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

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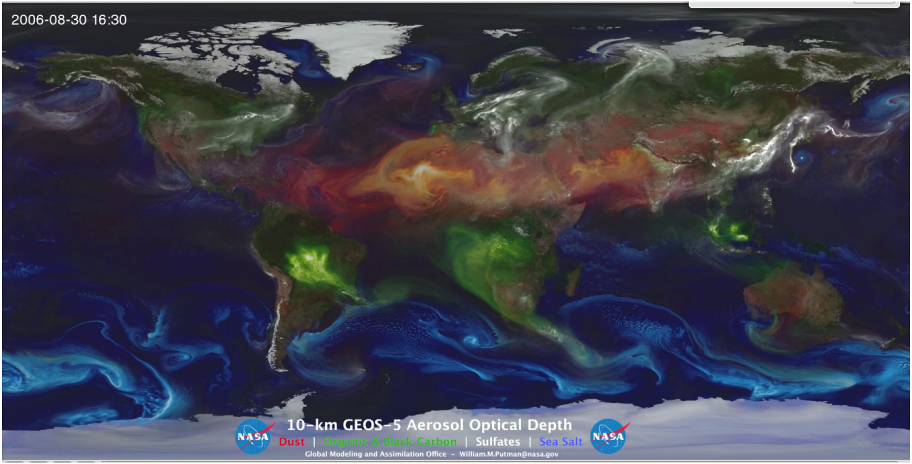
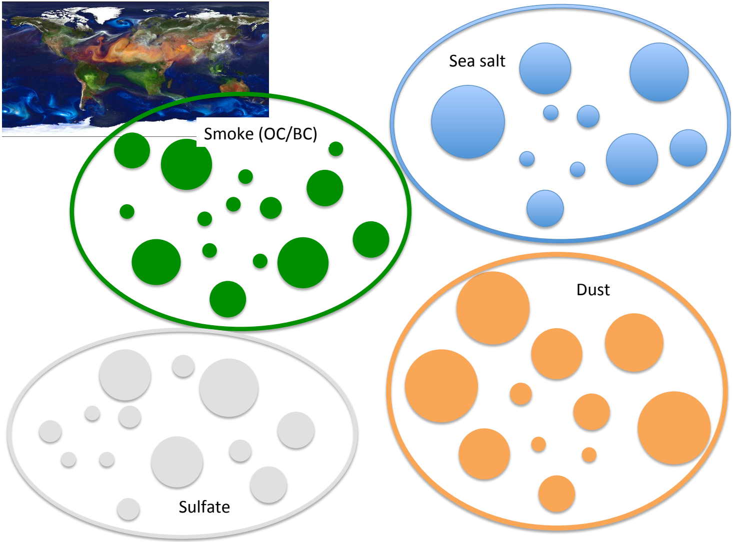
Simulated aerosol distributions

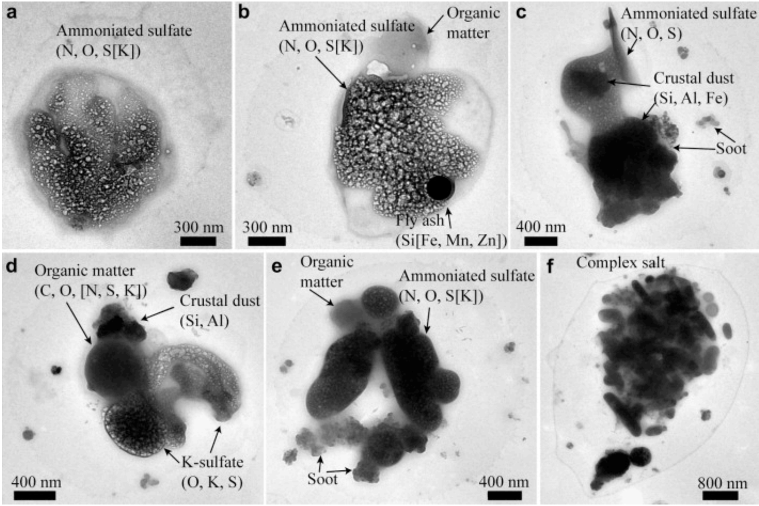


Underlying conceptual model of aerosol particles



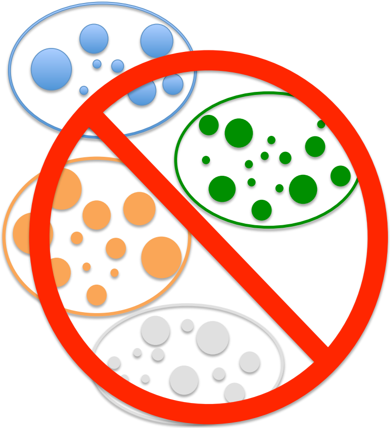
External mixture of different aerosol types

