Particle-resolved simulations for quantifying black carbon climate impact and model uncertainty

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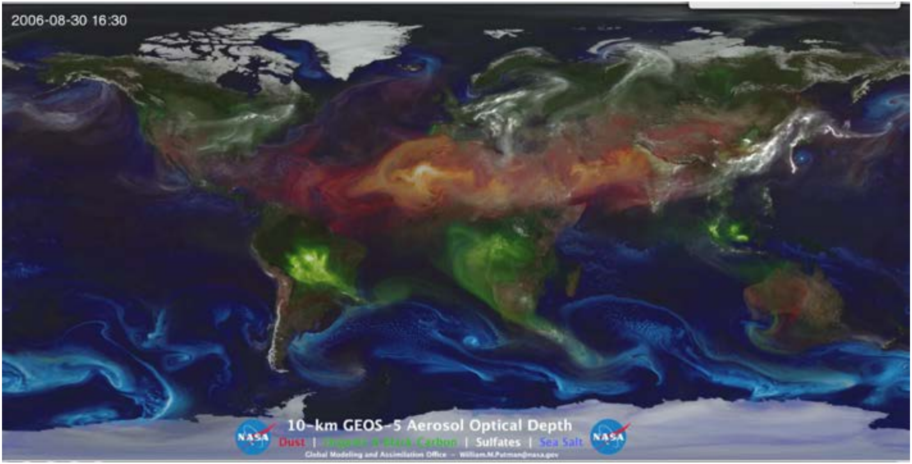
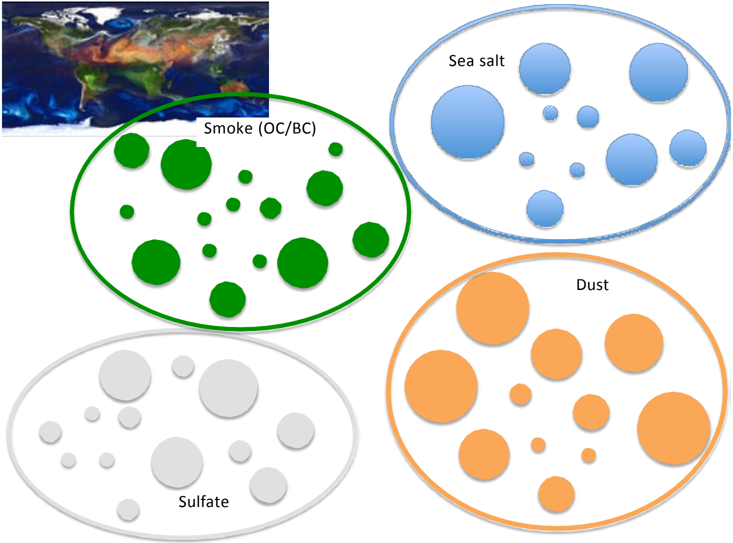
Global aerosol distributions

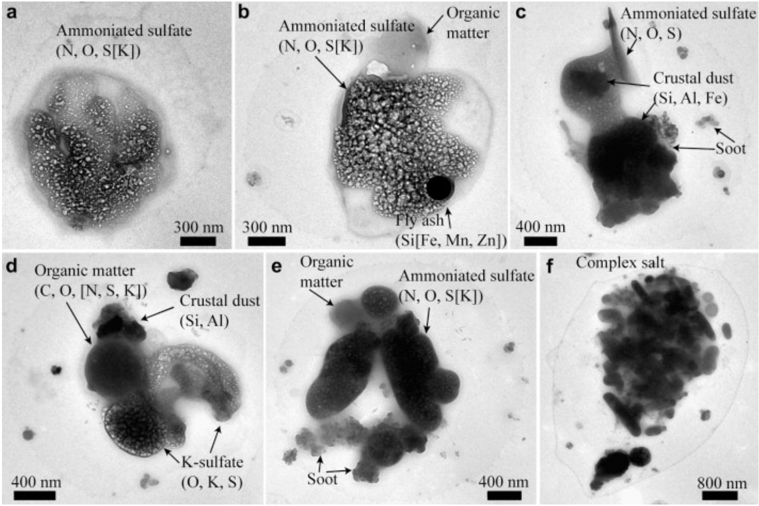


Underlying conceptual model of aerosol particles



External mixture of different aerosol types

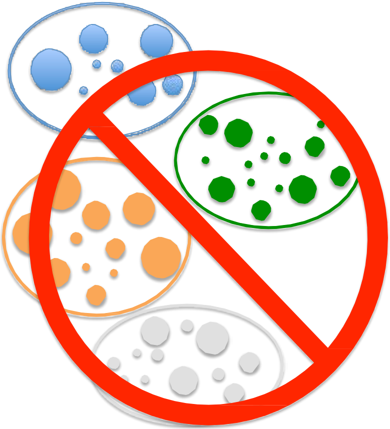
Real particles in the ambient atmosphere



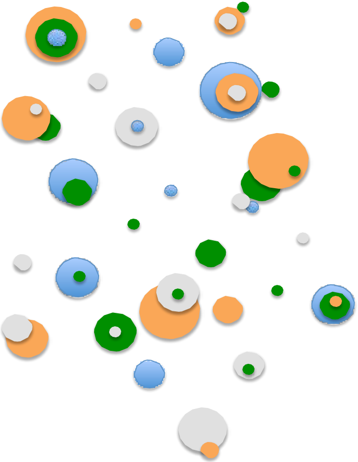
Li et al., Atmospheric Environment, 45, 2488–2495, 2011

