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# Particle-Resolved Simulations for Quantifying Black Carbon Climate Impact and Model Uncertainty

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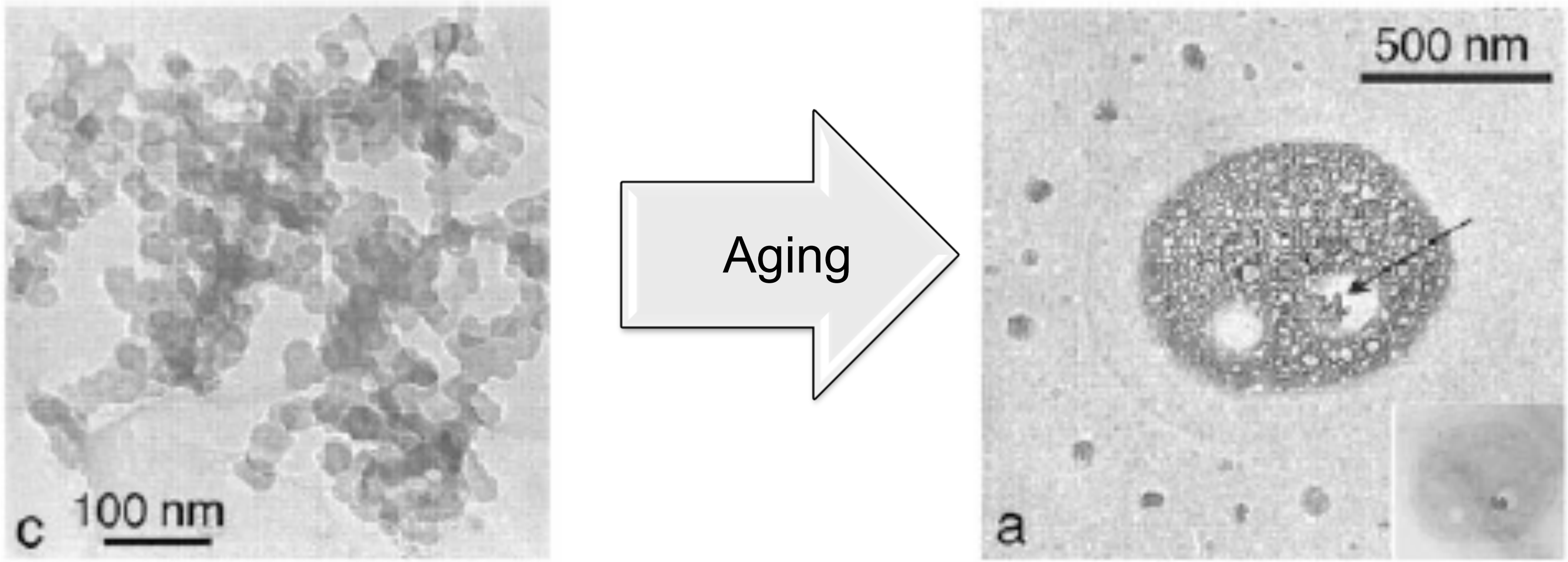
# Transformation of Black Carbon in the Atmosphere

Freshly emitted diesel soot

"aged" diesel soot: Internal mixture of soot and sulfate




*Buseck and Pósfai, 1999*

- Freshly emitted soot is hydrophobic.
- Transfer into internal mixture by condensation and coagulation.
- Impact on optical properties, growth behavior and life time.
- Challenge for models: **Need to represent mixing state**