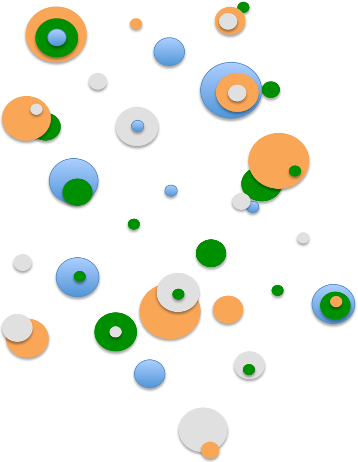
Real particles in the ambient atmosphere



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

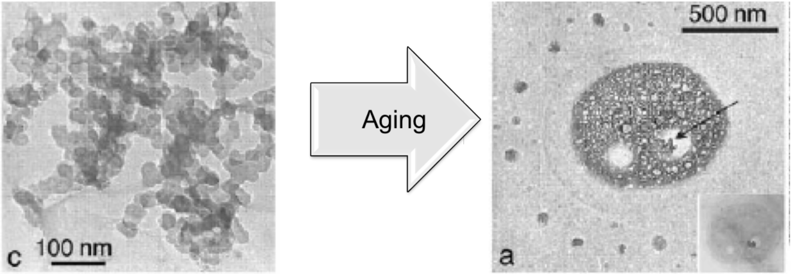
Revised underlying model of aerosol particles





Transformation of black carbon in the atmosphere

Freshly emitted diesel soot "aged" diesel soot



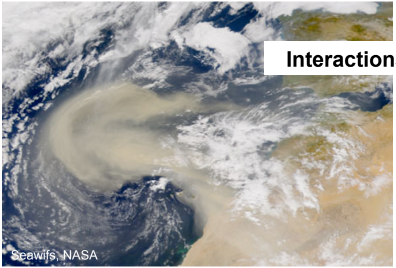
External Internal *Buseck and Pósfai, 1999*

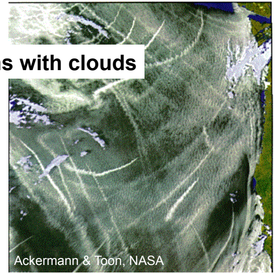
- Freshly emitted soot is hydrophobic.
- Aging due to coagulation and condensation.
- This changes optical properties and hygroscopicity, hence climate impacts.

Are these details important?

Scatter and absorb solar radiation

Interactions with clouds

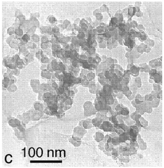


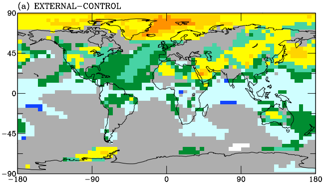


Are these details important?

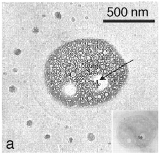
Change in equilibrium annual mean surface air temperature (K)

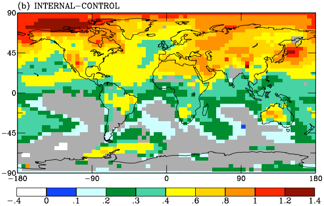
External





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

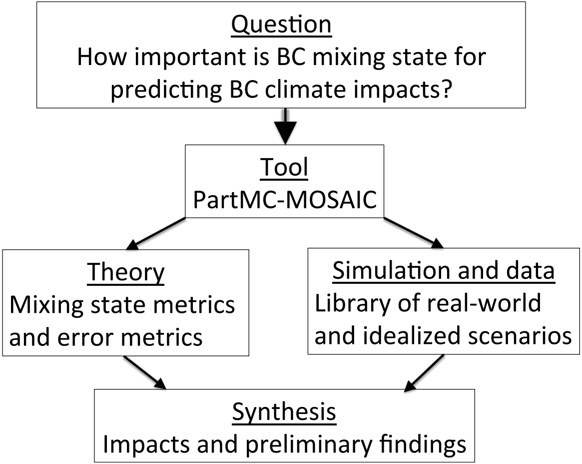
Are these details important?

Table 2. GISS model sensitivity studies.

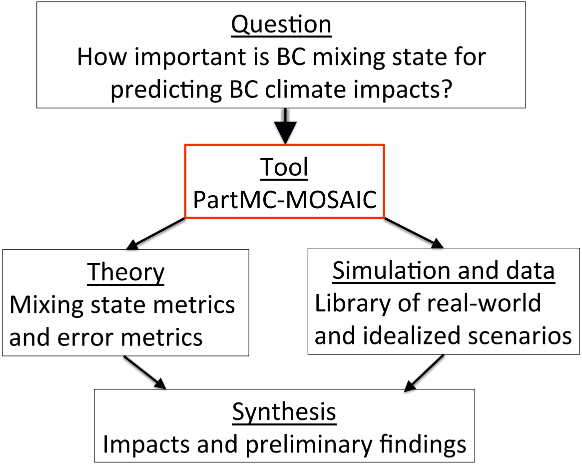
Description	Emission $Tg\,yr^{-1}$	Burden $mg\,m^{-2}$	Lifetime, d	AAOD x100 550 nm
Standard run, see text	7.2 (4.4 energy, 2.8 biomass burning)	0.36	9.2	0.55
EDGAR32 emission	7.5	0.37	9.3	0.58
IIASA emission	8.1	0.41	9.5	0.60
BB 1998	8.2	0.38	8.7	0.58
2x (Faster aging)	7.2	0.29	7.6	0.50
2x (Slower aging)	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