Revised underlying model of aerosol particles

External mixture



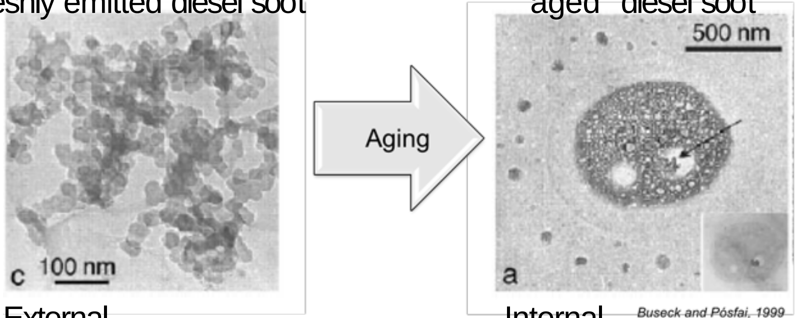
Complex external and internal mixtures



Transformation of black carbon in the atmosphere

Freshly emitted diesel soot

“aged” diesel soot



External

Internal

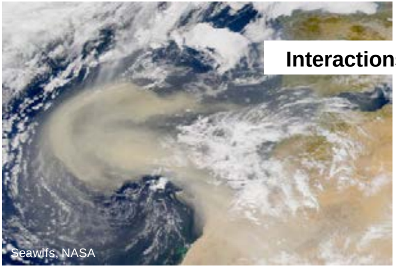
- Freshly emitted soot is hydrophobic.
- Aging due to coagulation and condensation.
- This changes optical properties and hygroscopicity, hence climate impacts.

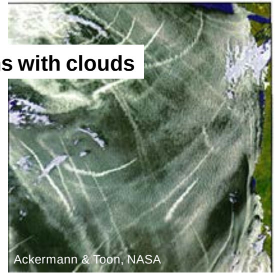
Are these details important?

Health Impacts

Scatter and absorb solar radiation

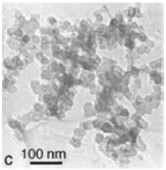
Interactions with clouds

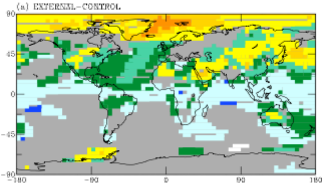




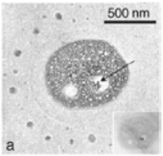
Change in equilibrium annual mean surface air temperature (K)

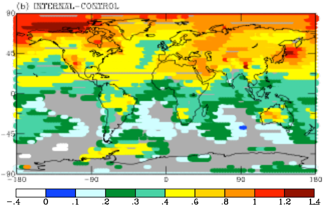
External





Internal





"[...] These results confirm that the mixing state of BC with other aerosols is important in determining its climate effect."

Chung and Seinfeld, JGR 2005

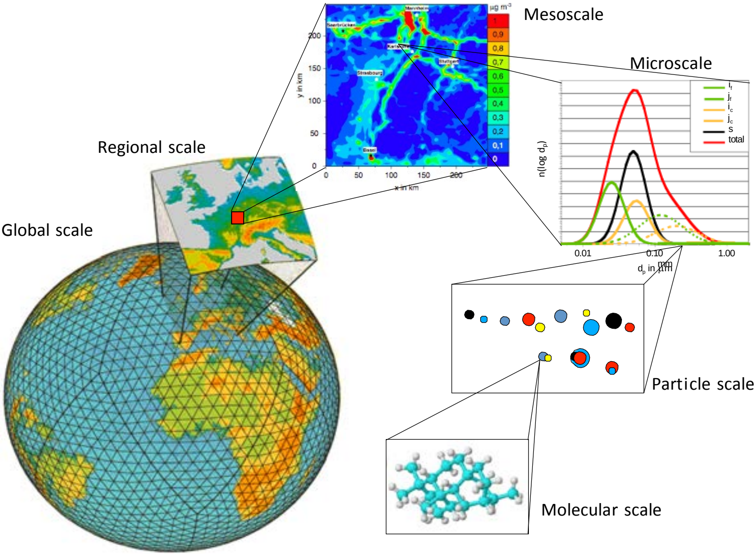
Are these details important?

Table 2. GISS model sensitivity studies.

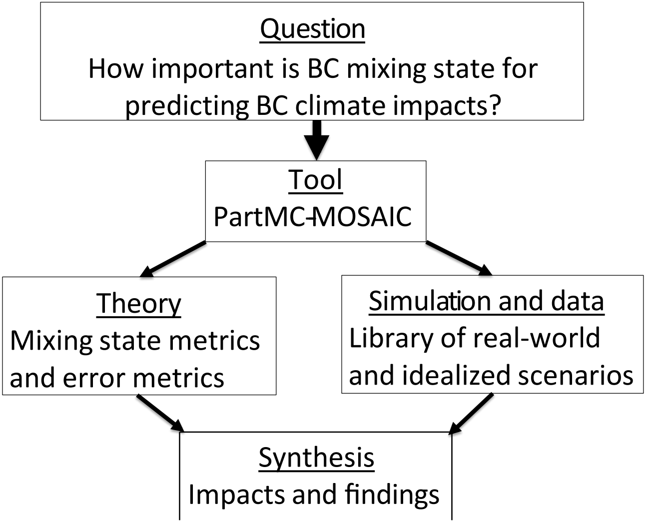
Description	Emission $Tg\,yr^{-1}$	Burden $mg\,m^{-2}$	Lifetime, d	AAOD x100 550 nm
Standard run, see text	7.2 (4.4 energy, 2.8 biomass burning)	0.36	9.2	0.55
EDGAR32 emission	7.5	0.37	9.3	0.58
IIASA emission	8.1	0.41	9.5	0.60
BB 1998	8.2	0.38	8.7	0.58
2x (Faster aging)	7.2	0.29	7.6	0.50
2x (Slower aging)	7.2	0.51	13	0.67
2x More ice-out	7.2	0.33	8.5	0.52
2x Less ice-out	7.2	0.38	9.8	0.57
Reff =0.1 μm	7.2	0.35	9.1	0.47
Reff =0.06 μm	7.2	0.36	9.3	0.70