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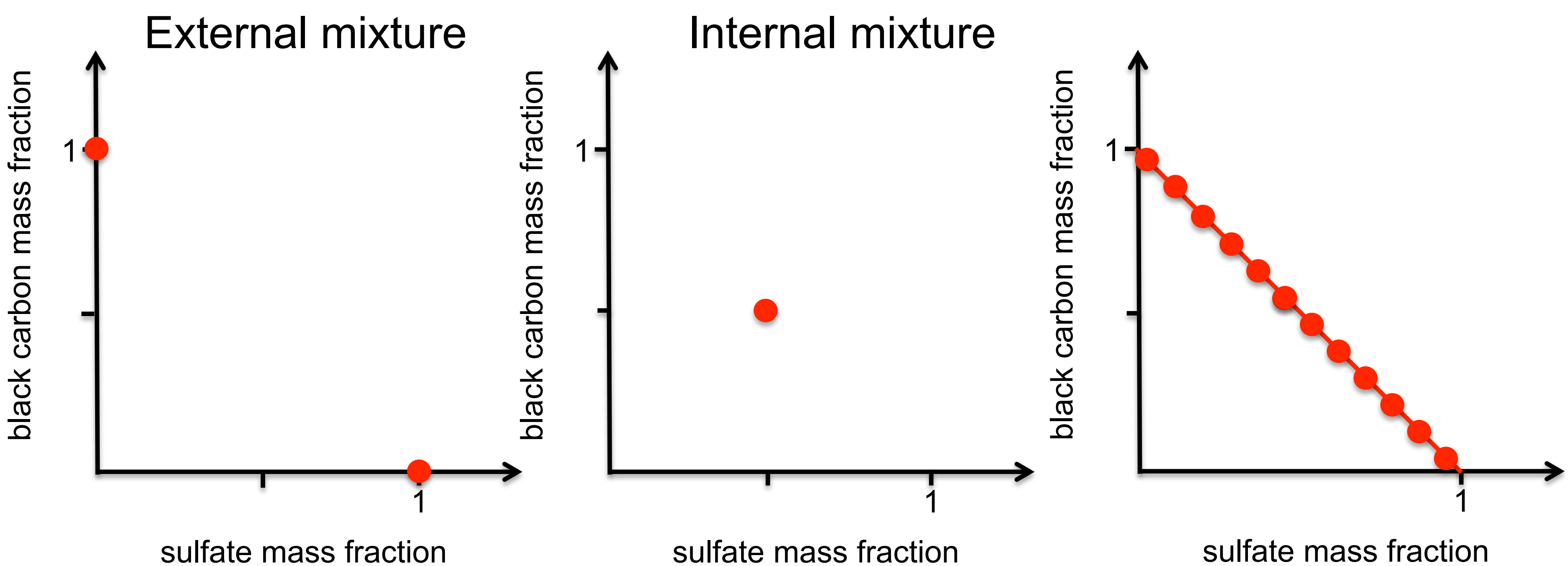
# Aerosol Mixing State

- Mixing state: Distribution of the per-particle composition
- Aging: Evolution of mixing state



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# Model Representation of Black Carbon

- Need atmospheric chemistry transport models on the regional and global scale to develop black carbon mitigation strategies
- Representation of black carbon in these models is a weak link:
  - Introduces unknown uncertainties in simulation
- This project addresses this on a fundamental level:
  - Parameter estimation
  - Error quantification
  - Testbed evaluations
- Key enabling tool: PartMC (Particle-resolved aerosol model)

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# Model Representation of Black Carbon

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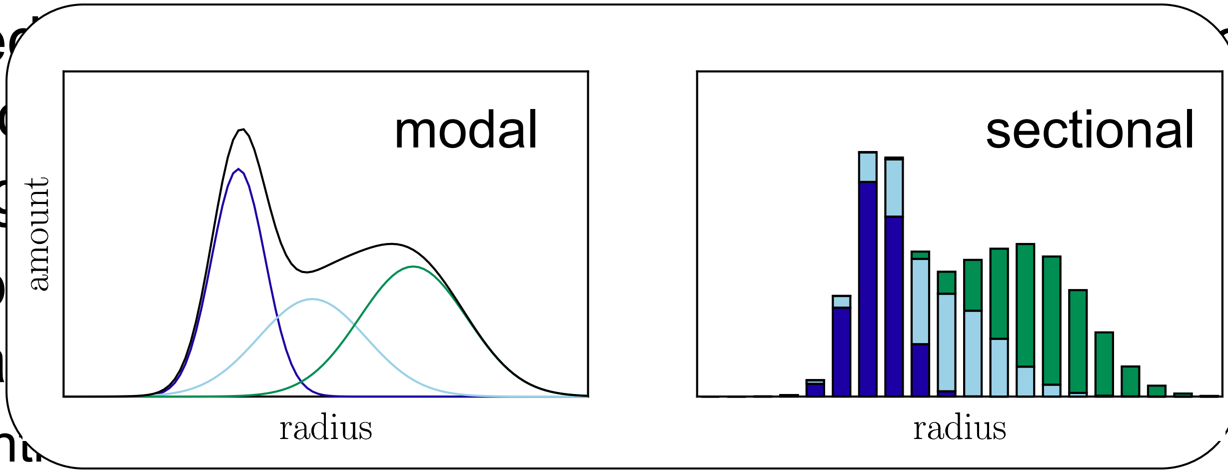