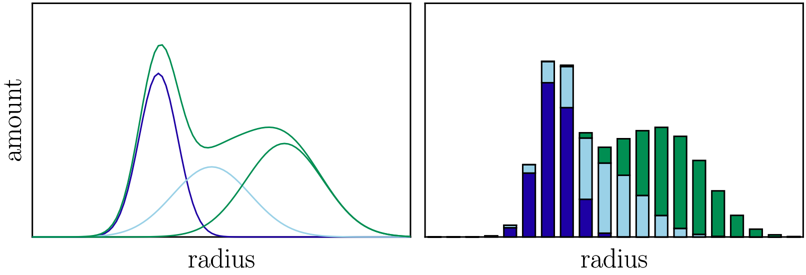
Central research question and strategy



Central research question and strategy



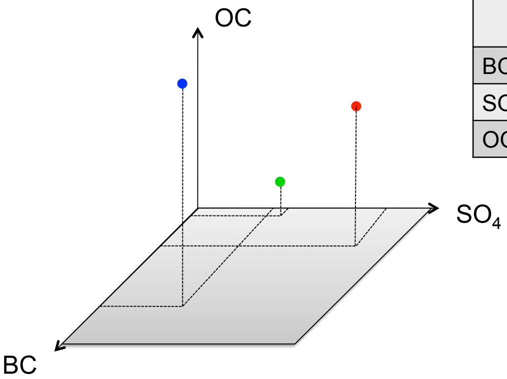
Model aerosol representation



- 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

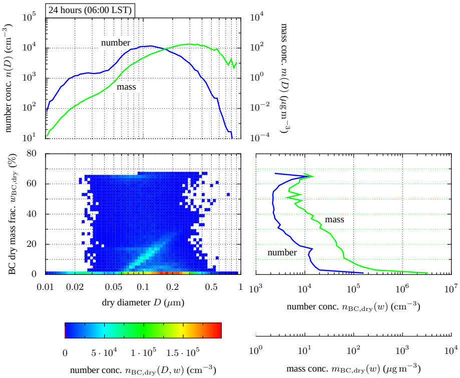
Benefits of particle-resolved models

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[1], MADE3[2], MATRIX, MOSAIC-ext).
3. Detailed studies on the particle scale and experimental intercomparison.

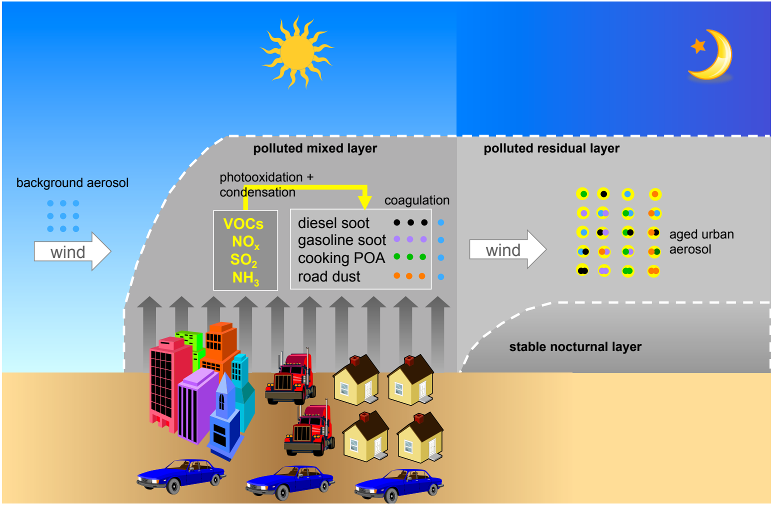
[1] McGraw et al., *Journal of Physics: Conference Series*, 2008
[2] Kaiser et al., *Geosci. Model Dev.*, 7, 1137–1157, 2014