Source: Koch et al., ACP 2009

Microscale processes determine macroscale impacts



Central research question and strategy



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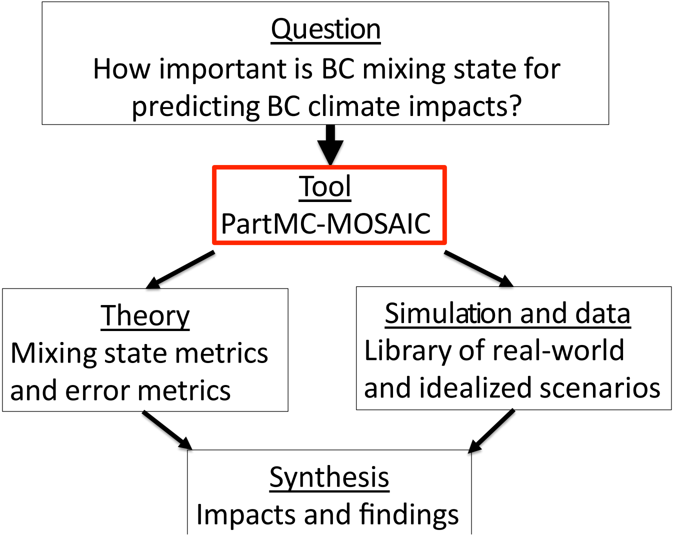
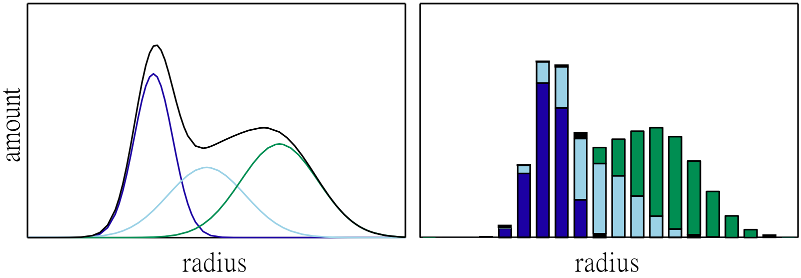


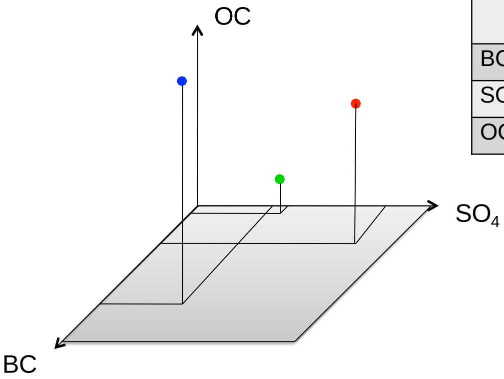
Model aerosol representation: Modes and bins



- Evolution of mixing state is challenging to represent.
- Each mode or size bin is treated as internally mixed.

What are particle-resolved aerosol models?

- No bins or modes
- Particles as vectors
- Treating multidimensional size distribution



	Particle 1	Particle 2	Particle 3
BC	3	10	1
SO_4	12	3	4
OC	5	8	2

Riemer et al., *J. Geophys. Res.*,114, D09202, 2009

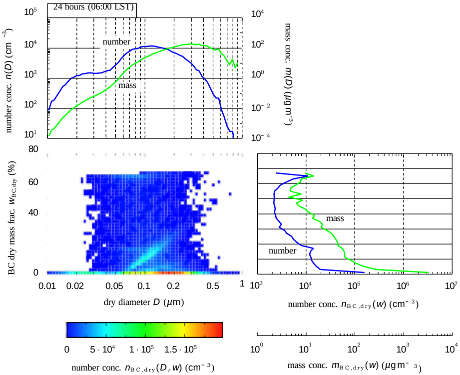
Benefits of particle-resolved models

No approximation needed for mixing state.

1. Coarse graining tool: deriving parameters for more approximate models (e.g. BC aging).
2. Benchmark and error quantification for more approximate models (e.g. QMOM, MADE3, MATRIX, MOSAIC-ext).
3. Detailed studies on the particle scale and experimental intercomparison.

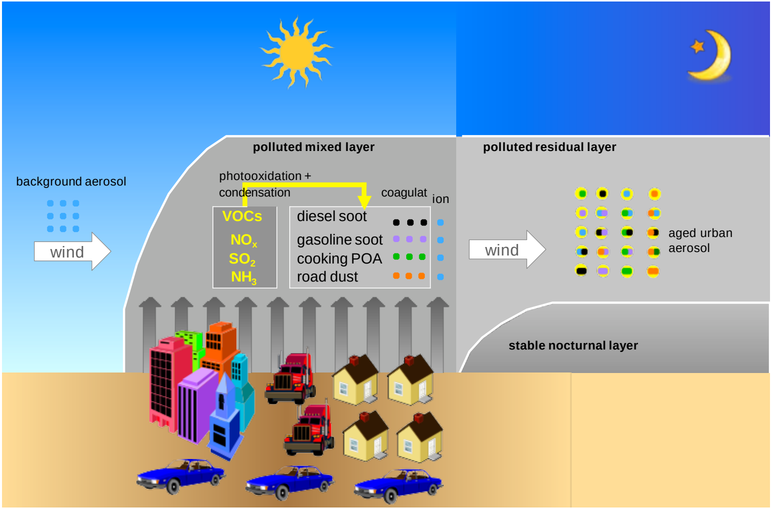
Riemer et al., *J. Aerosol Sci.*, 2010; Willis et al., *Atmos. Chem. Phys.*, 2016; Fierce et al., *Atmos. Chem. Phys.*, 2015, Fierce et al., *Nature Comm.* 2016; Ching et al., *J. Geophys. Res.*, 2012, 2016; Tian et al., *Atmos. Chem. Phys.*, 2014 McGraw et al., *Journal of Physics: Conference Series*, 2008; Kaiser et al., *Geosci. Model Dev.*, 2014

