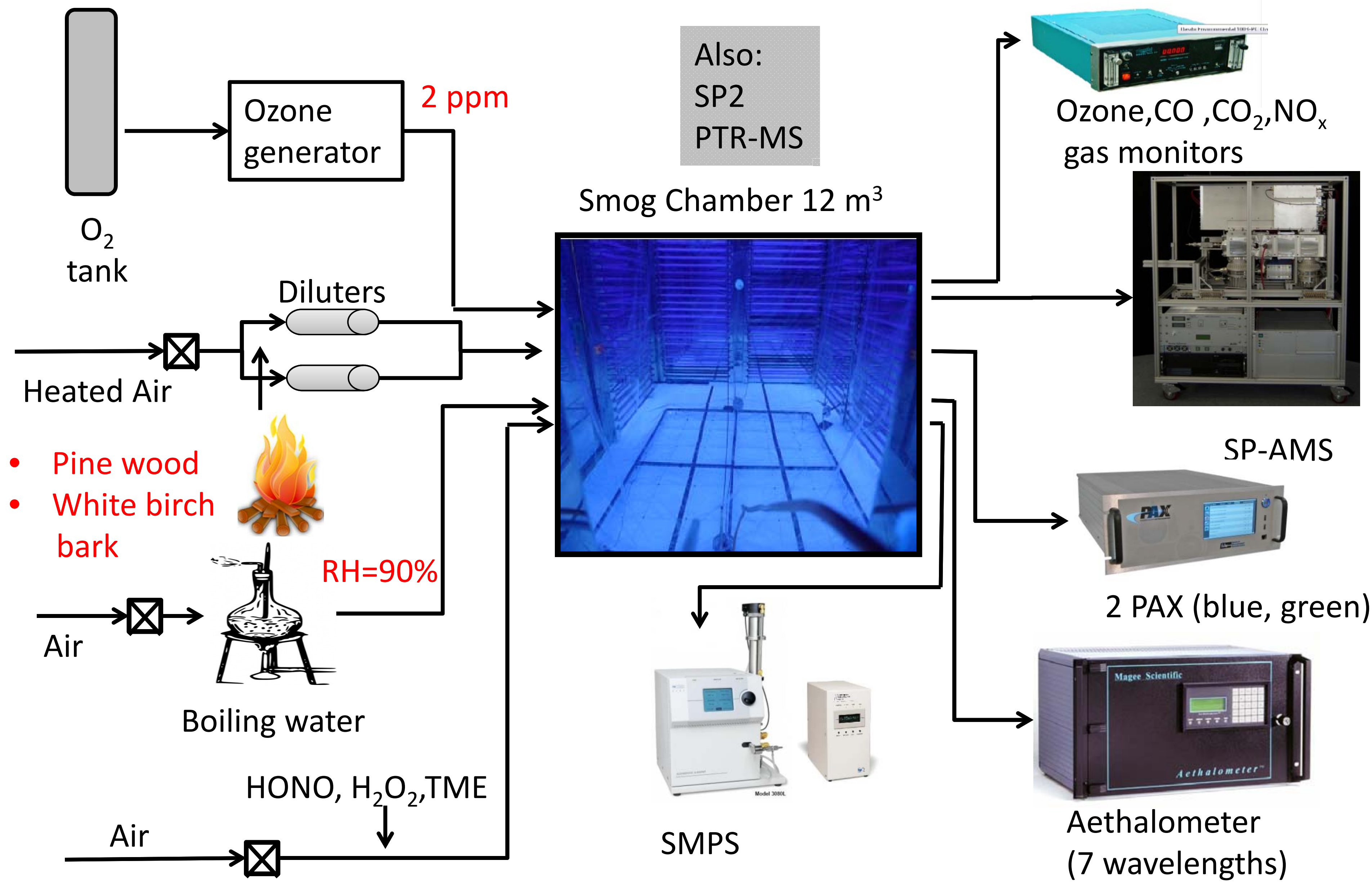
BrC and OA Volatility



BrC and OA Volatility

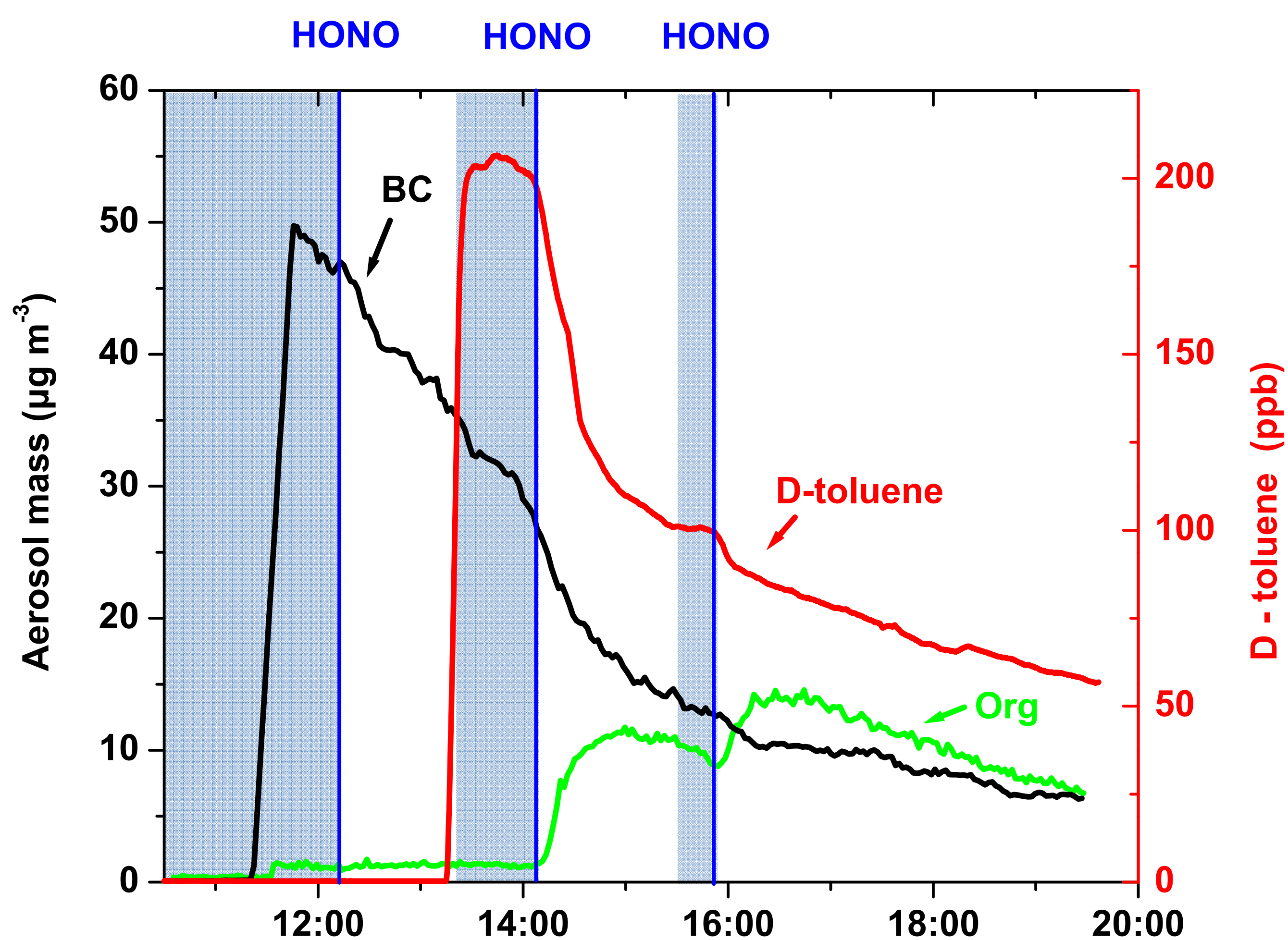


CMU Smog Chamber



Coating of BC with D-toluene SOA

(fuel: White birch bark)