Limitation of particle-resolved models



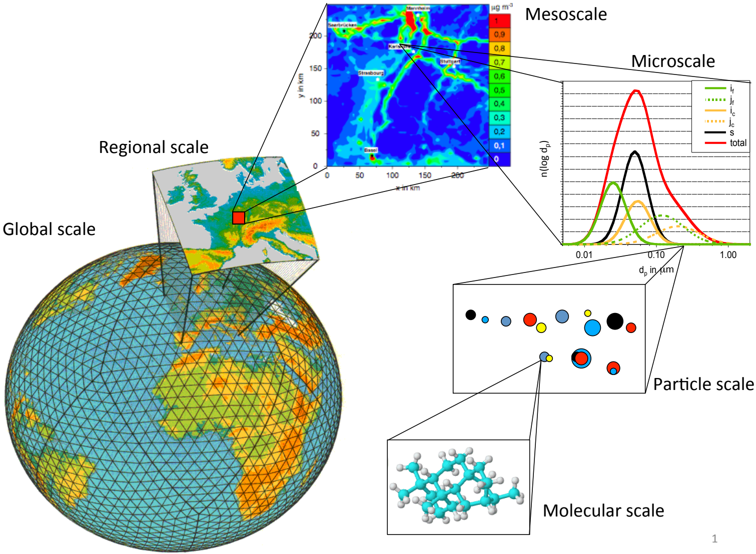
- Resolution: Only finite number of computational particles available per grid cell $(10^4 - 10^7)$.
- On-going research to develop more efficient algorithms, e.g. "weighted particles" (DeVille et al., *J. Comp. Phys.*, 2011) and parallel methods.