Limitation of particle-resolved models



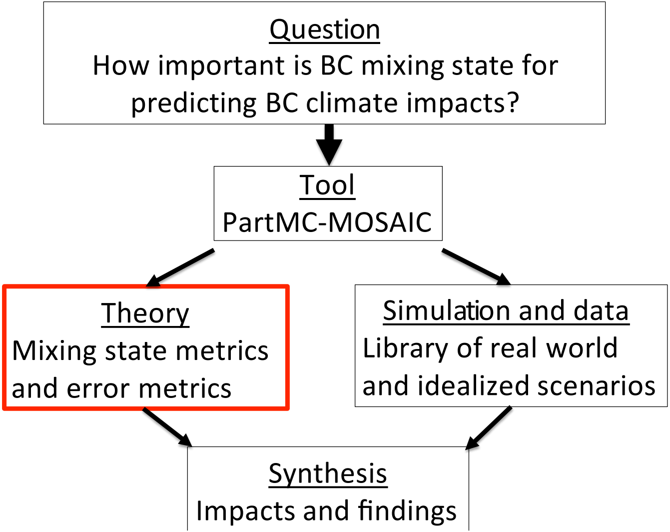
- Resolution: Only finite number of computational particles available per grid cell ($10^4 - 10^7$).
- On-going research to develop more efficient algorithms, e.g. “weighted particles” (DeVille et al., *J. Comp. Phys.*, 2011) and parallel methods.

Typical model setup

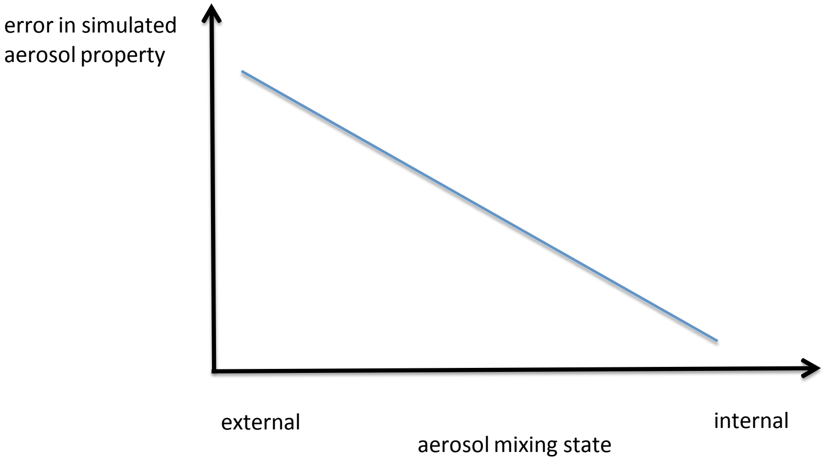


Zaveri, Easter, Riemer, West, JGR 2010

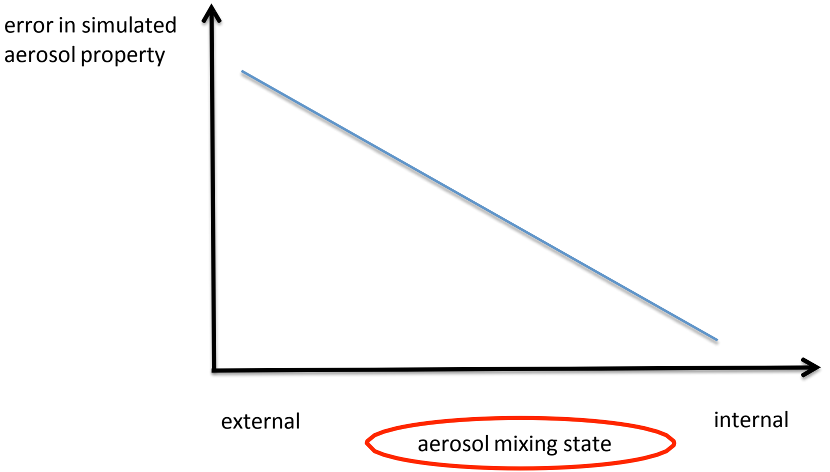
Central research question and strategy



Hypothesis: Error versus mixing state metric

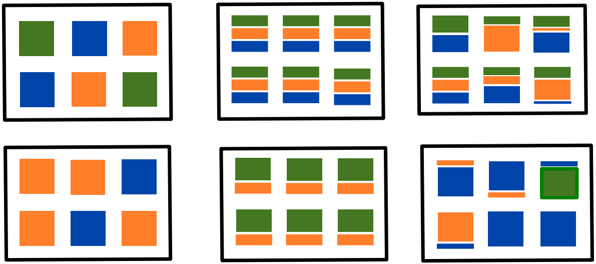


Hypothesis: Error versus mixing state metric



Mixing state terminology

- **Population mixing state** and **Morphological mixing state**
- Here we will only consider the population mixing state.



1. How "complex" are the particles, i.e. how many species are present in one particle?
2. How different are the particles from each other?