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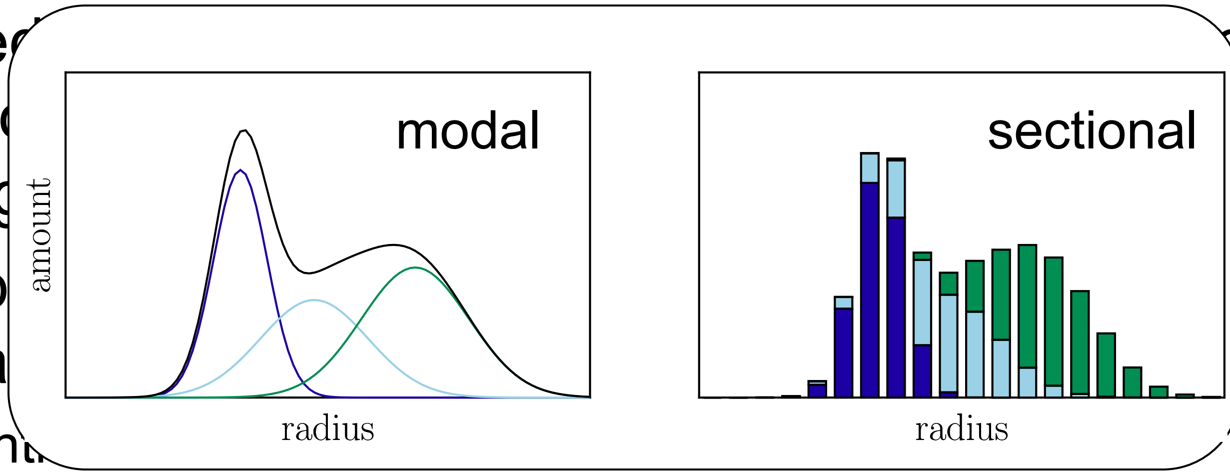


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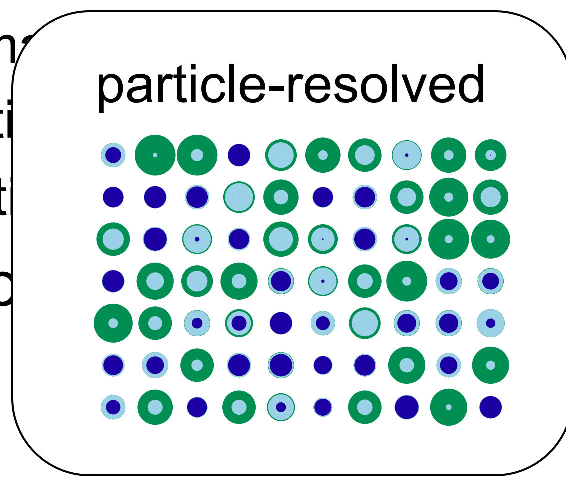


- Key enabling too … resolved aerosol model)

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# Objectives

- Objective 1:
  - Calculate key quantities for modeling black carbon effects relevant for climate simulations:
    - aging timescale
    - optical properties
    - cloud condensation nuclei (CCN) number concentration
- Objective 2:
  - Quantify the error in these quantities in climate predictions resulting from inadequate representation of black carbon aerosol mixing state in existing models.
- Objective 3:
  - Provide a testbed for the evaluation of proposed new approximate aerosol modeling algorithms, including multi-dimensional sectional models.