Typical model setup

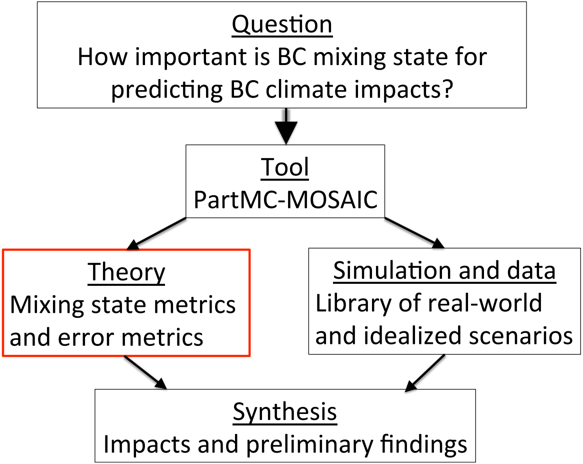


Zaveri, Easter, Riemer, West, JGR 2010

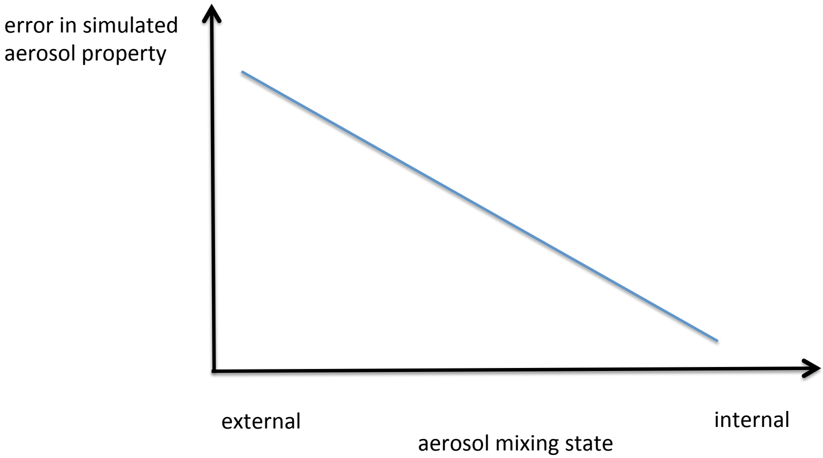
Multiscale model hierarchy



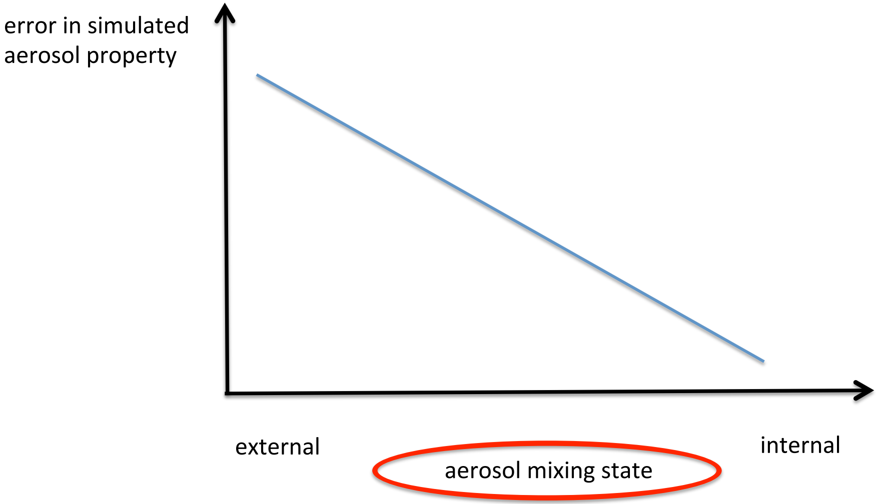
Central research question and strategy



Hypothesis: Error versus mixing state metric

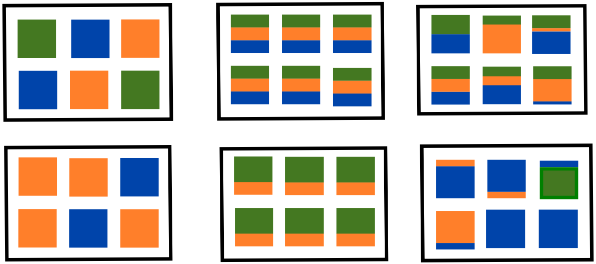


Hypothesis: Error versus mixing state metric



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



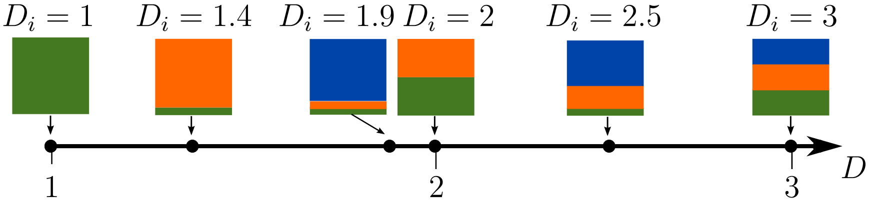
Internally mixed waterholes



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

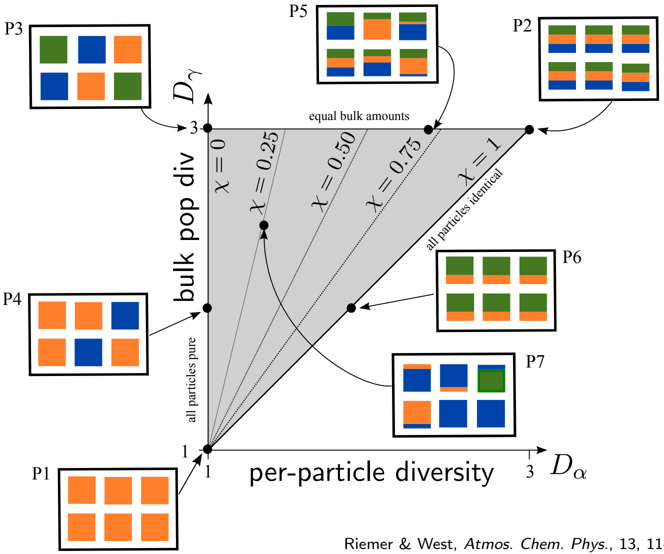
Single-particle diversity

- p_i^a = mass fraction of species a in particle i
- mixing entropy of ith particle: $H_i = -\sum_a p_i^a \ln p_i^a$
- diversity of ith particle: $D_i = e^{H_i}$ (units of *effective species*)



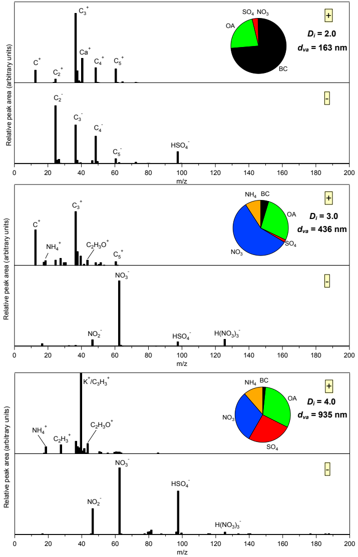
- average particle diversity: $D_\alpha = e^{H_\alpha}$, $H_\alpha = \sum_i p_i H_i$
- bulk population diversity: $D_\gamma = e^{H_\gamma}$, $H_\gamma = -\sum_a p^a \ln p^a$
- Mixing state index: $\chi = \frac{D_\alpha - 1}{D_\gamma - 1}$
- χ varies between 0 (externally mixed) to 1 (internally mixed).

