Unraveling aerosol mixing state: Enhancing climate impact predictions through particle-resolved simulations

Nicole Riemer

Department of Atmospheric Sciences, University of Illinois at Urbana-Champaign

with Jeff Curtis, Yu Yao, Laura Fierce, Tami Bond, Joseph Ching, Zhonghua Zheng, Feng Xu, Matt West

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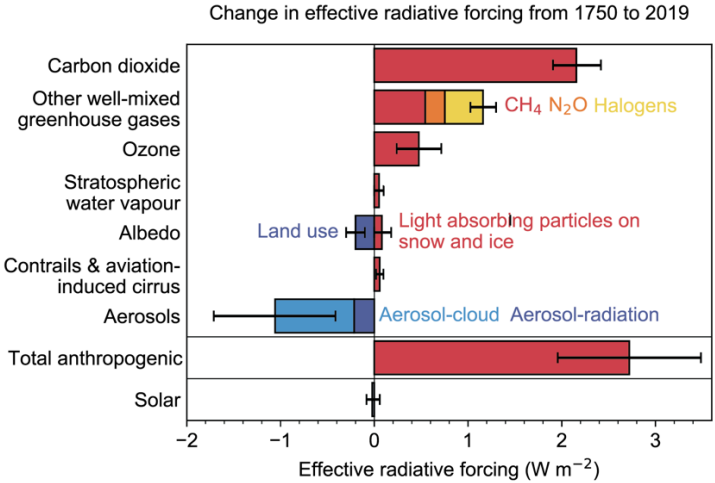
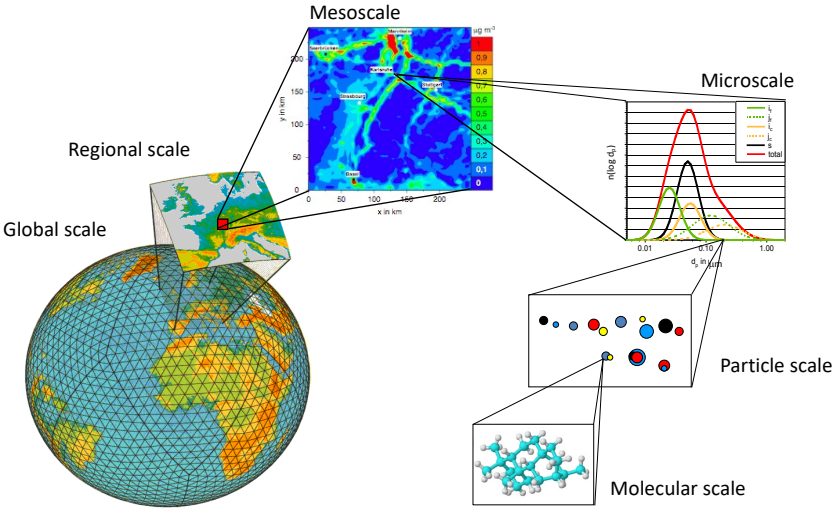
Microscale processes impact large-scale climate

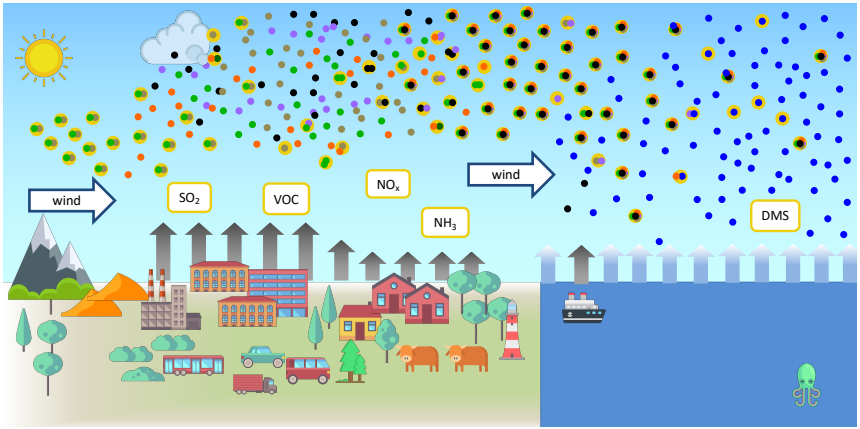


IPCC report, 2022.