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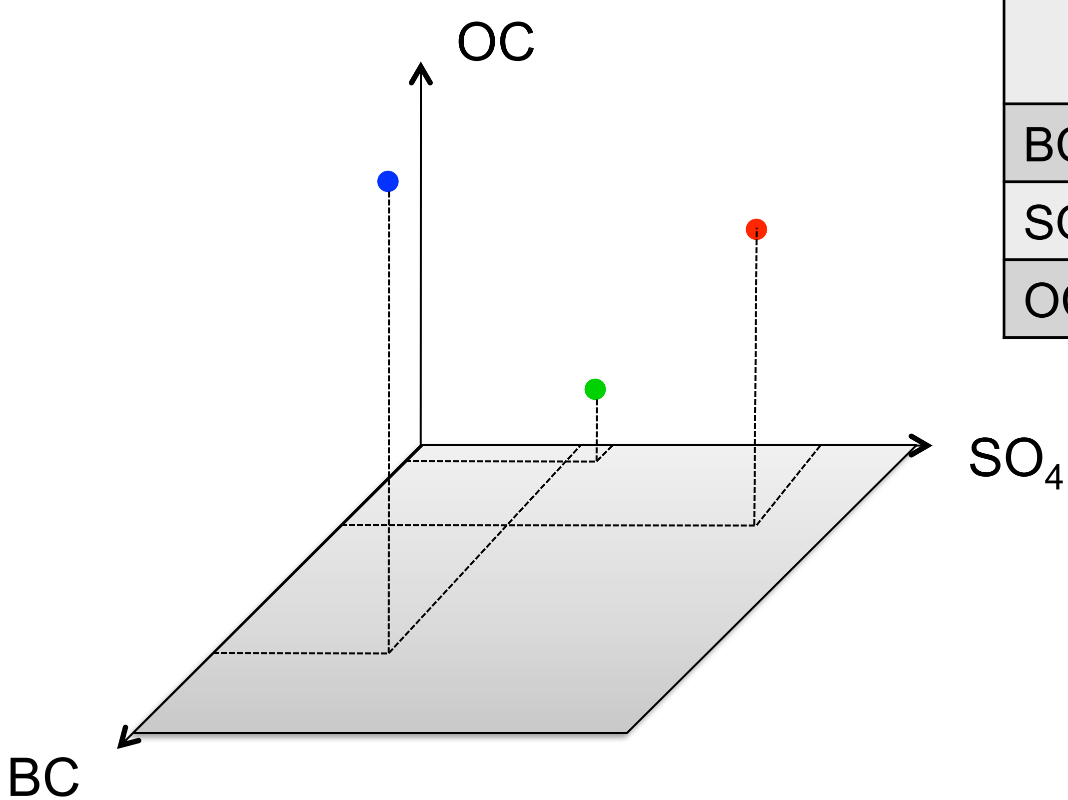
# What Are Particle-Resolved Aerosol Models?