Problem solved in ecology: Species diversity

Externally mixed waterholes

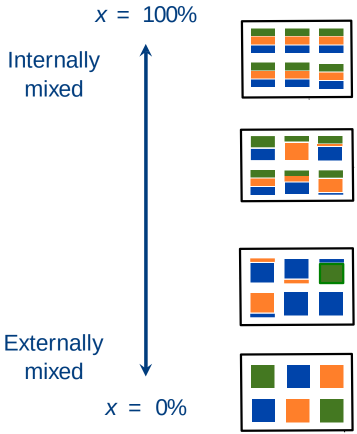


Internally mixed waterholes



Good, *Biometrika*, 1953
MacArthur, *Ecology*, 1955
Whi> aker, *Ecol. Monogr.*, 1960; *Science* 1965; *Taxon*, 1972

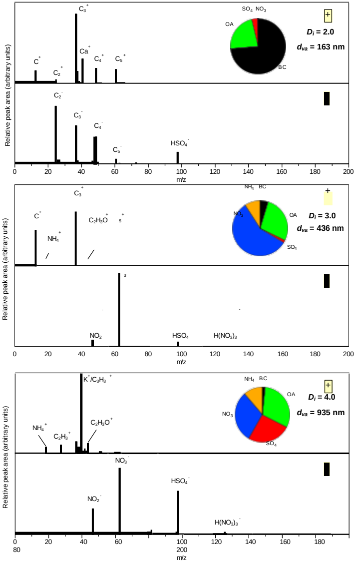
Mixing state metric χ



- Based on diversity metrics framework used in ecology.
- Need to know per-particle mass fractions.

Riemer and West, *Atmos. Chem. Phys.*, 2013; Healy et al., *Atmos. Chem. Phys.*, 2014; O'Brien et al., *J. Geophys. Res.*, 2015

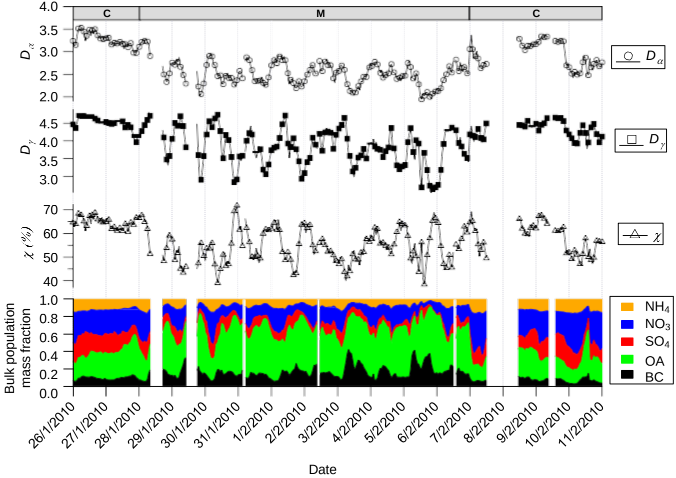
Application to field data: MEGAPOLI



- Collaboration with Robert Healy, University of Toronto.
- Paris, France, winter 2010.
- Mass fractions estimated based on ATOFMS data, supplemented by AMS, SMPS and MAAP.
- Five species: OA, BC, SO_4, NO_3, NH_4.

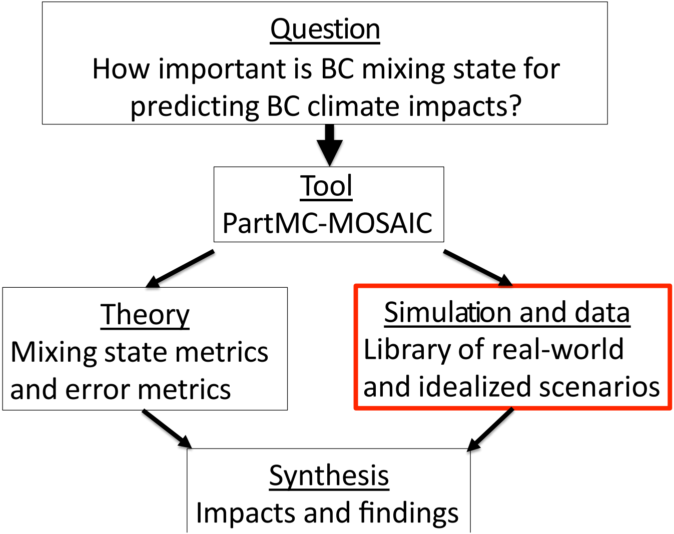
Healy et al., *Atmos. Chem. Phys.*, 4, 6289–6299, 2014