Mixing state diagram



Riemer & West, *Atmos. Chem. Phys.*, 13, 11423–11439, 2013

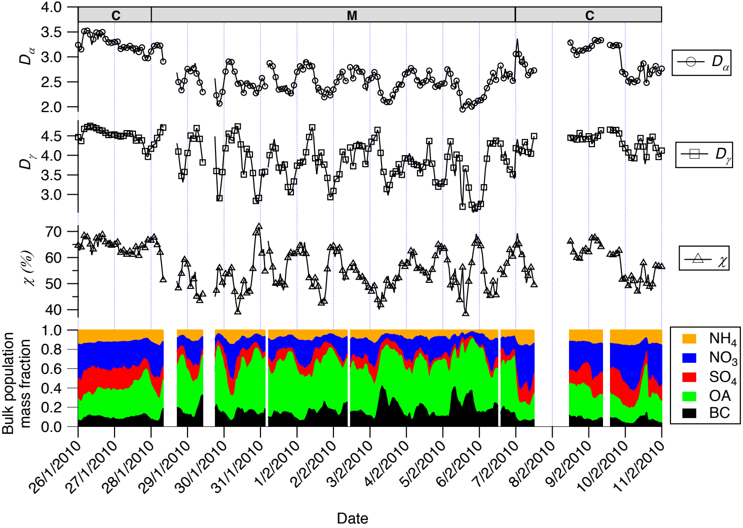
Application to field data: MEGAPOLI



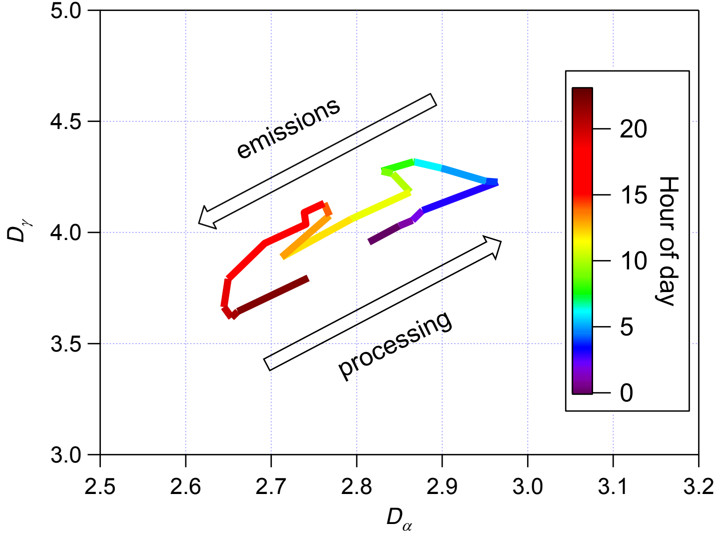
- Collaboration with Robert Healy, University of Toronto.
- Paris, France, winter 2010.
- Mass fractions estimated based on ATOFMS data, supplemented by AMS, SMPS and MAAP.
- Five species: OA, BC, $\mathrm{SO_4}$, $\mathrm{NO_3}$, $\mathrm{NH_4}$.

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

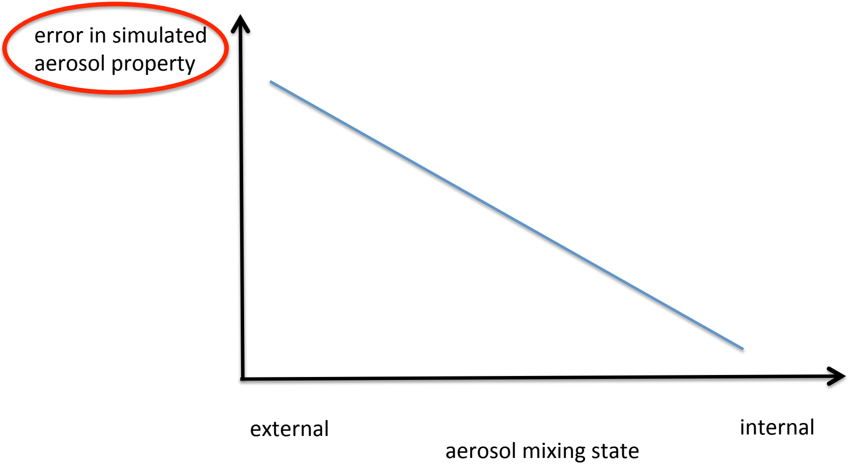
Mixing state parameters during the campaign