Aerosol science — a multiscale problem



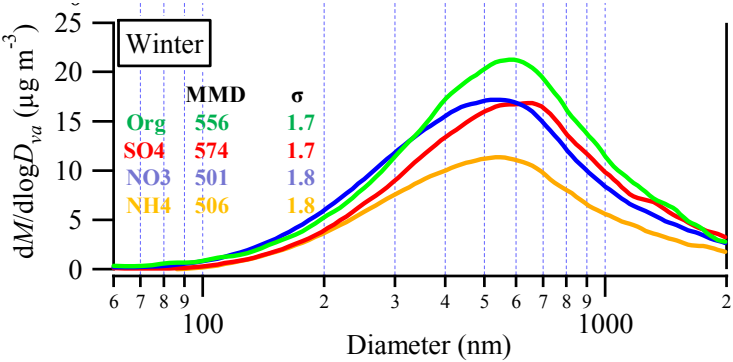
Particle composition evolves in the atmosphere



- Aerosols are complicated mixtures.
- Important for CCN properties, optical properties, IN properties.

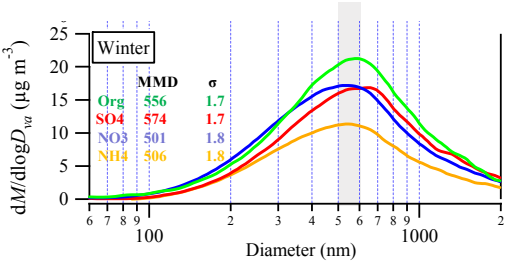
Riemer, Ault, Craig, Curtis, West, *Rev. Geophys.*, 2019

From distributions to particles



Zhang et al., *Atmospheric Chemistry and Physics*, 2014

What is the composition of the particles?



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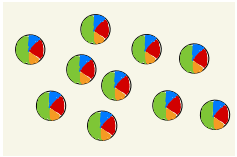
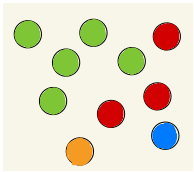
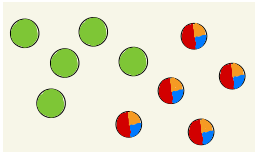


B.



C.



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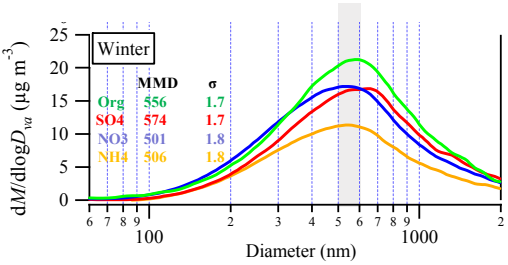

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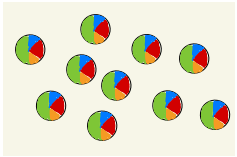

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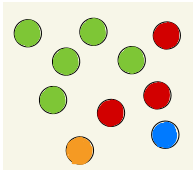
B.



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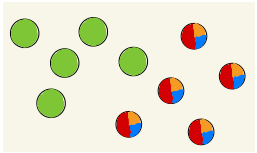
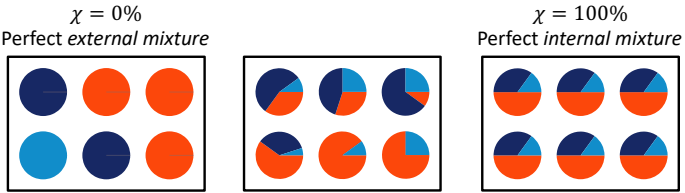