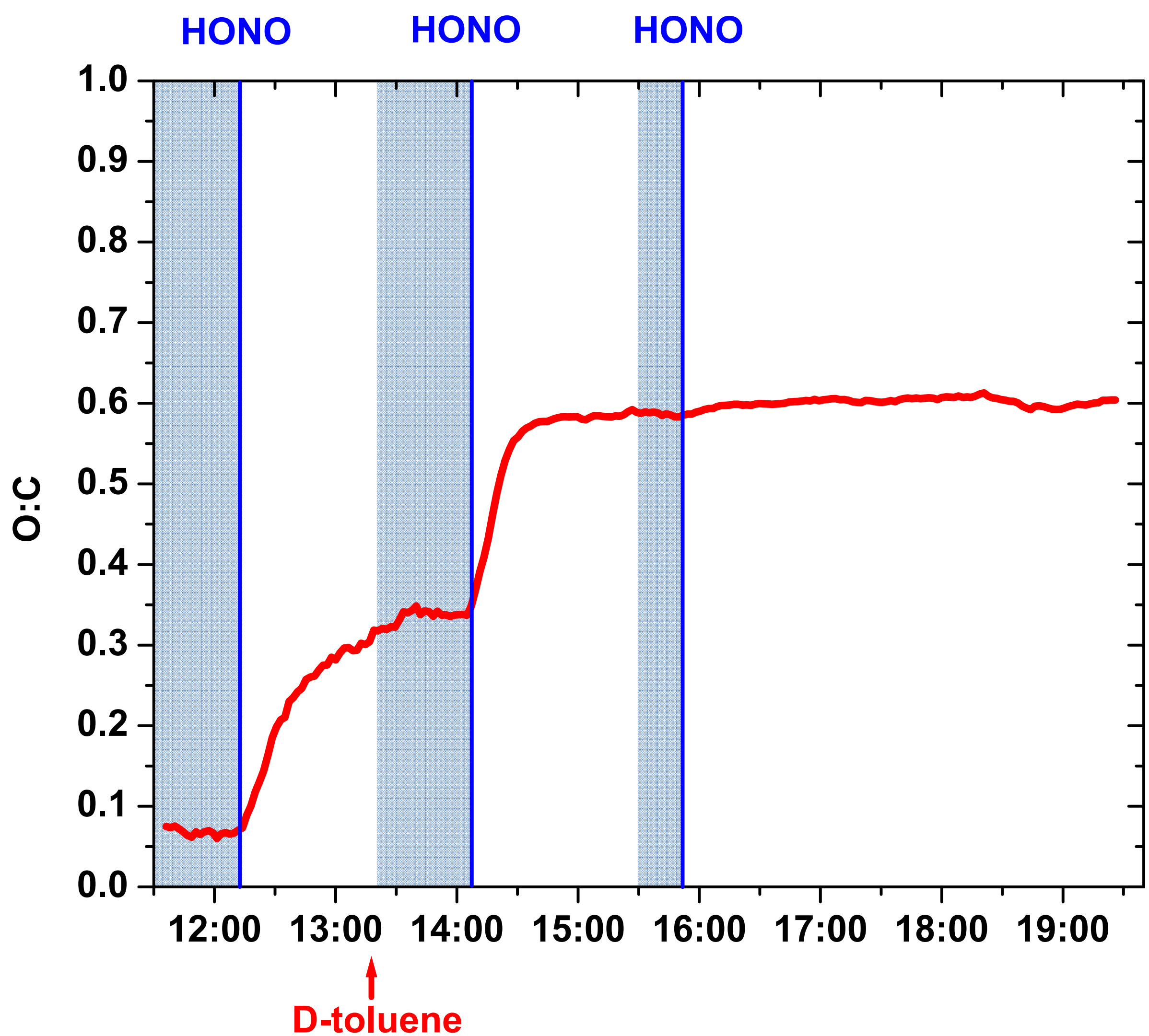


CE_{BC}= 0.4

CE_{org}=1

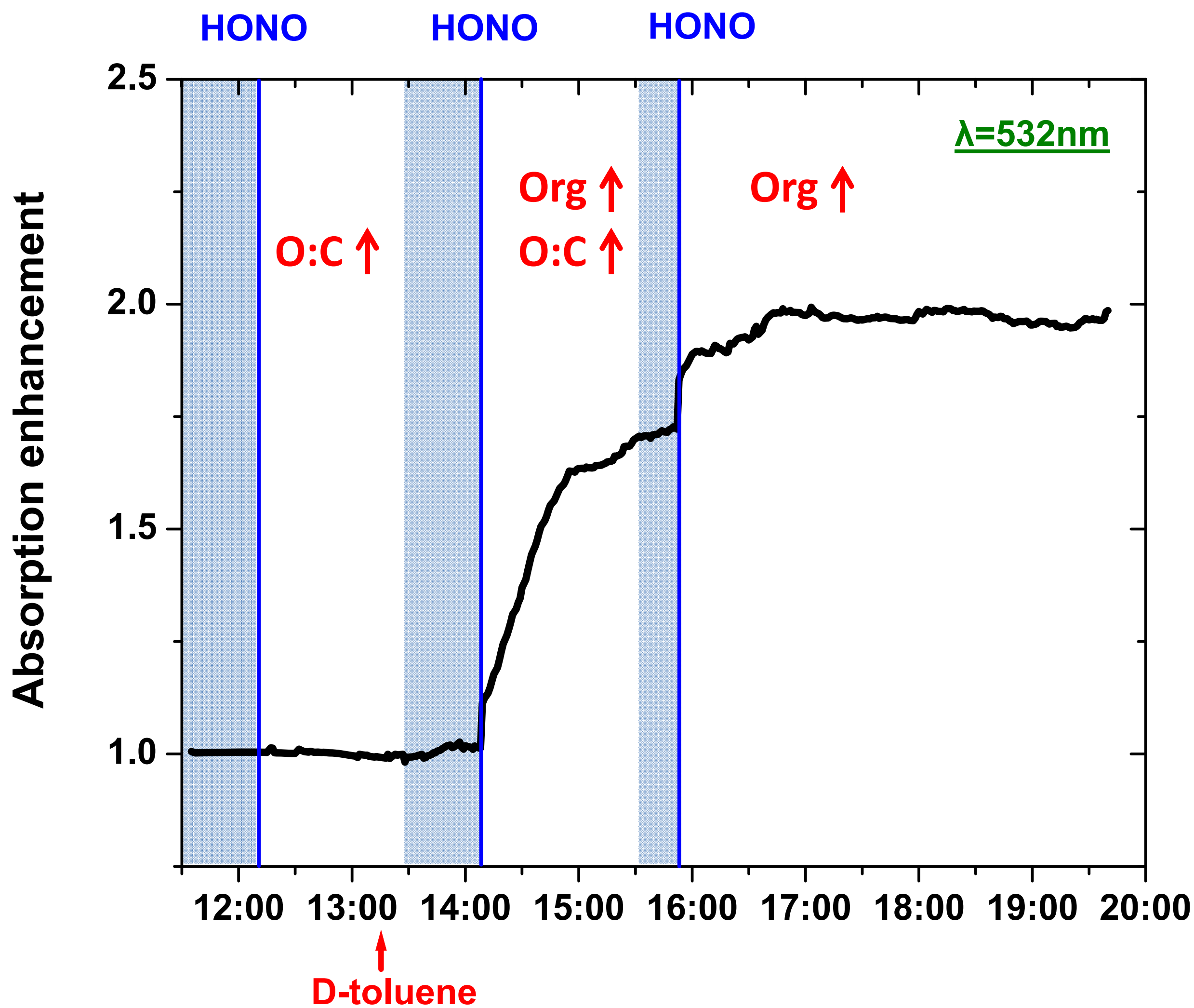
O/C during D-toluene SOA formation

(fuel: White birch bark)



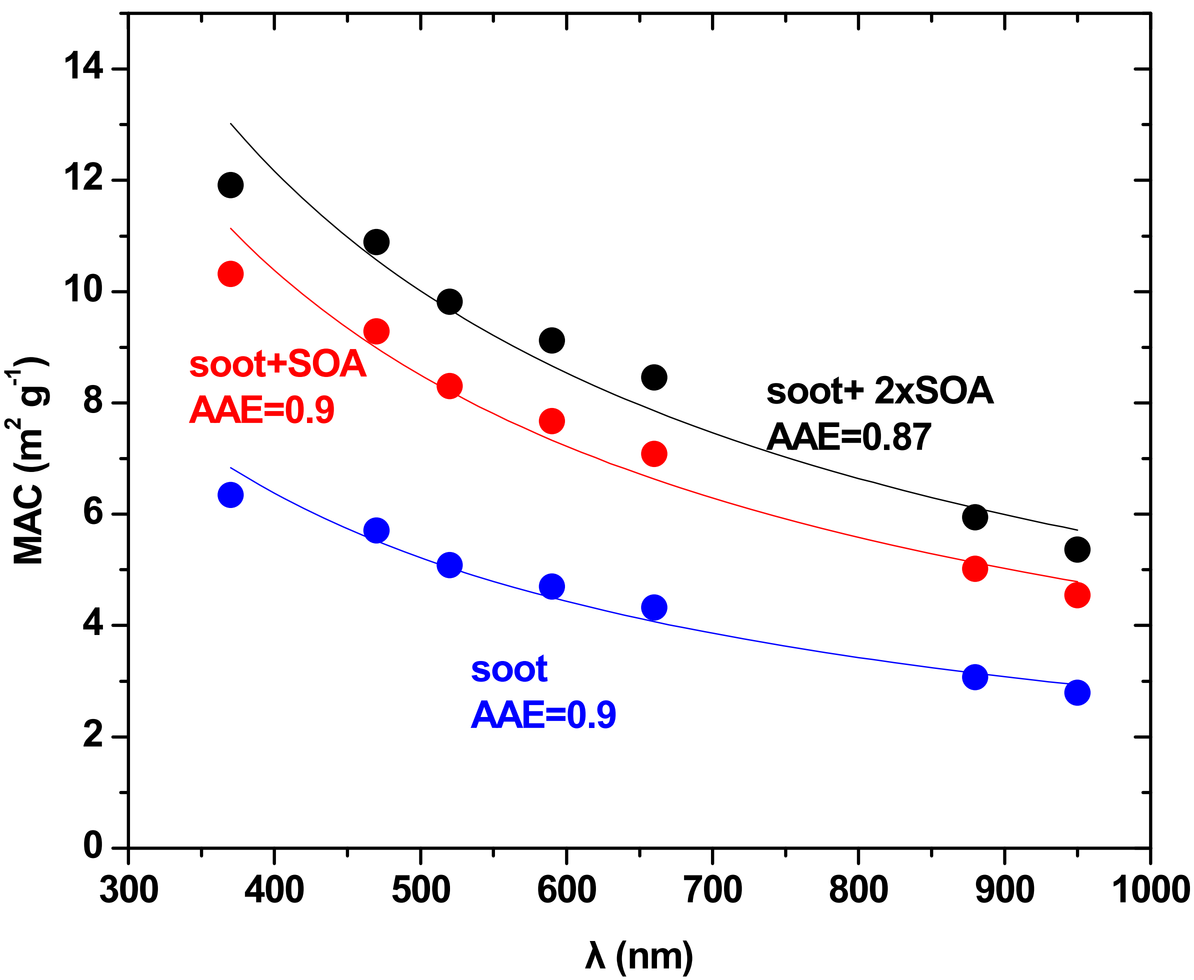
Absorption during D-toluene SOA formation

(fuel: White birch bark)



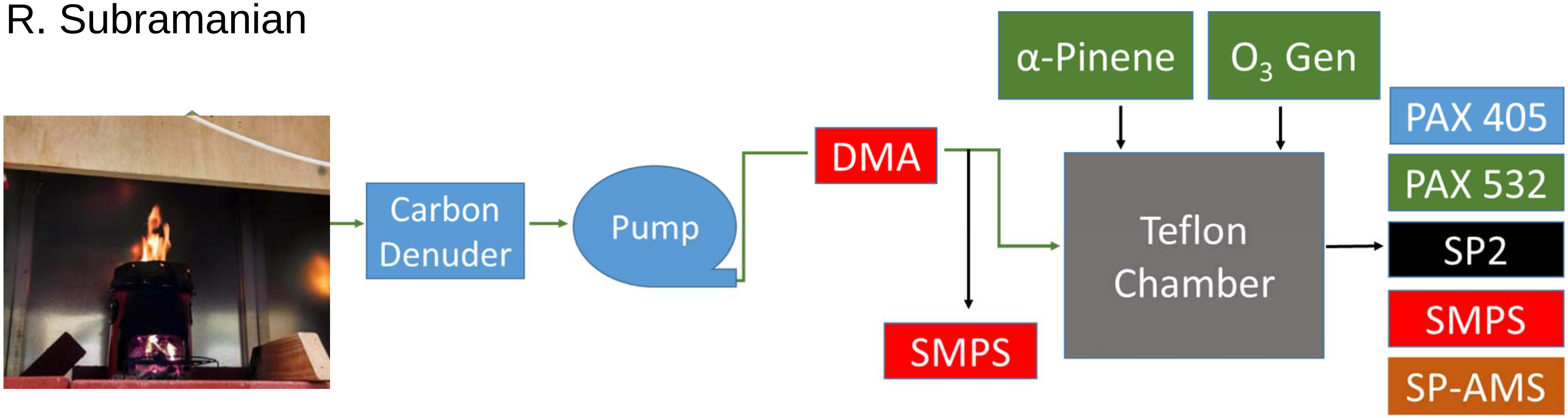
Absorption Angstrom exponent during D-toluene SOA formation