Mixing state parameters during the campaign

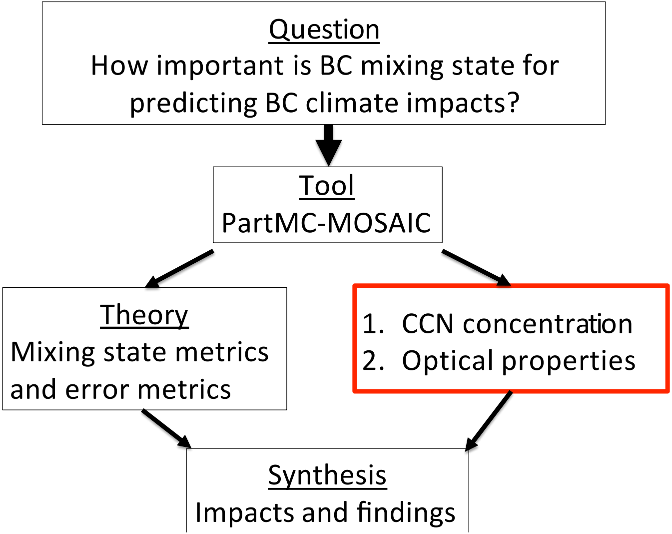


Healy et al., *Atmos. Chem. Phys.*, 4, 6289–6299, 2014

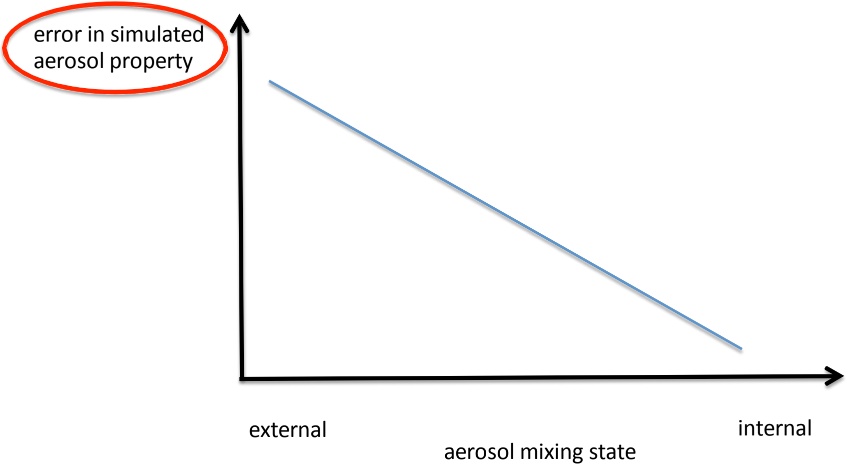
Central research question and strategy



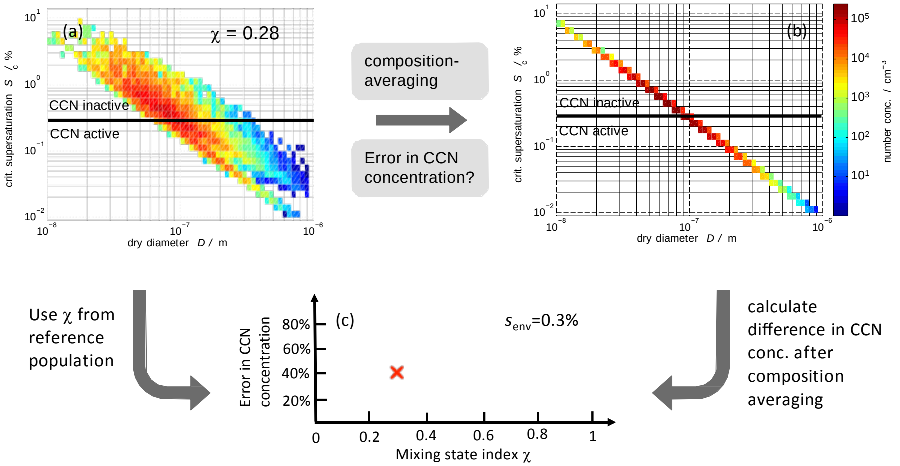
Central research question and strategy



Back to the hypothesis

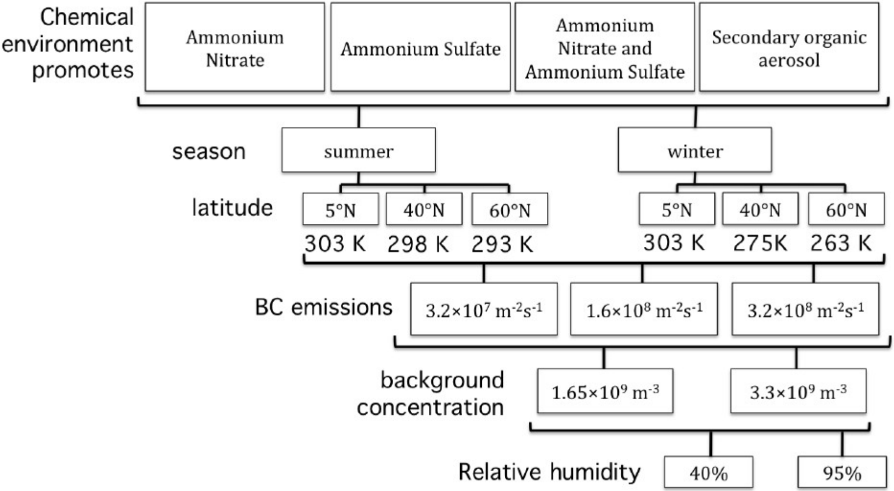


Importance of mixing state for CCN properties



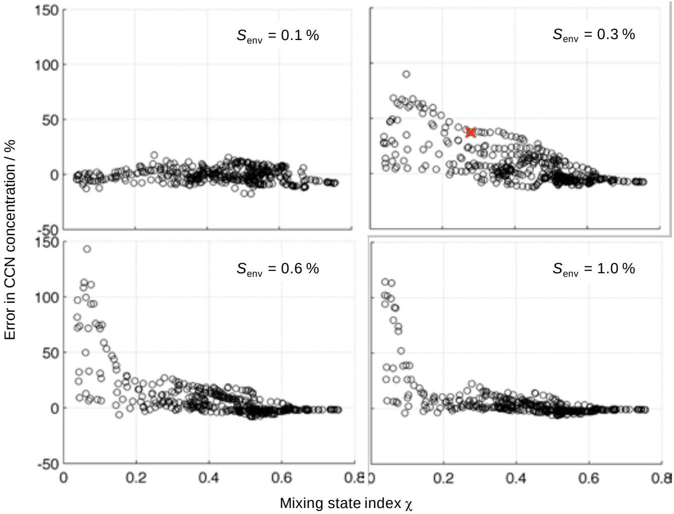
Ching et al., ACP, in prep., 2016

Scenario library to sample relevant parameter range



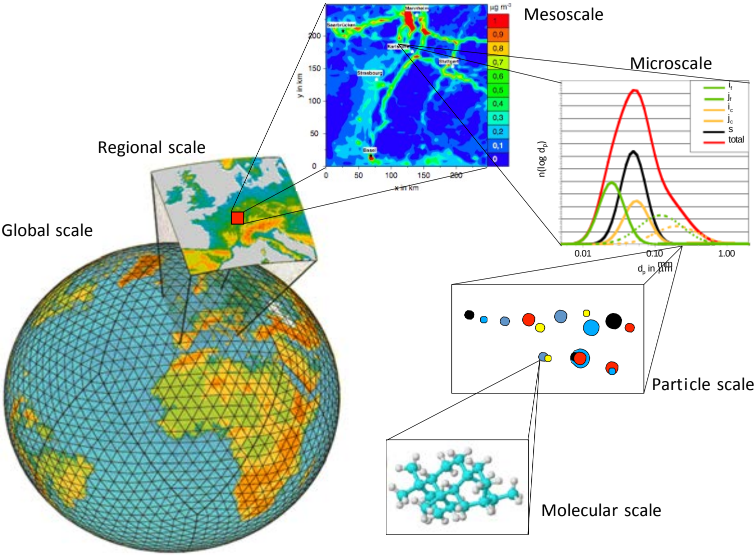
Fierce et al., ACP, 2015; Fierce et al., Nature Comm., 2016; Ching et al., JGR, 2016