- 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 |

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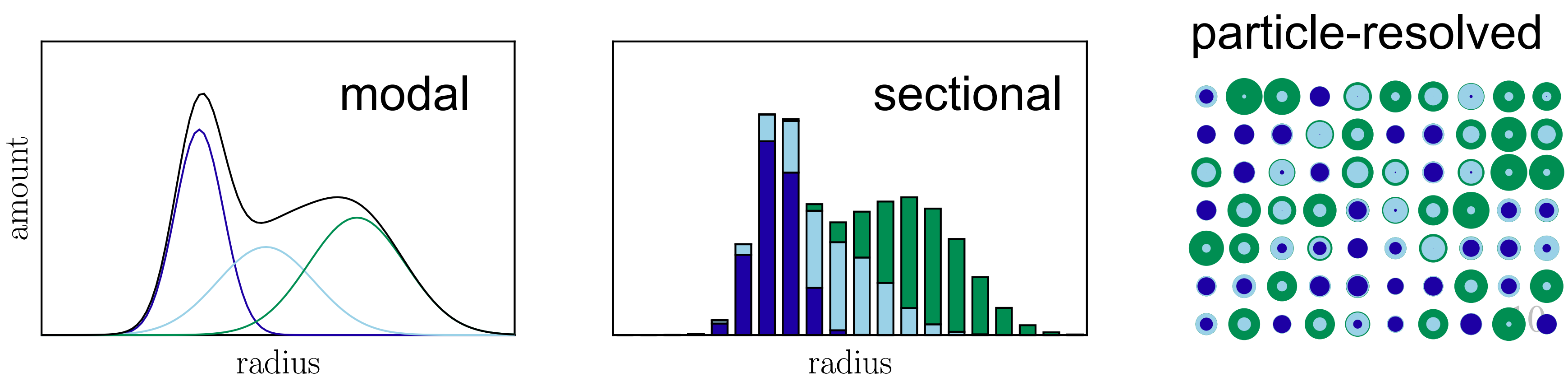
# Why Particle-Resolved Aerosol Models?

- Advantages:
  - Efficient representation of multidimensional size distribution
  - Advantageous whenever the per-particle-composition (= “mixing state”) is of interest: No assumptions about mixing state are needed.
- Disadvantage: Expensive to run
  - Each particle costs the same as one section or mode.
  - Need 1,000 to 100,000 particles



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# Box Model PartMC-MOSAIC

- **PartMC: Particle-resolved Monte Carlo aerosol model**
  - Stochastic treatments for emission, dilution, and coagulation
  - Riemer, N., M. West, R.A. Zaveri, and R.C. Easter, Simulating the evolution of soot mixing state with a particle-resolved aerosol model, *JGR.*, 114, D09202, 2009.

- **MOSAIC: Model for Simulating Aerosol Interactions & Chemistry**
  - Deterministic treatments for gas photochemistry, aerosol thermodynamics, and dynamic gas-particle mass transfer
  - Zaveri, R.A., R.C. Easter, J.D. Fast, and L.K. Peters, Model for Simulating Aerosol Interactions and Chemistry (MOSAIC), *JGR.*, 113, D13204, 2008.

- **Aerosol Properties Modules:**
  - **Kappa-Köhler Model for CCN:** Petters, M.D. and S.M. Kreidenweis, A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, *Atmos. Chem. Phys.*, 7, 1961-1971, 2007.
  - **ACKMIE Shell-Core Model for Optics:** Ackerman, T.P. and O.B. Toon, Absorption of visible radiation in atmospheres containing mixtures of absorbing and non-absorbing particles, *Appl. Optics*, 20, 3661-3668, 1981.