✅

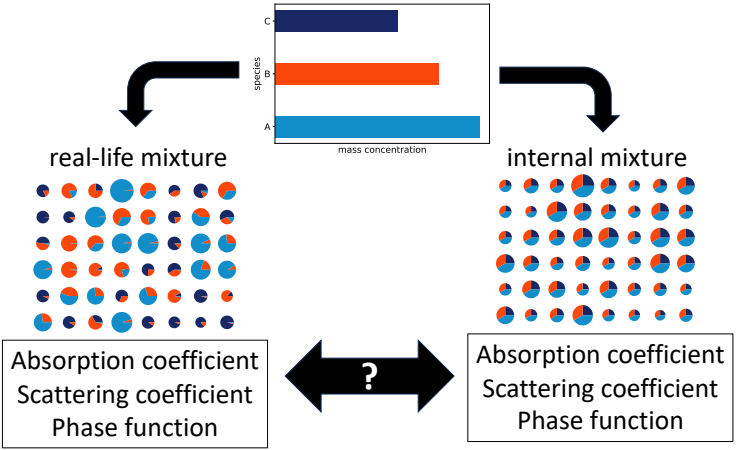
Definition of aerosol mixing state

- Aerosol mixing state: Distribution of chemical species across the population.
- Property of the population.

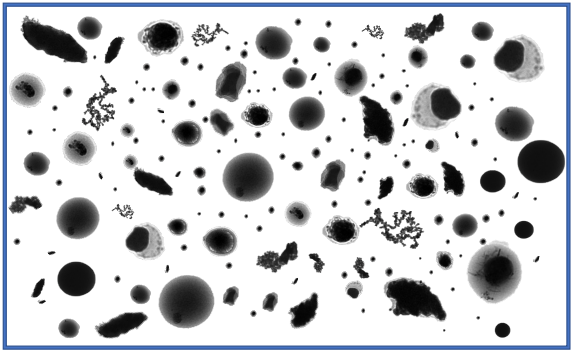
"The same net composition of an aerosol can be caused by an infinite variety of different internal distributions of the various compounds." (Winkler, 1973)



Why does this matter?

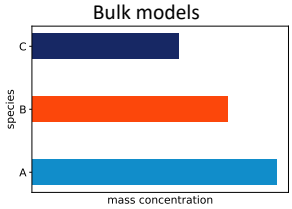


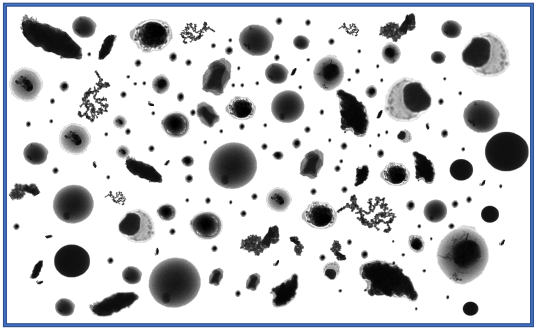
From aerosol state to model state

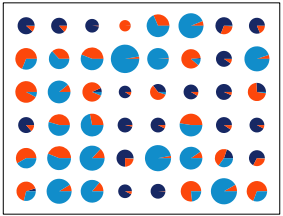


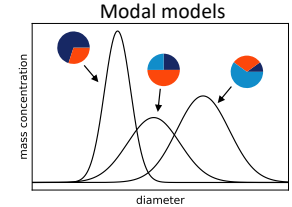
Particle images courtesy of Miriam Freedman