(fuel: White birch bark)



g of Monodisperse Cookstove Soot

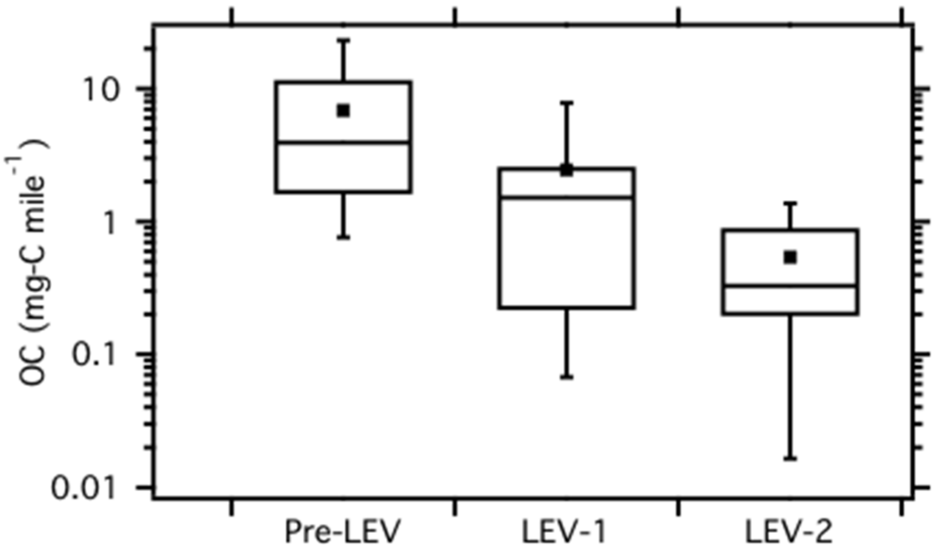
R. Subramanian

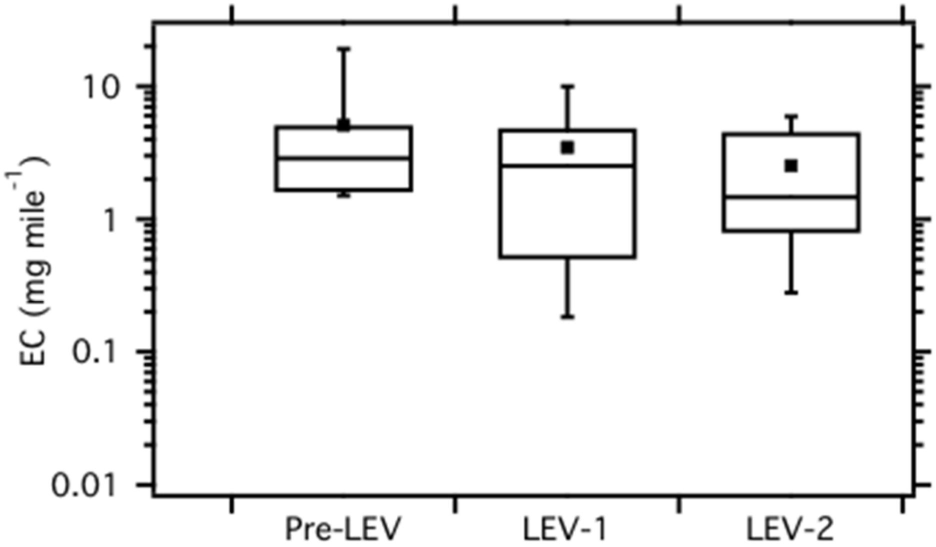


- Absorption enhancement of mono-disperse aged BC particles.
- Three nascent BC core diameters (100, 130, 150 nm mass equivalent diameters).
- Soot was coated with α-pinene SOA in stages till a shell/core diameter ratio of ~2.5
- SP2 for BC mass; SP-AMS for organic aerosol mass; PAXs for light absorption/scattering.

Black Carbon Emissions

Organic/Elemental Carbon Emissions



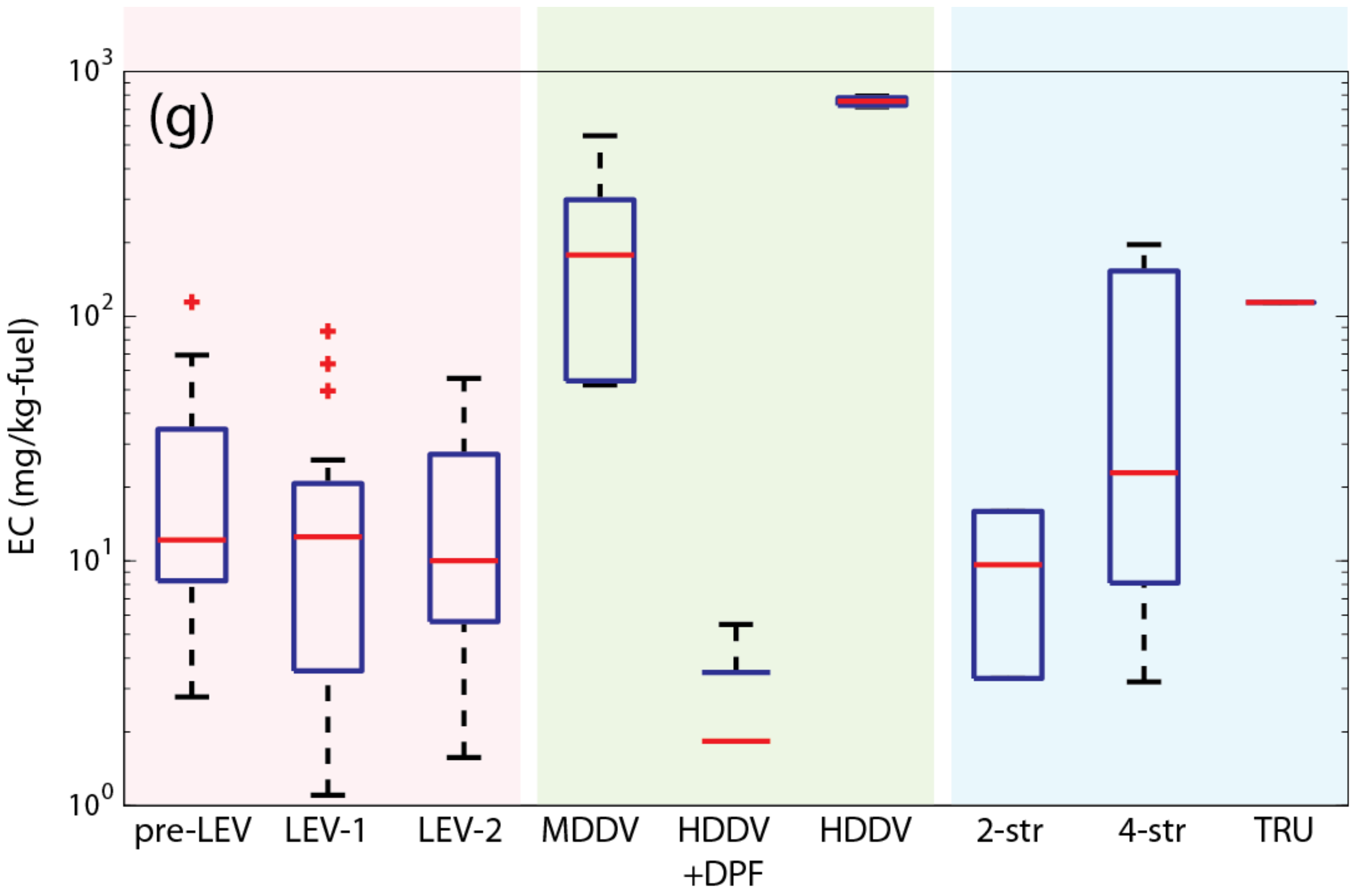


OC emissions decrease with newer vehicles.

EC much less sensitive to more stringent standards.

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

EC Emissions

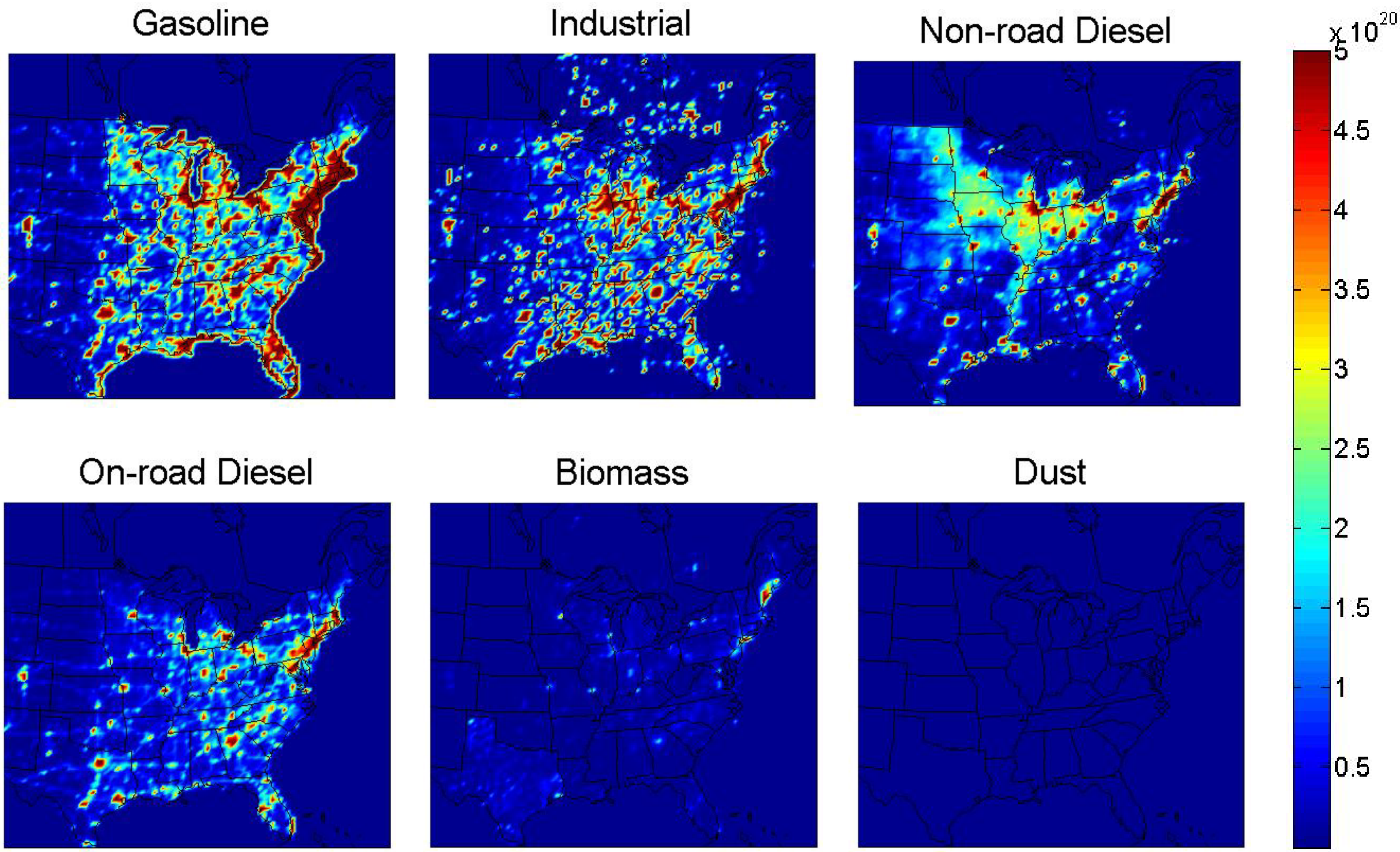


May et al. (Atmos. Environ, 2014)