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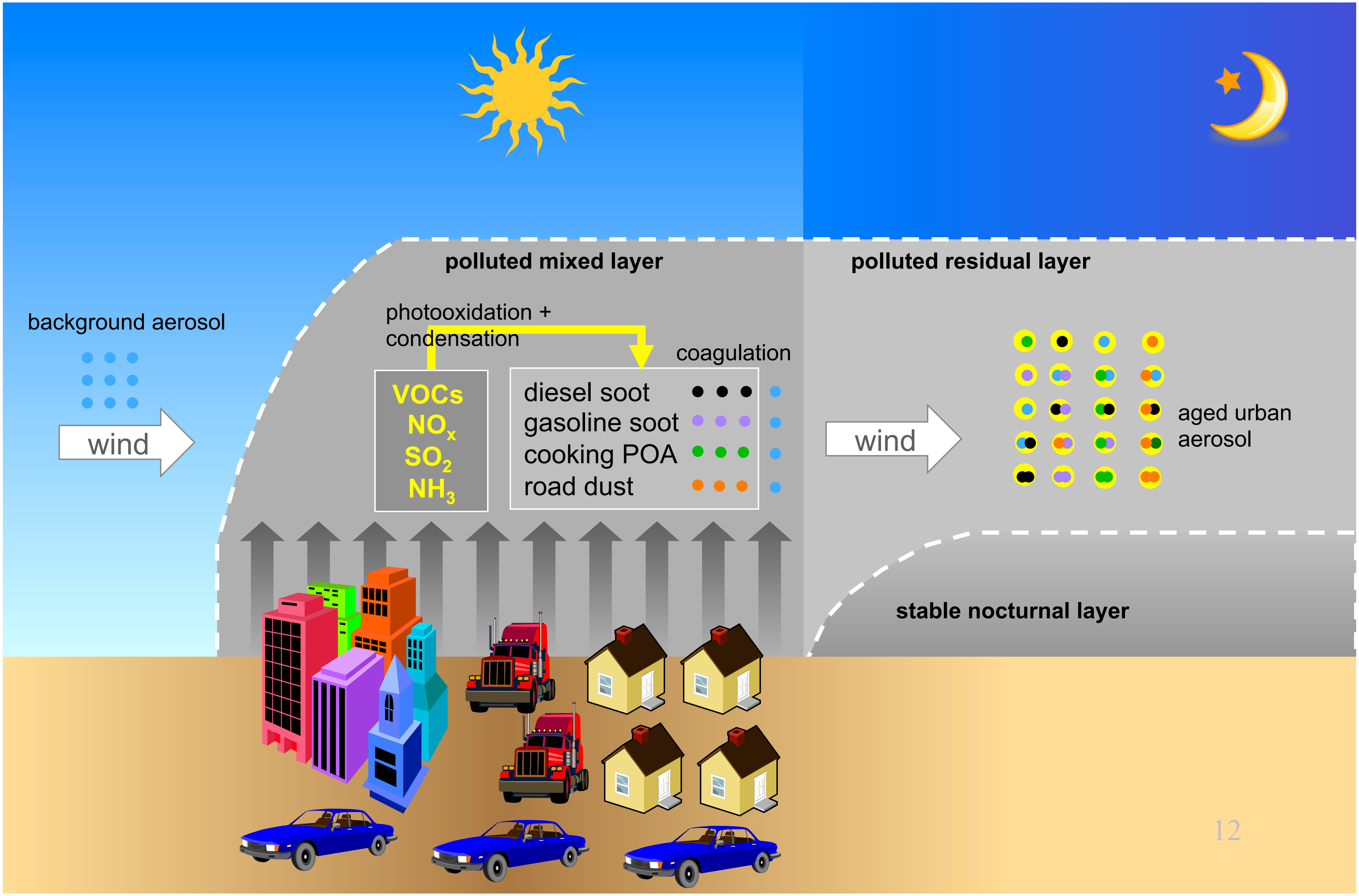
# Idealized Urban Plume Scenario