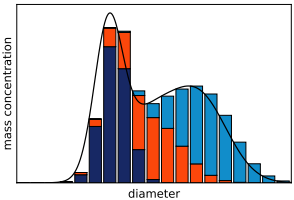
From aerosol state to model state











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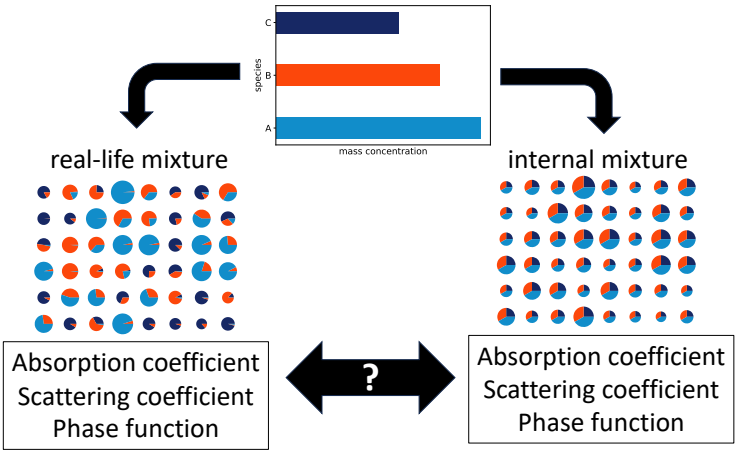
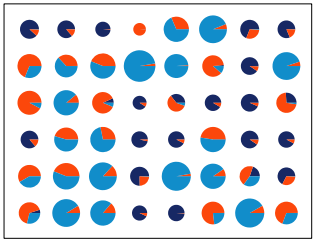


- Modal and sectional models are distribution-based.
- Inherent assumption: All particles in one mode/bin have the same composition.
- Mixing state is an "unknown unknown" — structural uncertainty of the model.

Particle-resolved modeling 101



PartMC

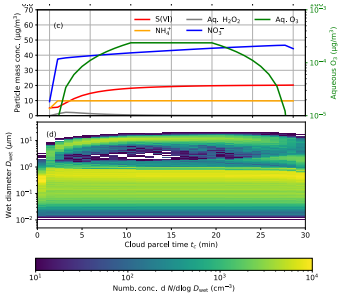
- No modes or bins
- Instead: discrete computational particles

- **PartMC:** simulates coagulation, particle emissions, dilution, deposition, nucleation stochastically (Riemer et al., 2009, DeVille et al., 2011, 2019, Curtis et al., 2016, 2017, 2023).
- **MOSAIC:** simulates gas phase chemistry, aerosol thermodynamics deterministically (Zaveri et al., 2008).

- Evolution of mixing state is straight-forward to represent.
- Computationally expensive!

Impacts of aqueous phase chemistry in cloud droplets

Yao et al., JGR, 2021



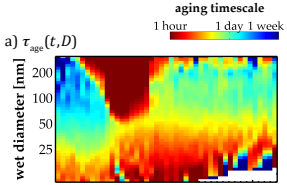
Water uptake and optical properties of mixed organic-inorganic particles

Nandy et al., AST, 2021

AST figures removed to respect copyright protections

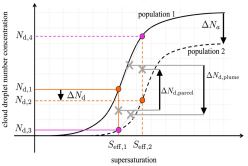
Variance in black carbon's aging timescale

Fierce et al., ACP, 2015



Black carbon mixing state impacts on clouds

Ching et al., JGR, 2016



Mixing state impacts on CCN spectra

Razafindrambinina et al., AST, 2021

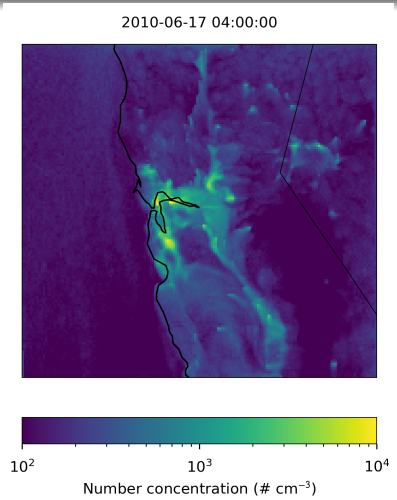
How many particles do we need to estimate mixing state?