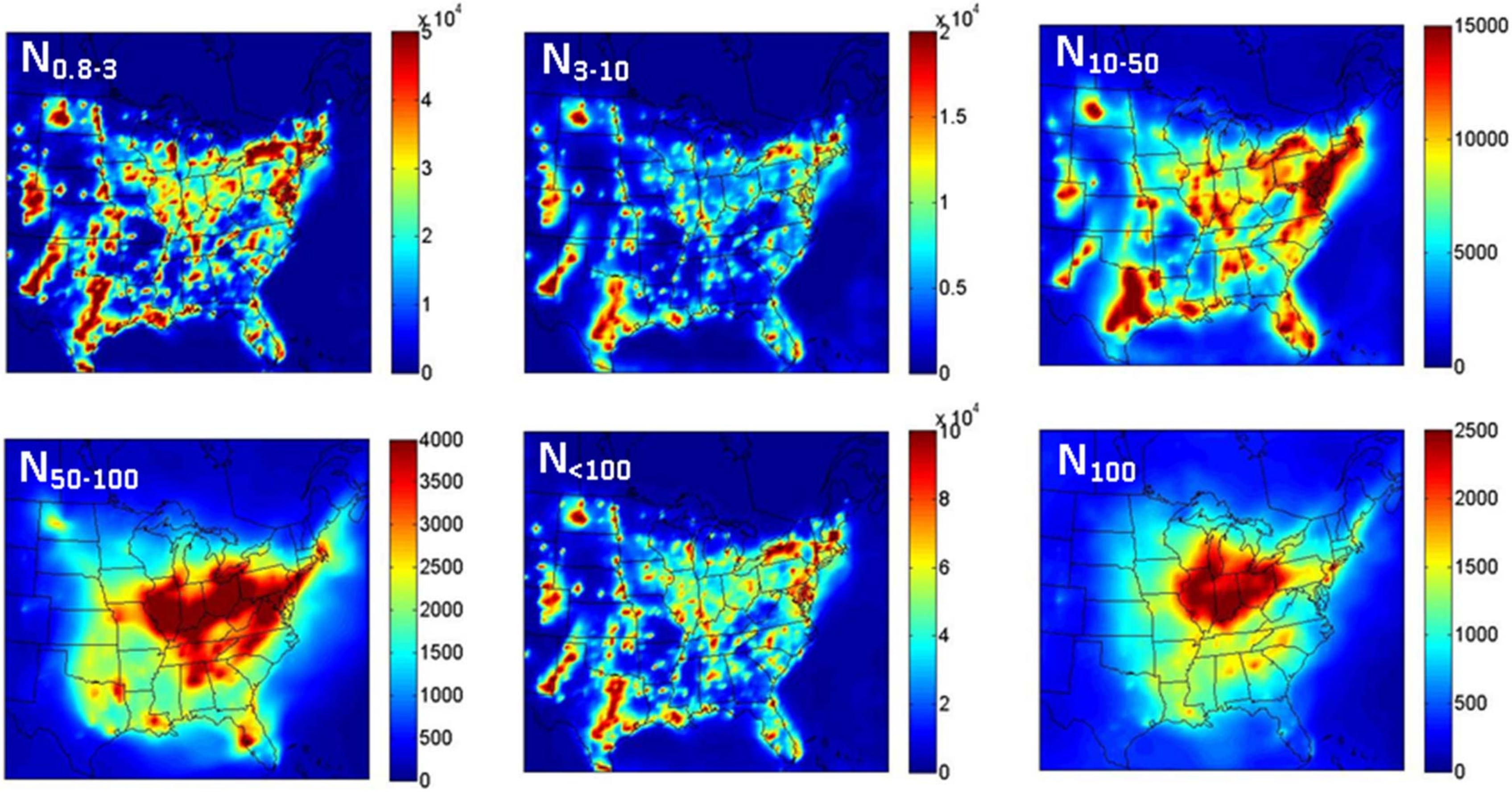
BC and Aerosol Number Concentrations

Source-Resolved Total Number Emissions

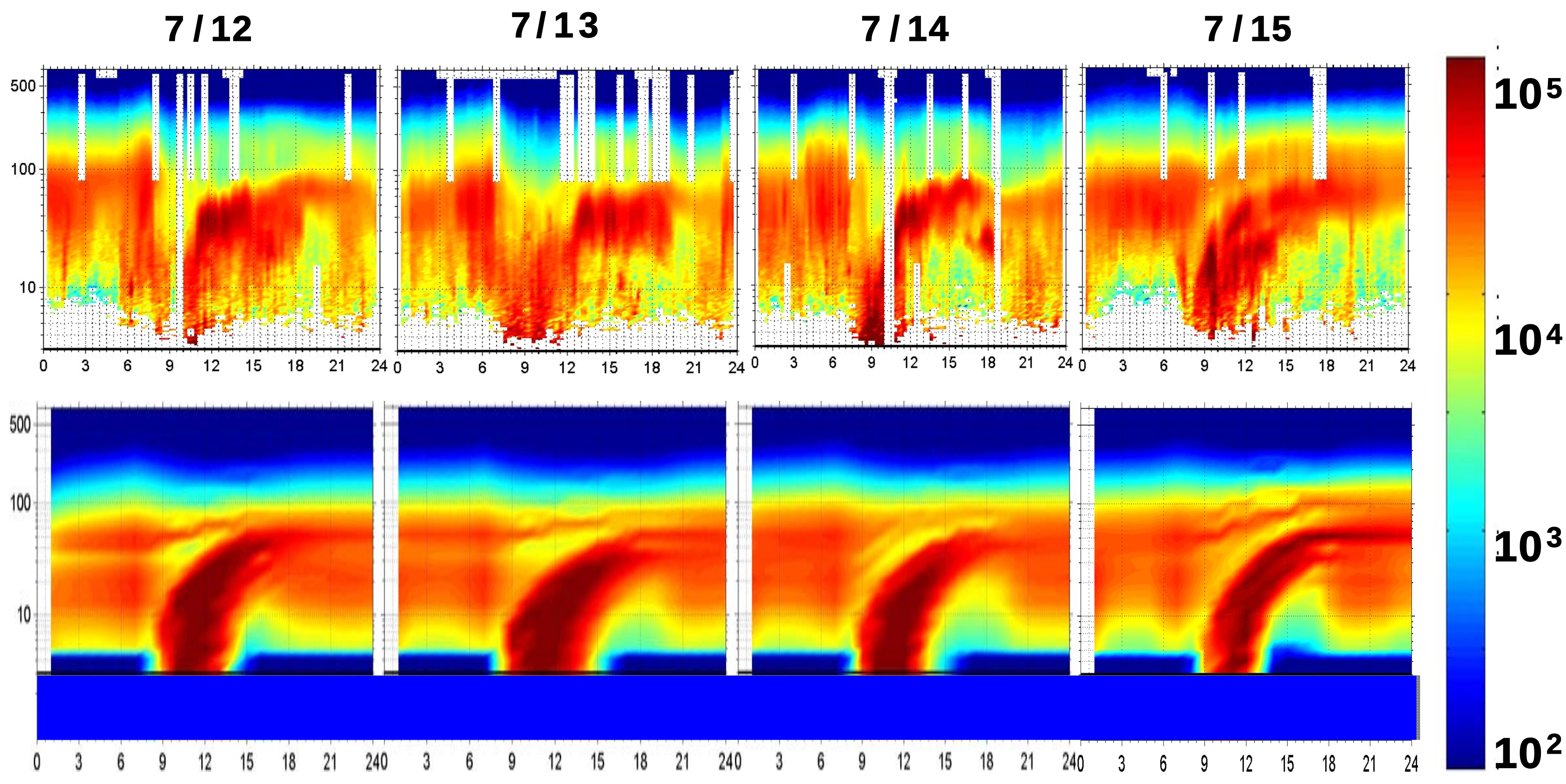
(particles d^{-1} km^{-2})



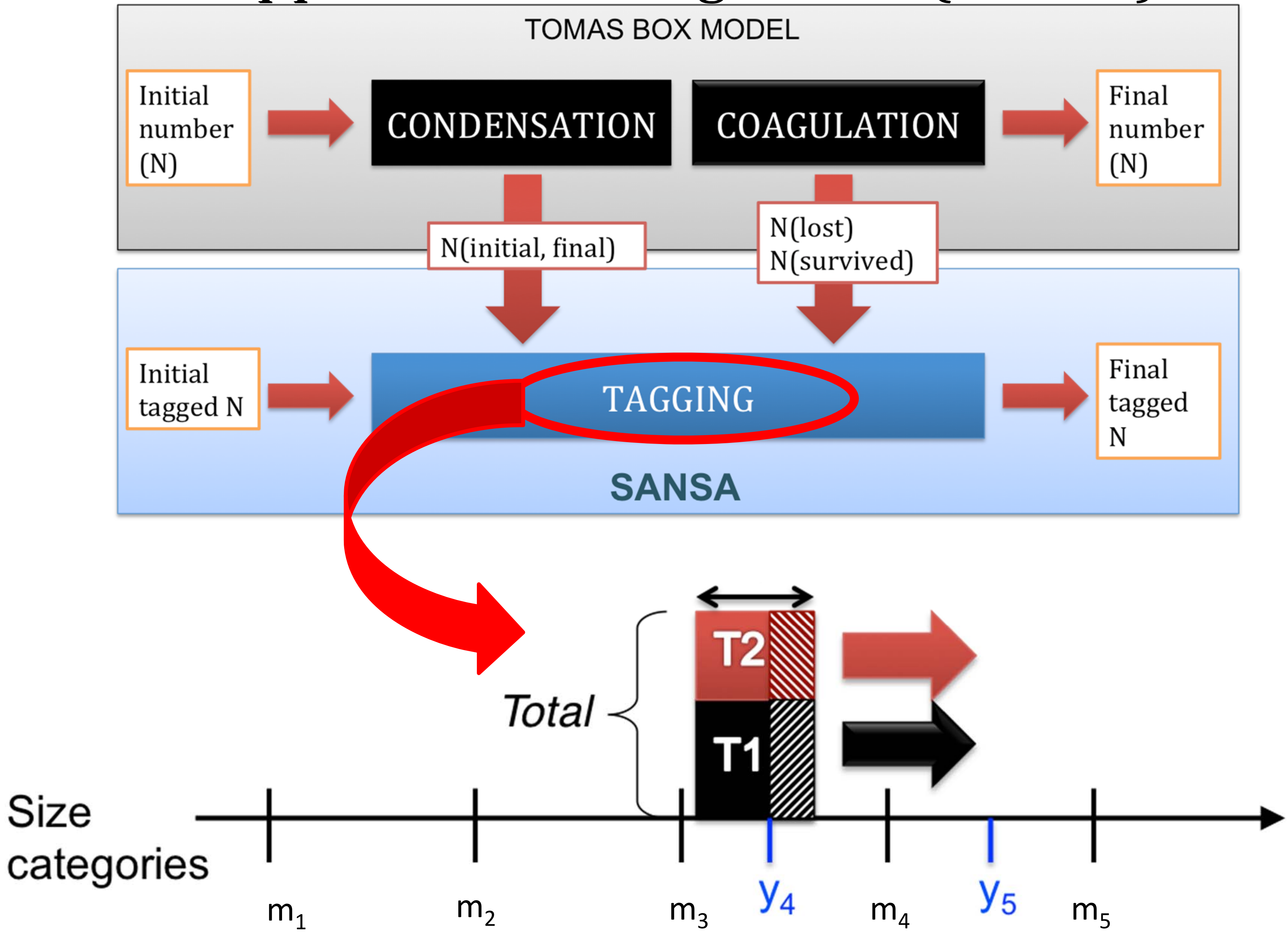
PMCAMx-UF base number concentration (particles cm^{-3})