Relationship of χ and error in CCN concentration

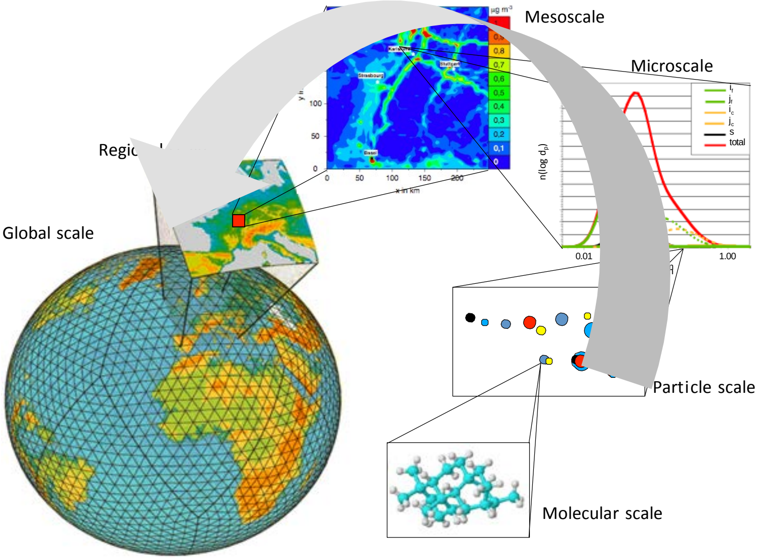


Ching et al., ACP, in prep., 2016

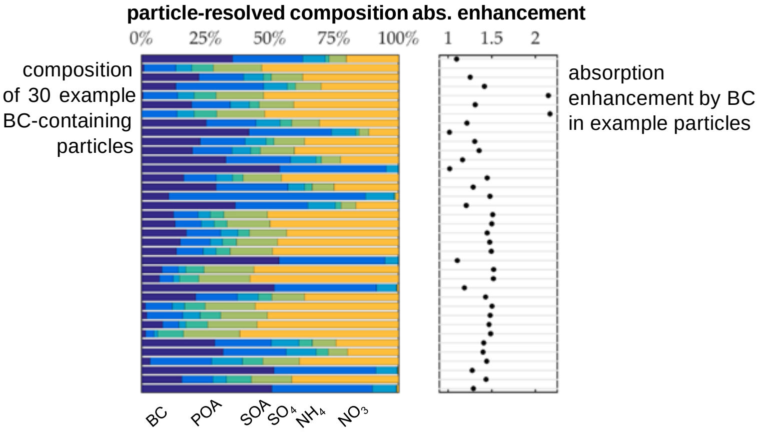
Back to the multiscale model hierarchy



Back to the multiscale model hierarchy



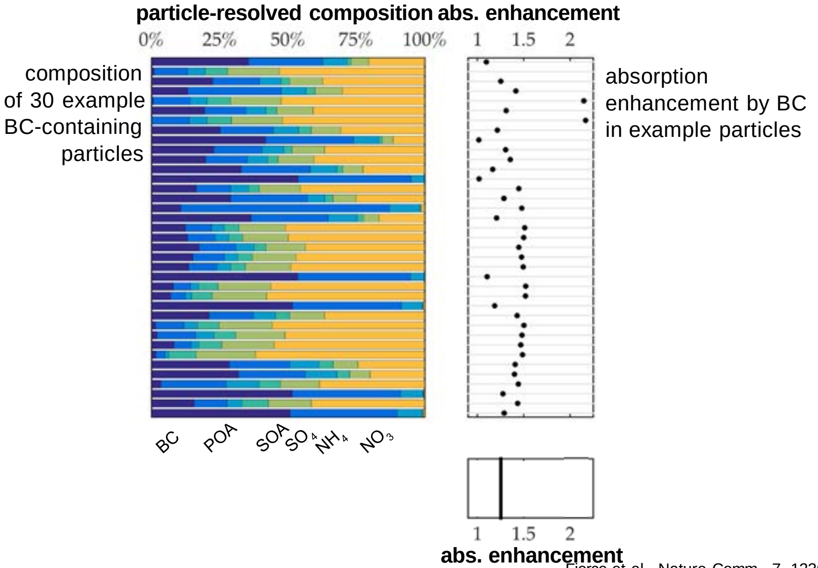
Diversity in composition affects BC absorption



Per-particle composition → *Per-particle absorption*

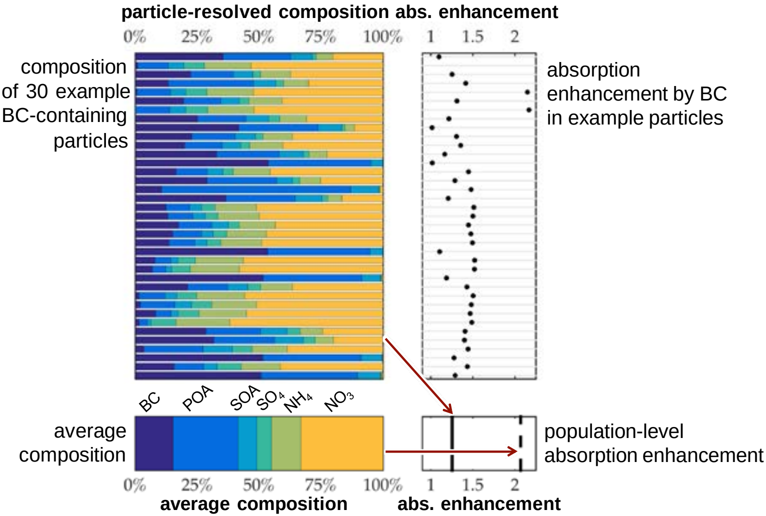
Fierce et al., Nature Comm., 7, 12361, 2016.