Average diurnal cycle

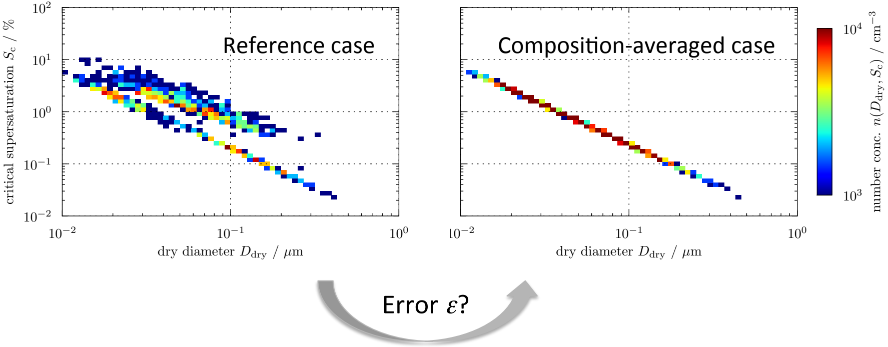


Back to the hypothesis

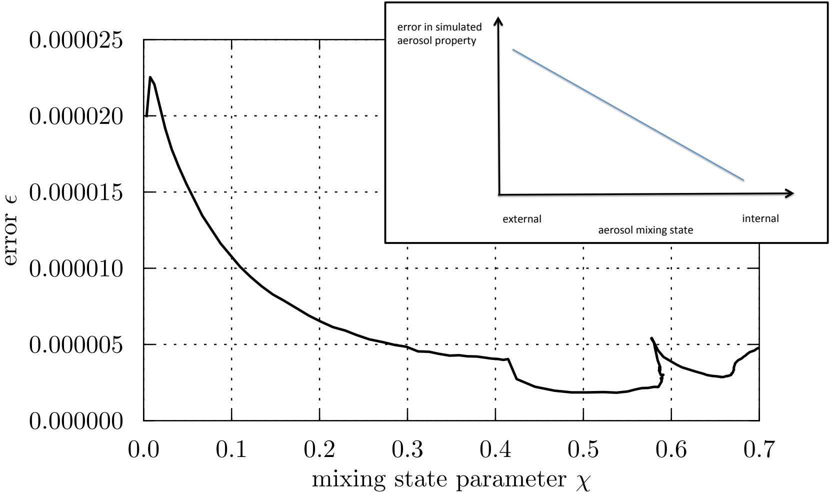


Quantifying the importance of mixing state

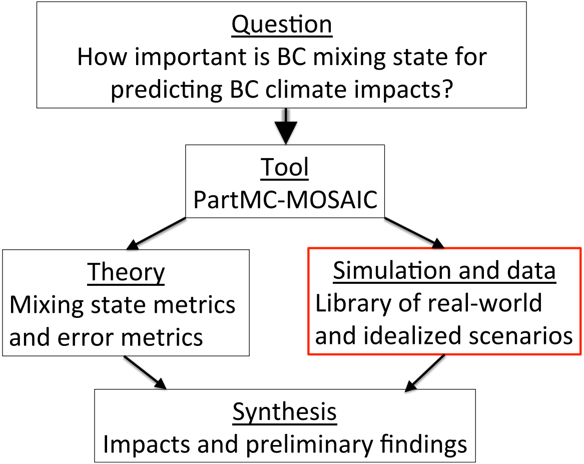
Here: for CCN properties



Error ϵ and mixing state parameter χ

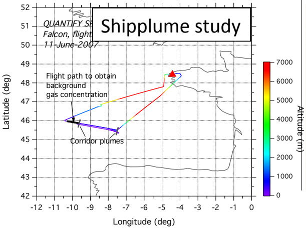


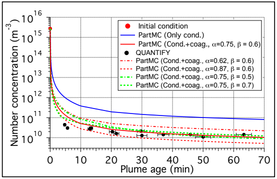
Central research question and strategy



Real-world scenarios

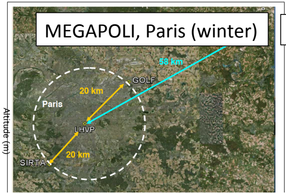
Shipplume study

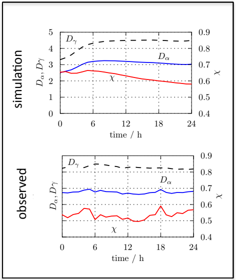




Aging: Combustion particles coagulate with small sulfuric acid particles.

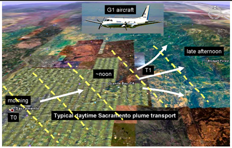
MEGAPOLI, Paris (winter)

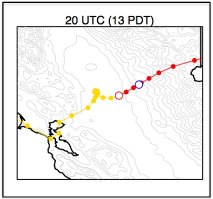




Aging: Ammonium nitrate formation during the night (if any).

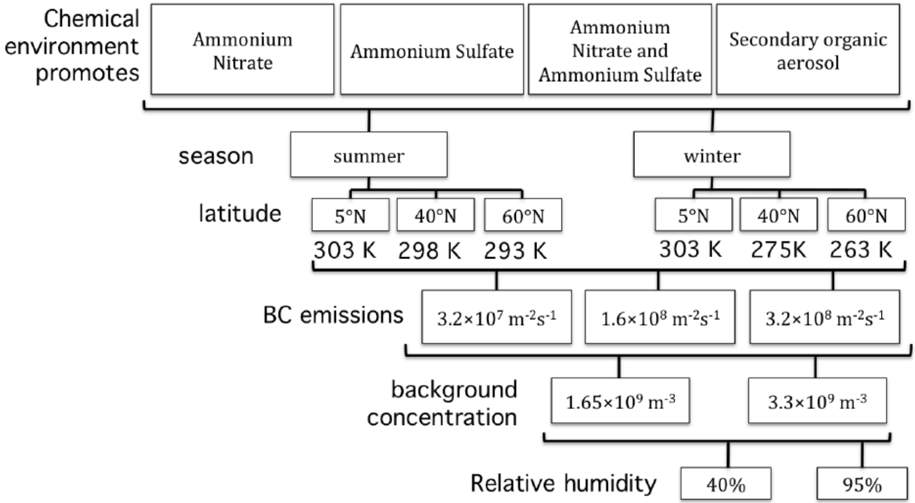
CARES, Sacramento (summer)