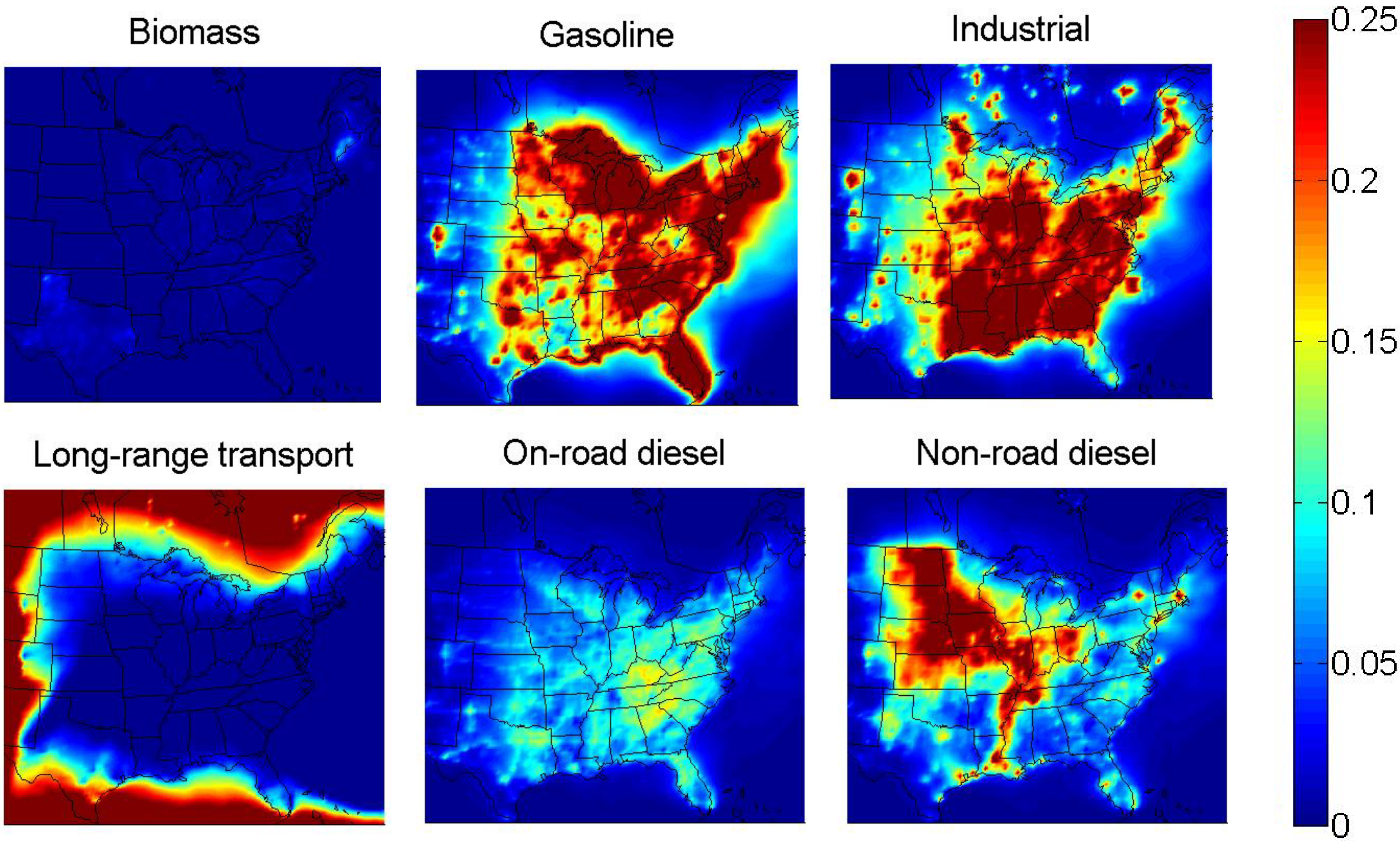
PMCAMx-UF Evaluation (Pittsburgh)



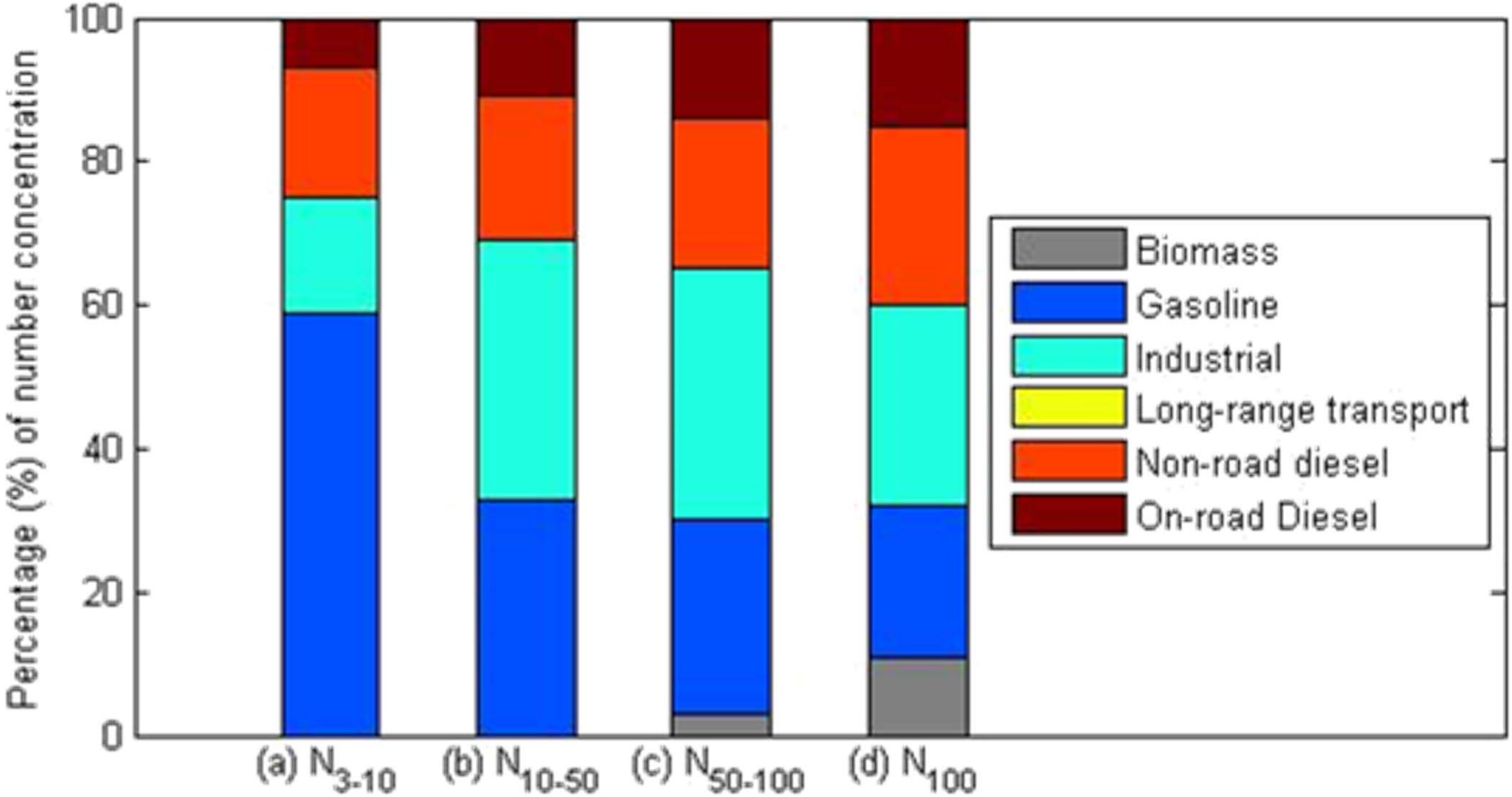
Size-resolved Aerosol Number Source Apportionment algorithm (SANSA)



Total primary particle number fractional source contributions

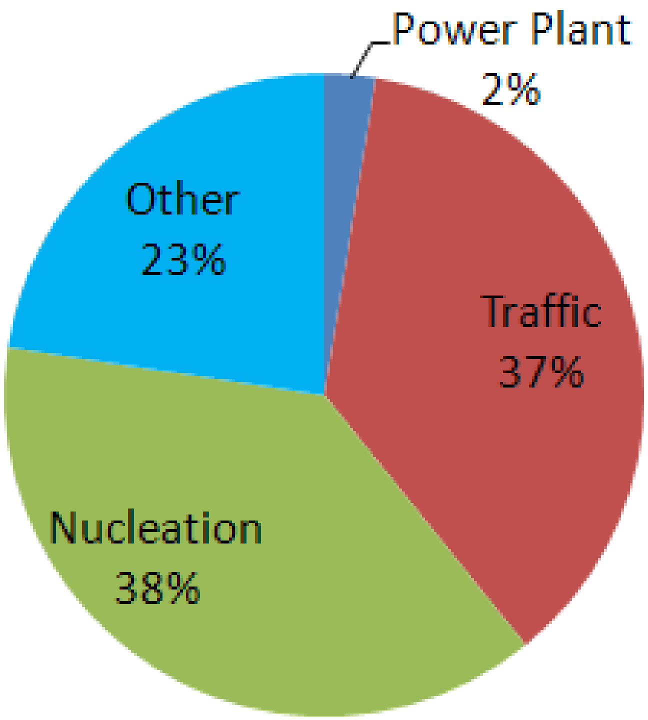


Primary particle number source apportionment in Pittsburgh



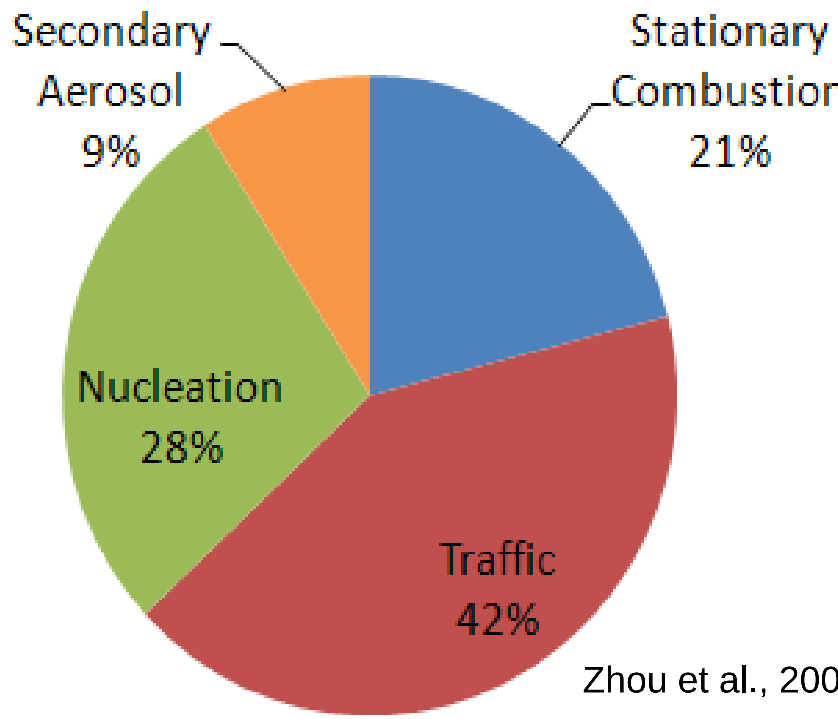
Sources of Measureable (>3 nm) Particle Number in Pittsburgh