Gasparik et al., AST, 2020

Mixing state evolution in a chamber

Shou et al., AST, 2019

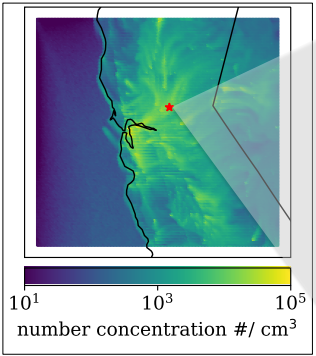
Particle-resolved modeling on the regional scale

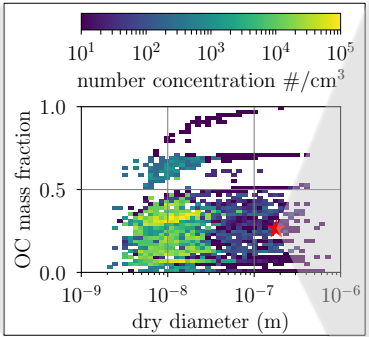


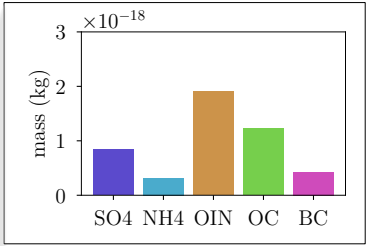
- Host model WRF: simulates windfield, temperature, humidity
- PartMC: simulates advection, turbulent diffusion stochastically (in addition to aerosol dynamics)
- MOSAIC: chemistry processes

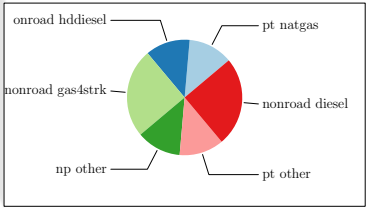
- 170 x 160 x 40 domain
- 6656 cores
- Simulate 5000 particles per grid cell to capture aerosol mixing state
- 10 billion total particles in the simulation domain

Each grid cell contains the full aerosol state information



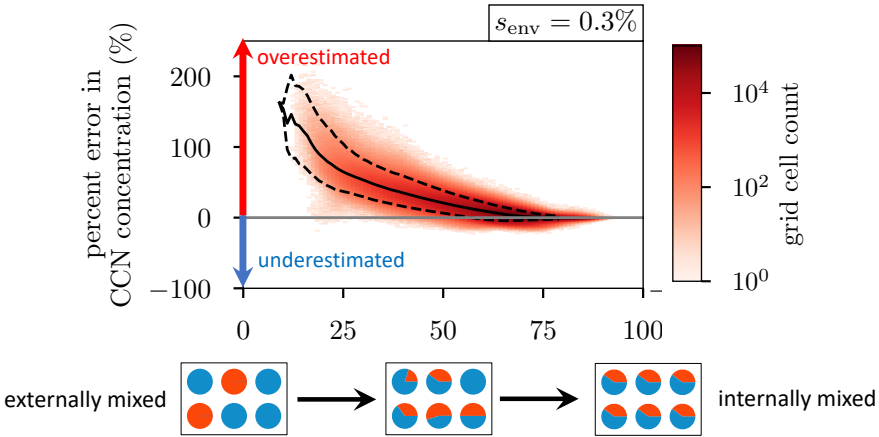






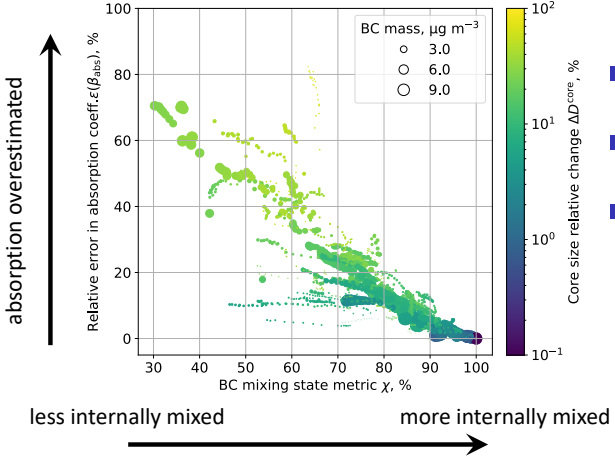
Curtis, Riemer, West, *GMD*, 2017; under review with *GMD* 2024