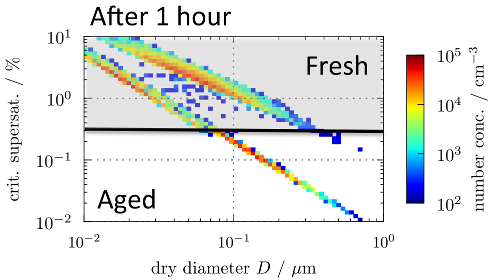


Aging: ???
Current work

Library of idealized scenarios



CCN-based aging criterion

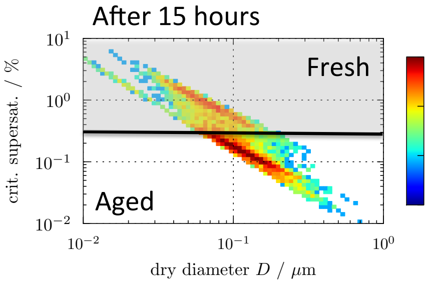


N_f = number fresh

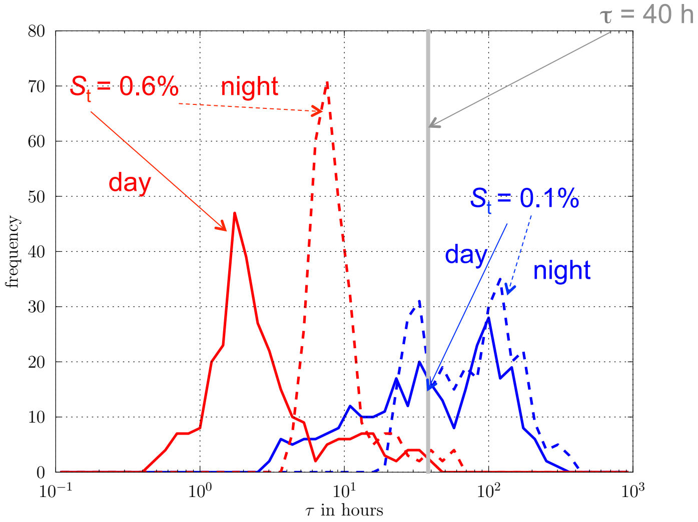
τ = aging time-scale

S_* = threshold supersat.

$$\left(\frac{dN_\mathrm{f}}{dt}\right)_\mathrm{aging} = -\frac{1}{\tau(t,S_*)} N_\mathrm{f}(t, S_*)$$



Distribution of aging time-scales



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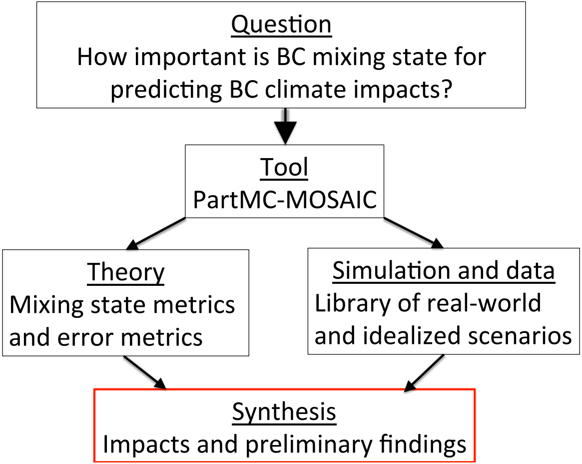
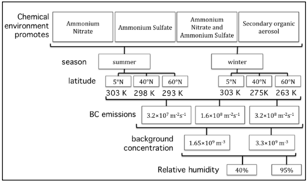
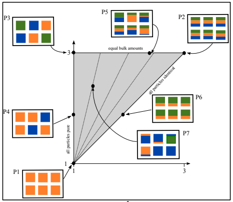


Impacts on community

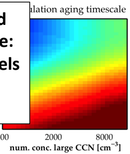
Scenario library

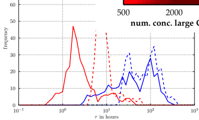


Mixing state metrics



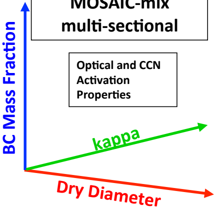
With Tami Bond and Laura Fierce: Predictive models of aging time scales



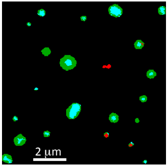


With PNNL: MOSAIC-mix multi-sectional

Optical and CCN Activation Properties



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



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Preliminary conclusions and current work

How important is BC mixing state for predicting BC climate impacts?

- Depends on target quantity: CCN properties or optical properties? (Zaveri, Barnard, Easter, Riemer, West, JGR, 115, D17210, 2010)
- Good news—sometimes—regarding CCN properties.
- Optical properties more complicated.