PMCAMx



Predicted: 29,000 cm^{-3}

Calculated from Measurements

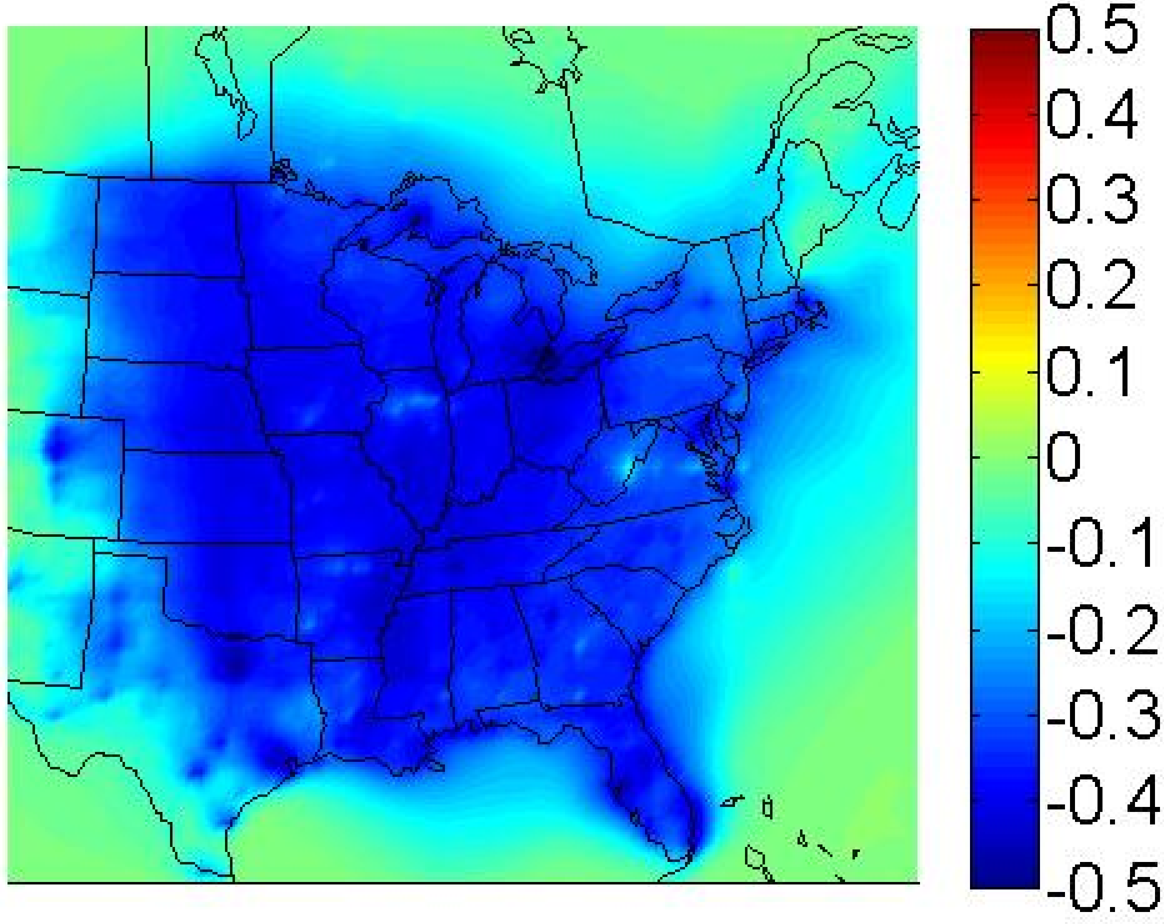


Zhou et al., 2005

“Measured”: 26,000 cm^{-3}

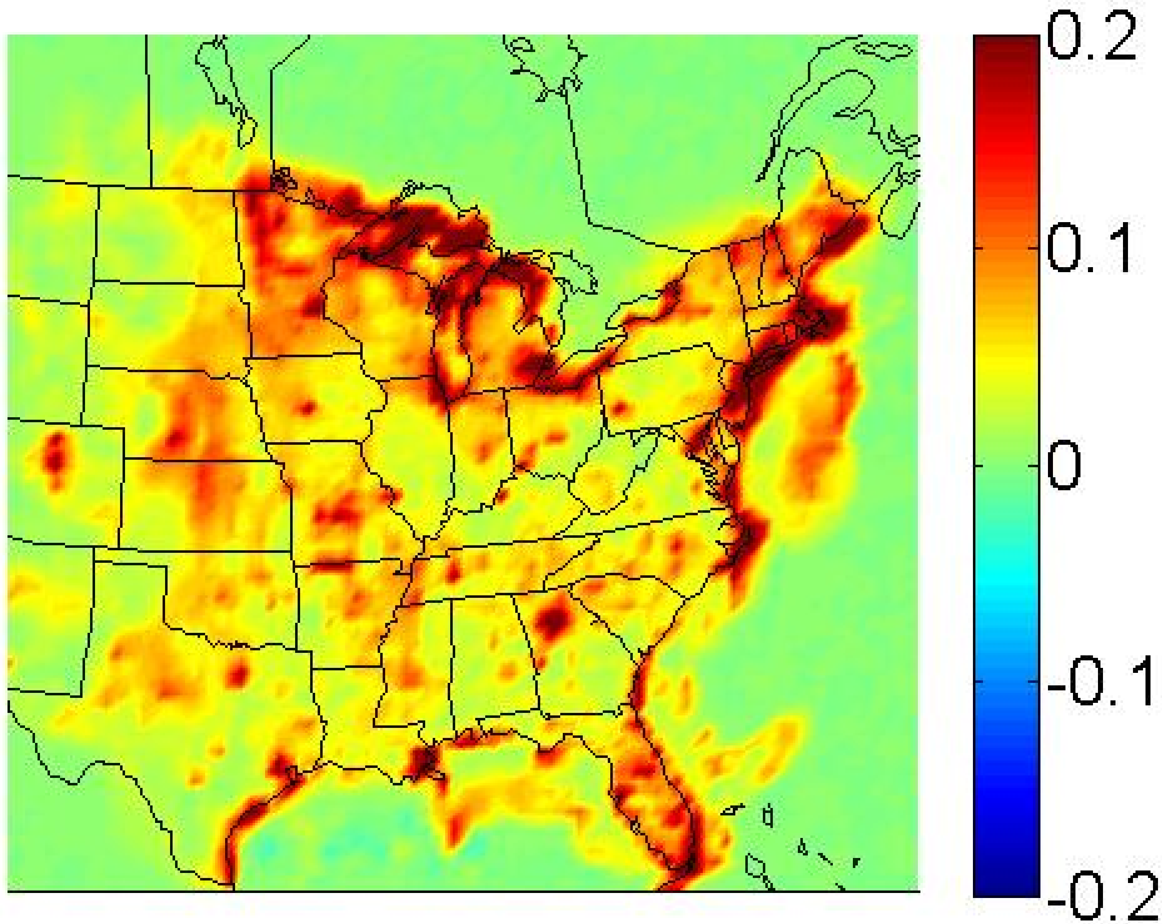
Effects of Controls of Diesel Particulate Emissions (-50% Scenario)

Fractional Change of EC



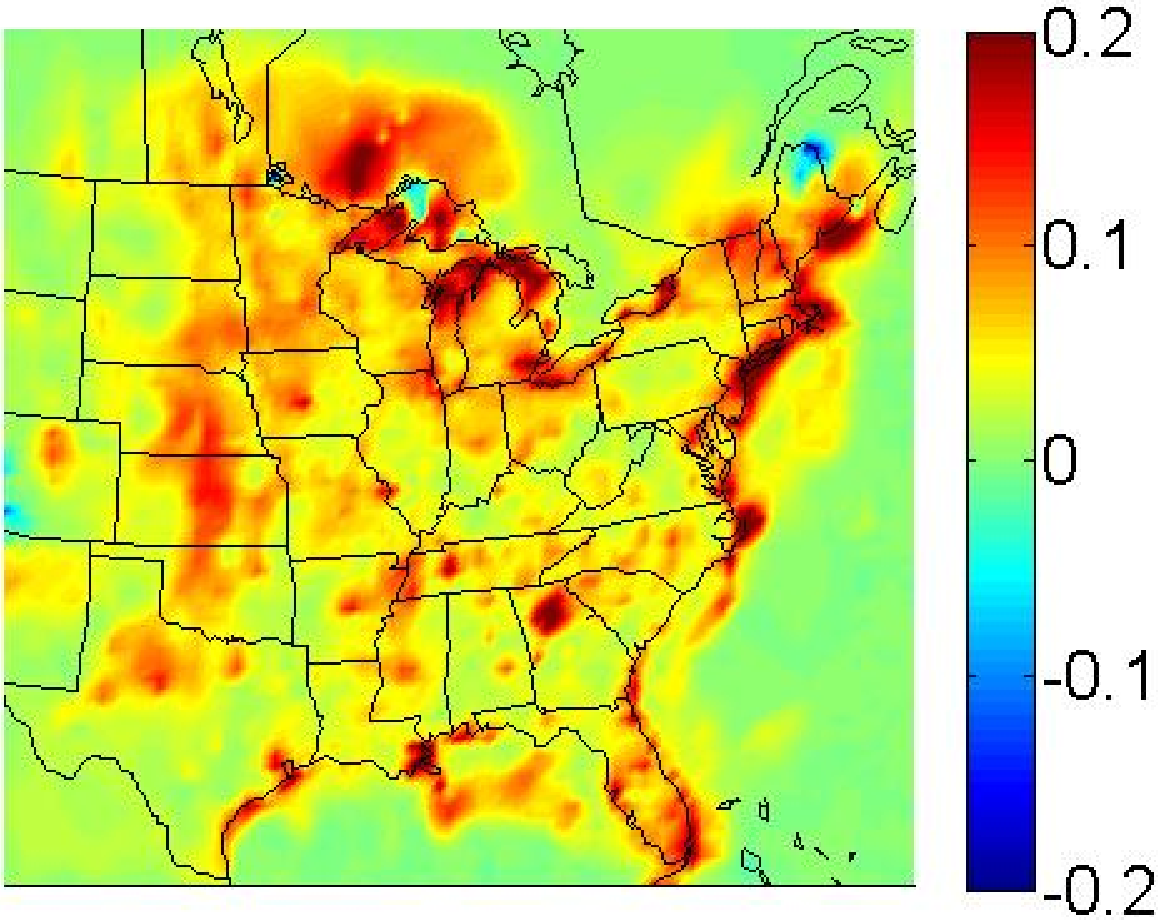
Average $PM_{2.5}$ reduction around 3%.