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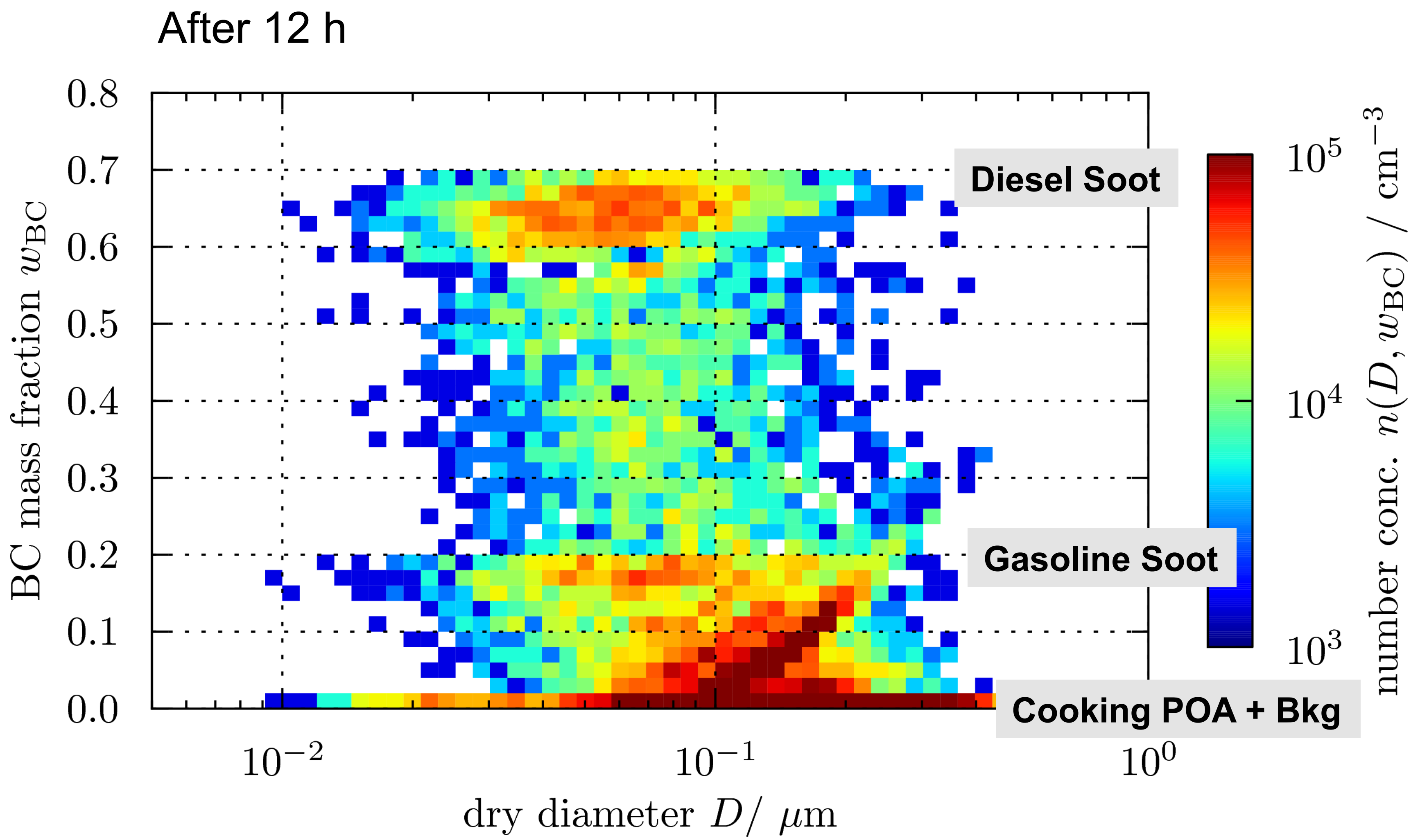
# Evolution of Black Carbon Mixing State



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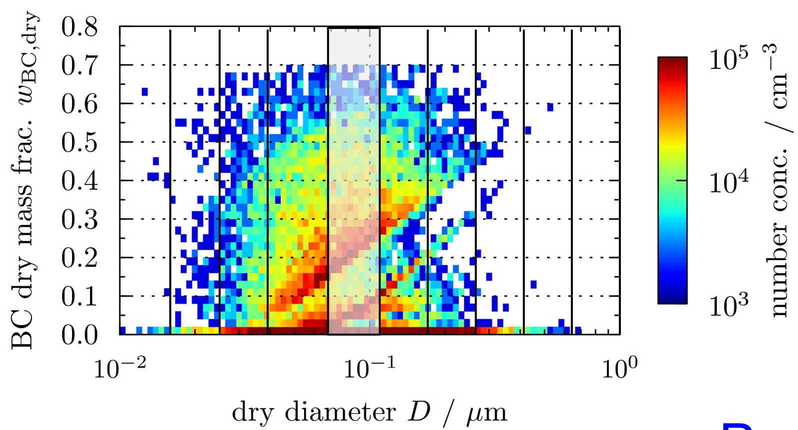
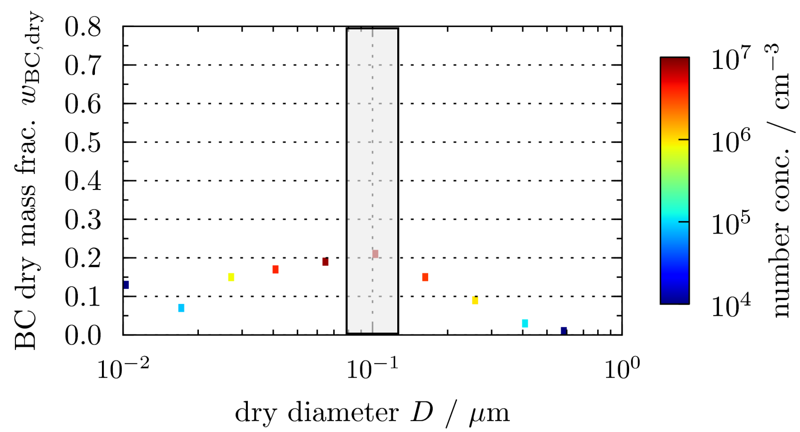
# Error Quantification