Error in CCN concentration depends on aerosol state



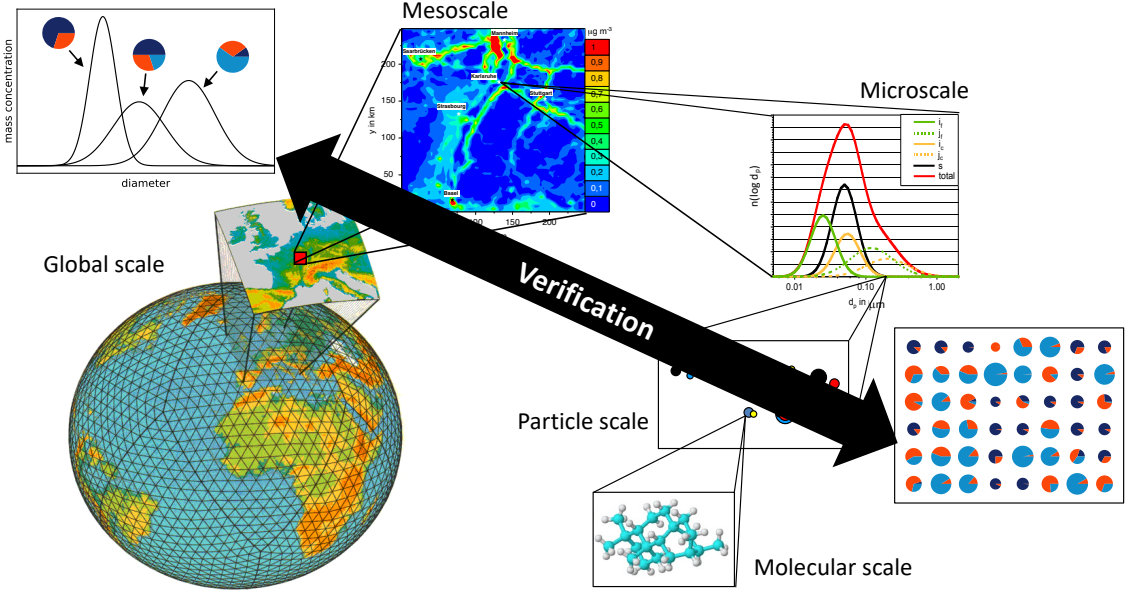
- Use all populations from WRF-PartMC California simulation.
- Assuming internal mixture leads to *overestimation* of CCN concentration.

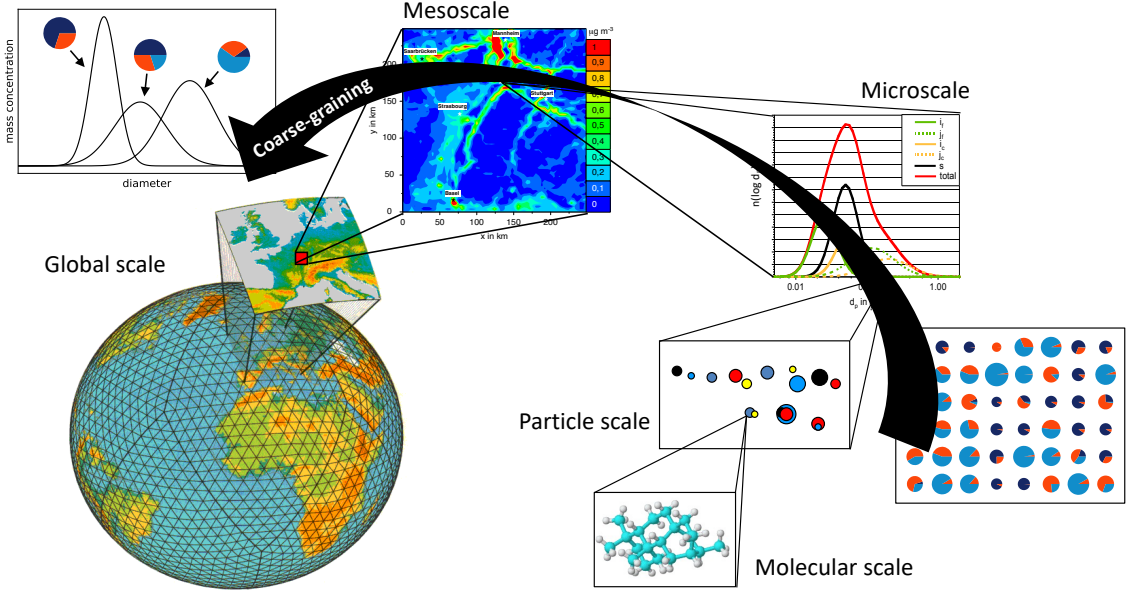
Assuming internal mixture leads to . . .