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



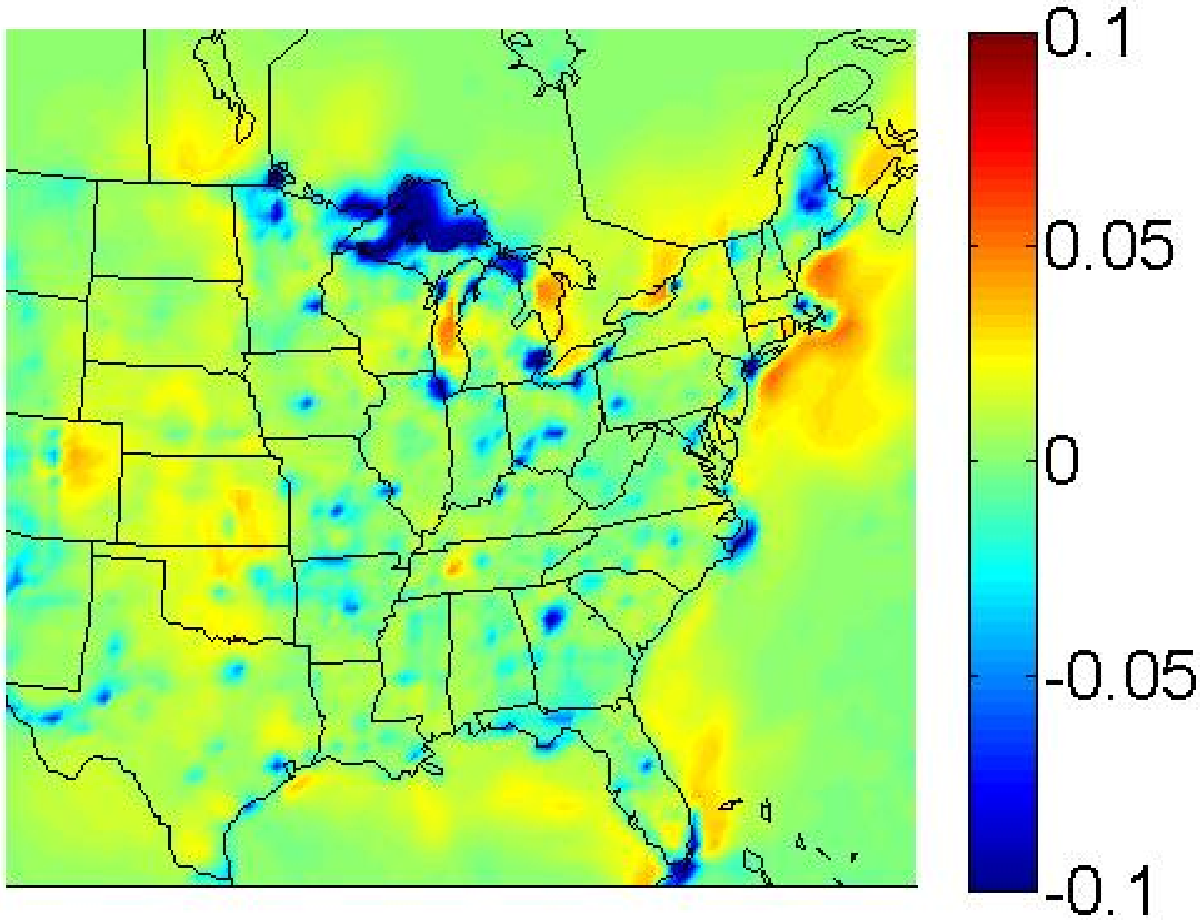
Nucleation increases, creating more smaller particles due to the decrease in the condensation sink.

Fractional Changes of N_{3-10}



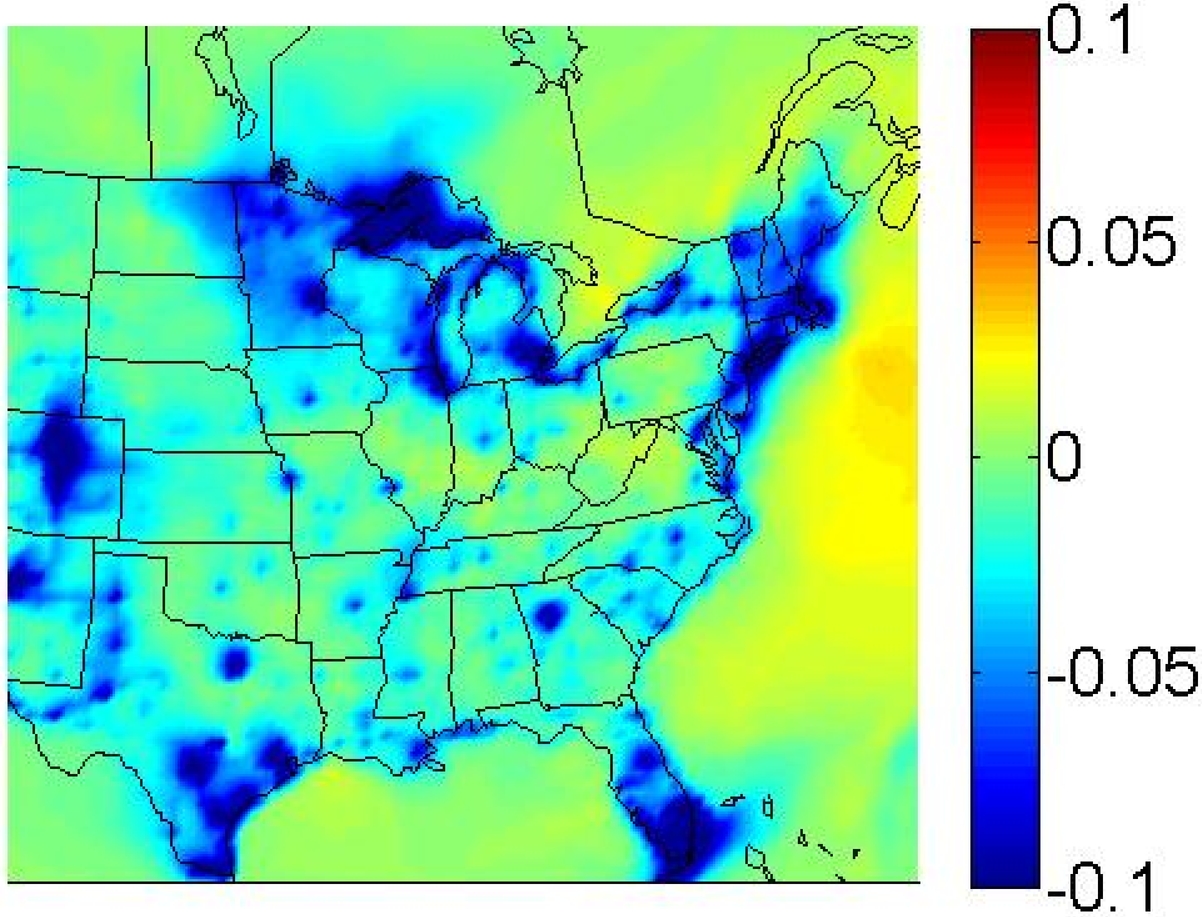
These increases also suggest that nucleation may increase and nucleated particles grow into this size range.

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

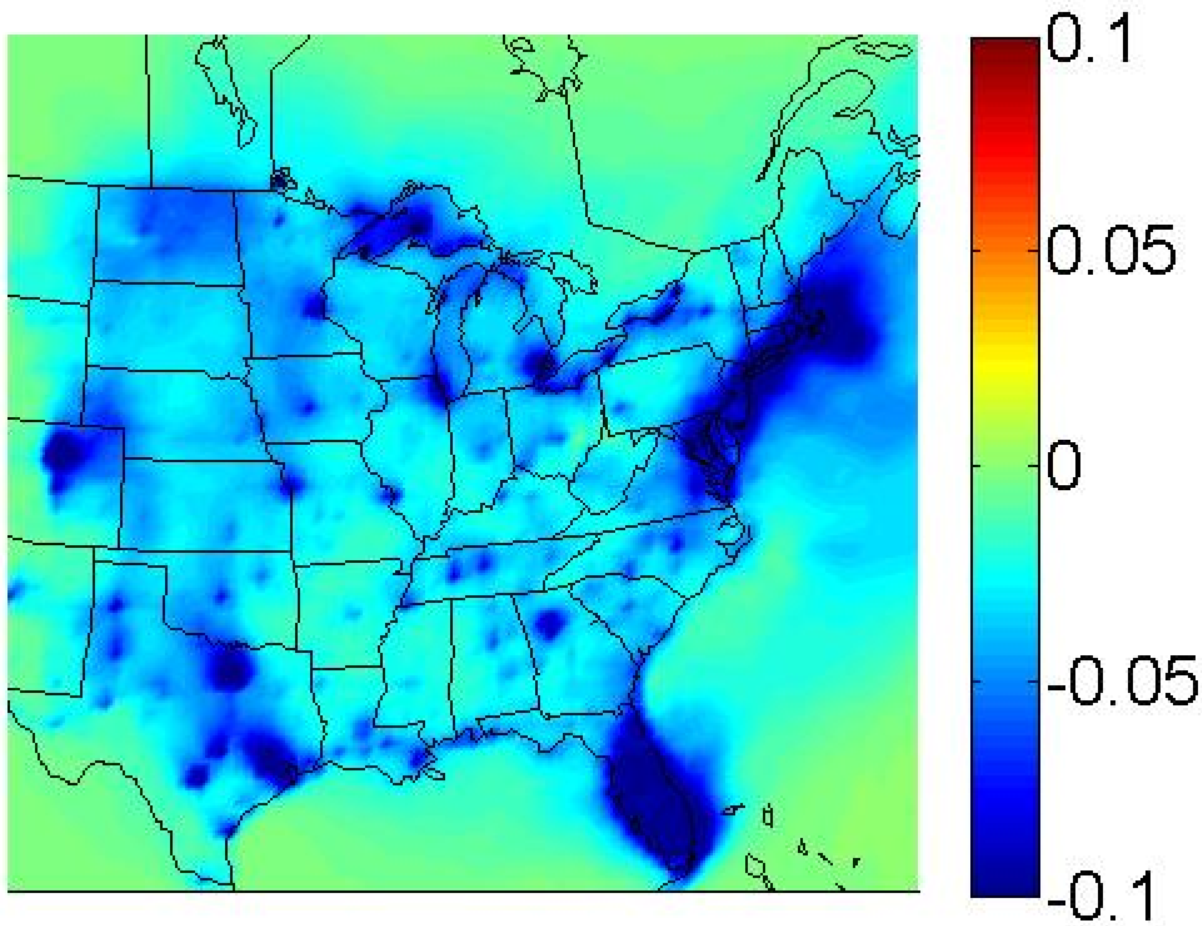


Particles in this size range are typically emitted or grown from nucleated particles, so they see increases (from nucleation) and decreases elsewhere.