Diversity in composition affects BC absorption



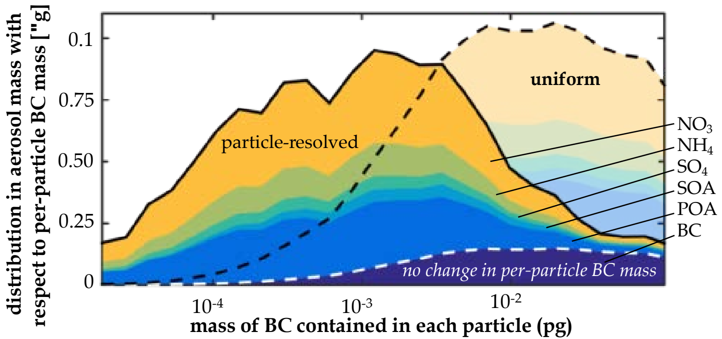
Fierce et al., Nature Comm., 7, 12361, 2016.

Diversity in composition affects BC absorption



Fierce et al., Nature Comm., 7, 12361, 2016.

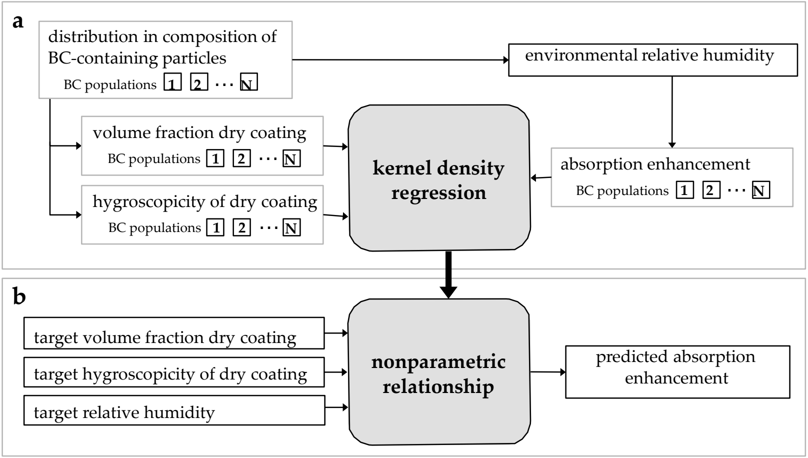
Why this bias?



- Uniform composition assumption: All particles contain the same volume fraction of each aerosol component.
- Causes an artificial redistribution of coating material onto particles containing large amounts of BC.

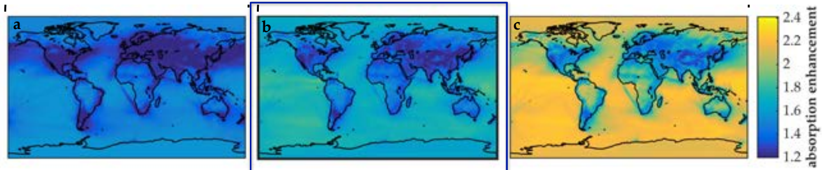
Fierce et al., Nature Comm., 7, 12361, 2016.

Coarse-graining of particle-resolved data



Fierce et al., Nature Comm., 7, 12361, 2016, Supplemental Material.

Taking this to the global scale



Accounting for particle-level composition diversity	Accounting for particle-level composition diversity	Assuming uniform composition
Neglecting water uptake	Accounting for water uptake	Accounting for water uptake

our best estimate

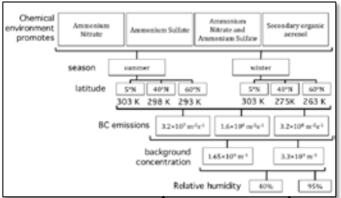
Fierce et al., Nature Comm., 7, 12361, 2016.

Outcomes

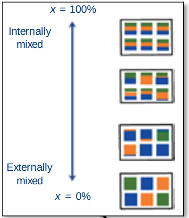
1. Framework to quantify the impact of aerosol mixing state on different target quantities (CCN properties and optical properties).
   - Mixing state metric and error metrics
   - Coarse-graining method: Use particle-resolved simulations to develop parameterization with inputs that large-scale models already track.
2. CCN properties: Small errors (< 10 %) are caused by internal-mixture assumption for populations with $\chi > 0.6$. Large errors (up to 150 %) for $\chi < 0.2$.
3. Optical properties: If particle-diversity is neglected, population-level absorption enhancement is overpredicted (up to 70%).

Impacts on community

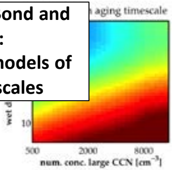
Scenario libraries



Mixing state metrics



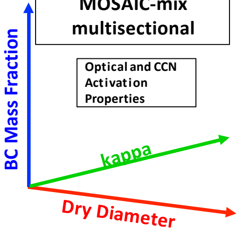
With Tami Bond and Laura Fierce: Predictive models of aging time scales



Fierce et al., ACP, 2015
Fierce et al., BAMS, 2015

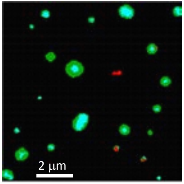
With PNNL: MOSAIC-mix multisectional

Optical and CCN Activation Properties



Ching et al., JGR, 2015

With O'Brien, Moffet, Gilles, Laskin: Spectro-microscopy and mixing state



O'Brien et al., JGR, 2015