- . . . overestimation in aerosol absorptivity,
- . . . underestimation in aerosol scattering,
- . . . underestimation in single scattering albedo.

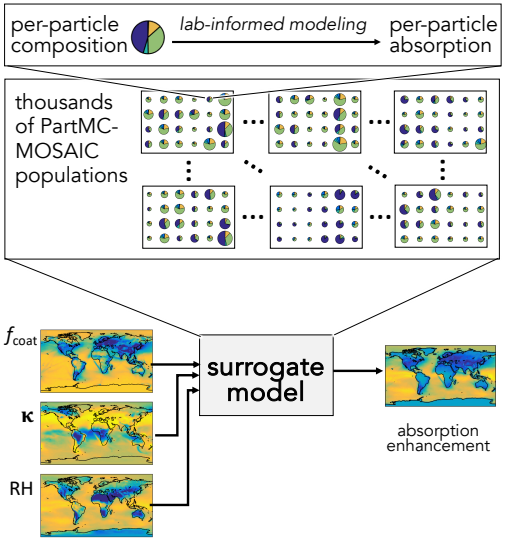
Yao, Curtis, Ching, Zheng, Riemer, *ACP*, 2022.


Mesoscale
Microscale
Global scale
Verification
Particle scale
Molecular scale
mass concentration
diameter
µg m⁻³
y in km
n(log d_p)
d_p in µm
total


mass concentration
diameter
Coarse-graining
Mesoscale
μg m-3
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
Saarbrücken
Mannheim
Stuttgart
Strasbourg
Basel
y in km
x in km
Microscale
n(log d
total
0.01
1.00
Global scale
Particle scale
Molecular scale

Diversity in composition affects BC absorption

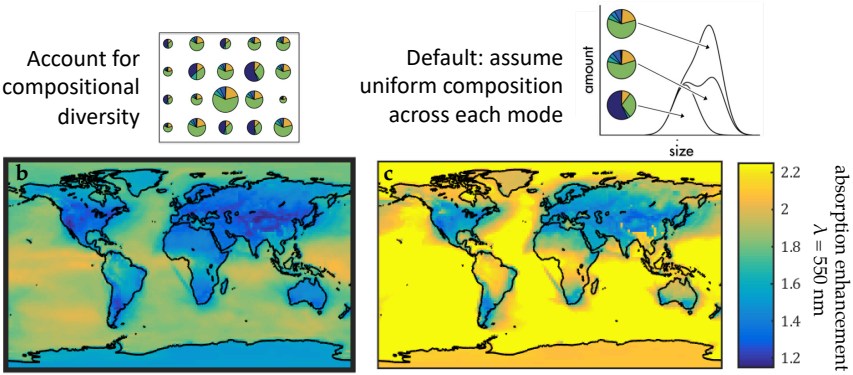
- **Challenge:** Particle-resolved model is expensive.
- **Solution:** Surrogate model that approximately reproduces particle-resolved model predictions.