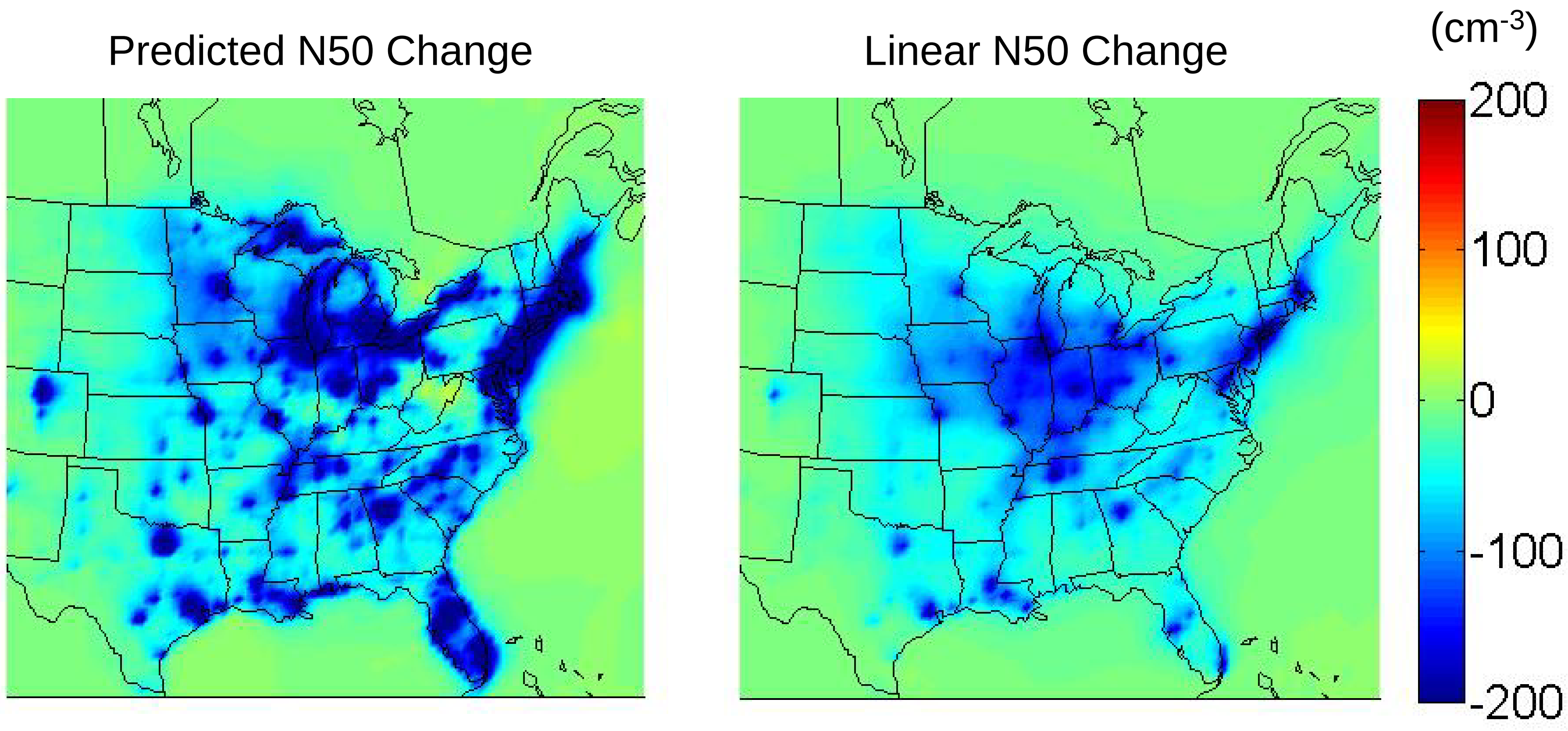
Fractional changes of N_{50-100}



Fractional changes of N_{100}



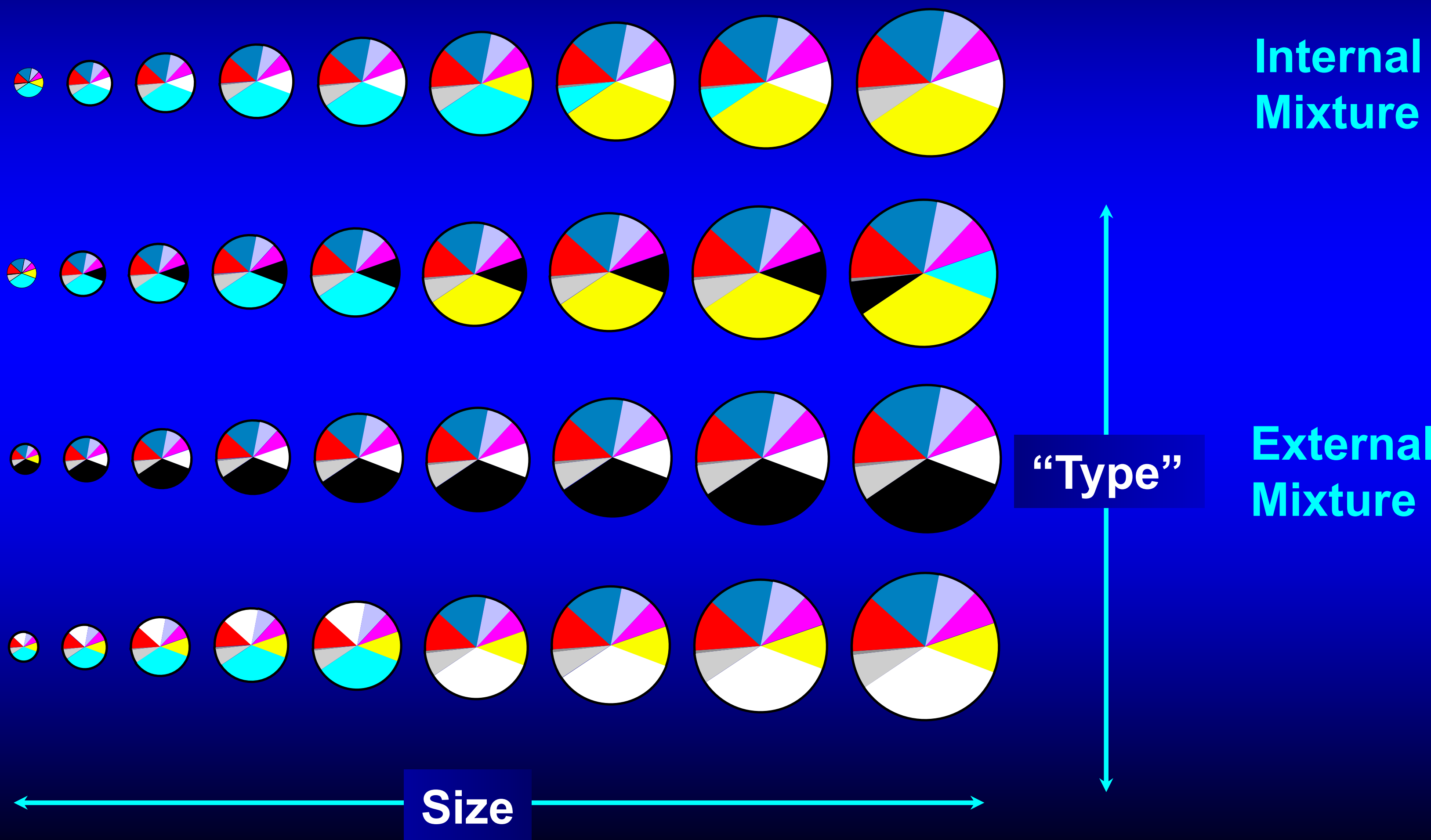
Non-linear Response of CCN to Diesel PM Controls



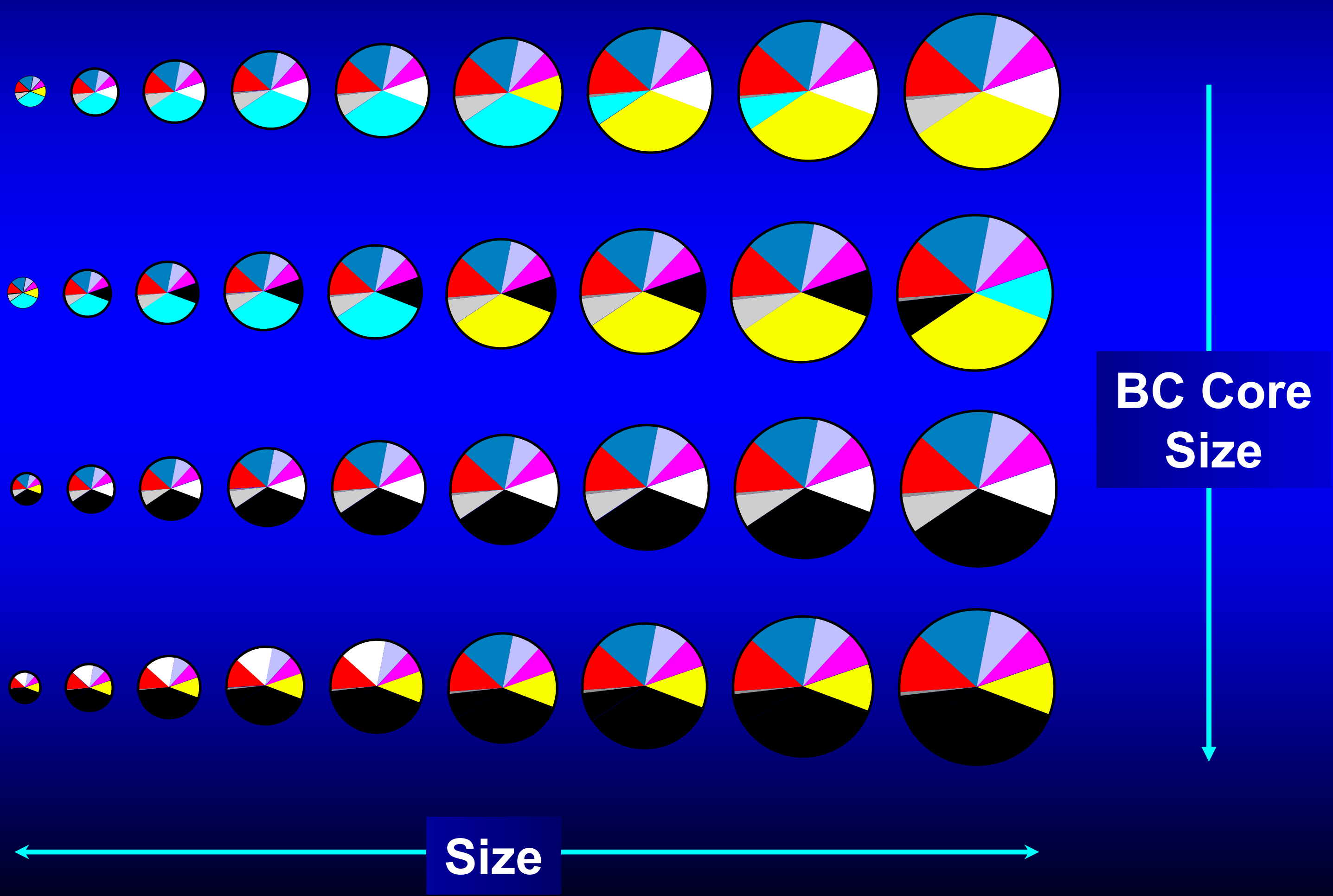
50-100% higher reduction in N50 and N100 than the linear response

Improving Regional Scale BC Models

Relaxing the Internal Mixture Assumption



Simulating BC Mixing State In PMCAMx



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.

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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