Fierce, Bond, Bauer, Mena, Riemer, *Nature Comm.*, 2016.

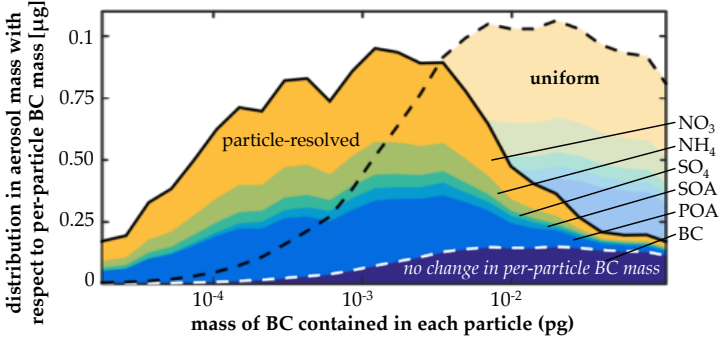
Surrogate model improves predictions of aerosol absorption

- Predict BC absorption enhancement with output from NASA GISS MATRIX model.



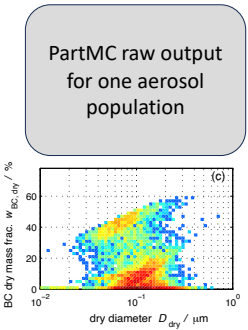
Fierce, Bond, Bauer, Mena, Riemer, *Nature Comm.*, 2016.

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.

Use PartMC data for satellite retrieval verification



PartMC raw output for one aerosol population

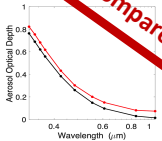
Mie code

Population-level (reference)
- AOD
- SSA

RT code

Radiance at TOA

Retrieval



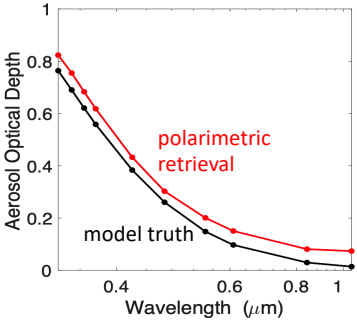
compare

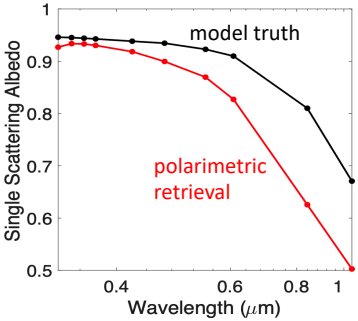
Population-level (retrieval)
- AOD
- SSA

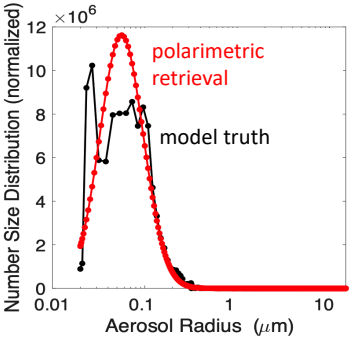
Assumption:
Two aerosol species – each with its own refractive index and log-normal size distribution

Collaboration with Feng Xu, Univ. of Oklahoma

Proof of concept for one example population

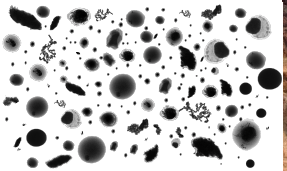


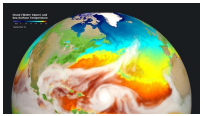




- PartMC particle populations mimic ambient populations with realistic mixing states.
- "Closure" exercise with PartMC data allows to assess validity of retrieval assumptions.

Particle-resolved modeling bridges scales in aerosol science





- Particle-resolved modeling (combined with machine learning) is a key tool in the model hierarchy to:
  - bridge from the particle scale to larger scales
  - quantify structural uncertainties in aerosol models.
- This modeling approach is now feasible for use in 3D Eulerian chemical transport models.

PartMC uses open source