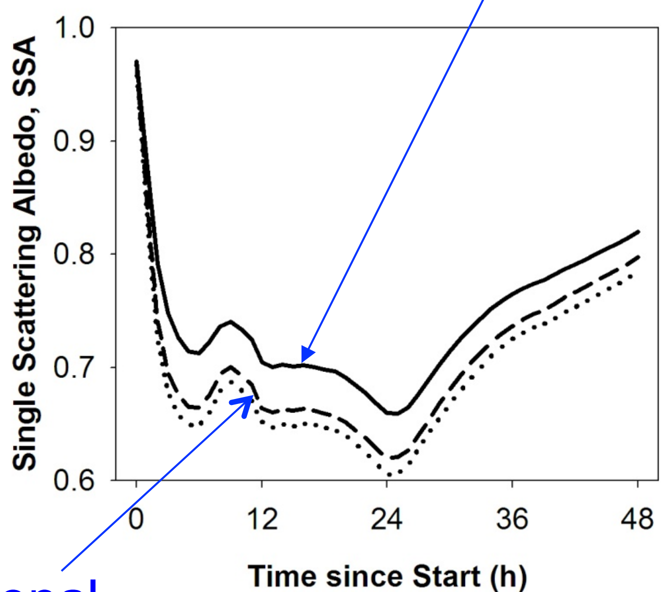
Zaveri, Barnard, Easter, Riemer, West, *JGR*, 115, D17210, 2010

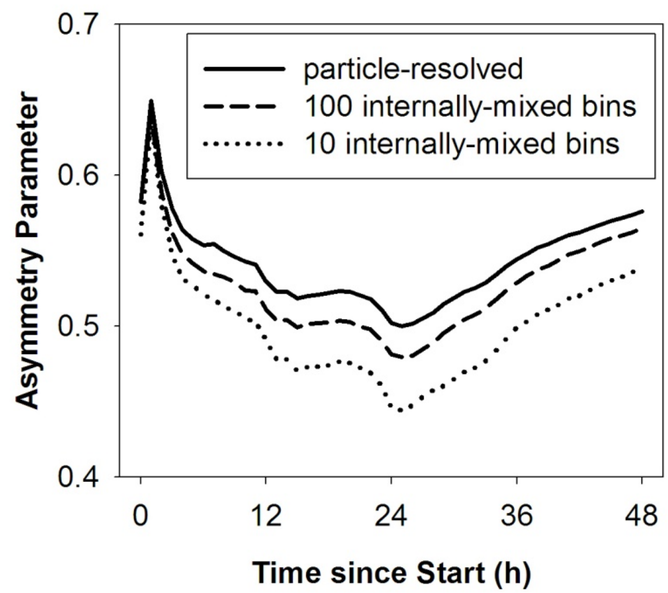



project to sectional




Particle-resolved




sectional

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# Parameter Estimation: Population Aging Timescales

(Riemer et al., *J. Aerosol Sci.*, 2010)

$$\dot{N}^{\mathrm{aging}}(t) = \boxed{\frac{1}{\tau_{\mathrm{N}}(t)}} N_{\mathrm{f}}(t)$$

known known

- Aging criterion: Activation at supersaturation threshold $S_t$
- Determine number of black carbon particles that transition from "fresh" to "aged" by comparing two successive time snapshots.
- Infer aging timescale.

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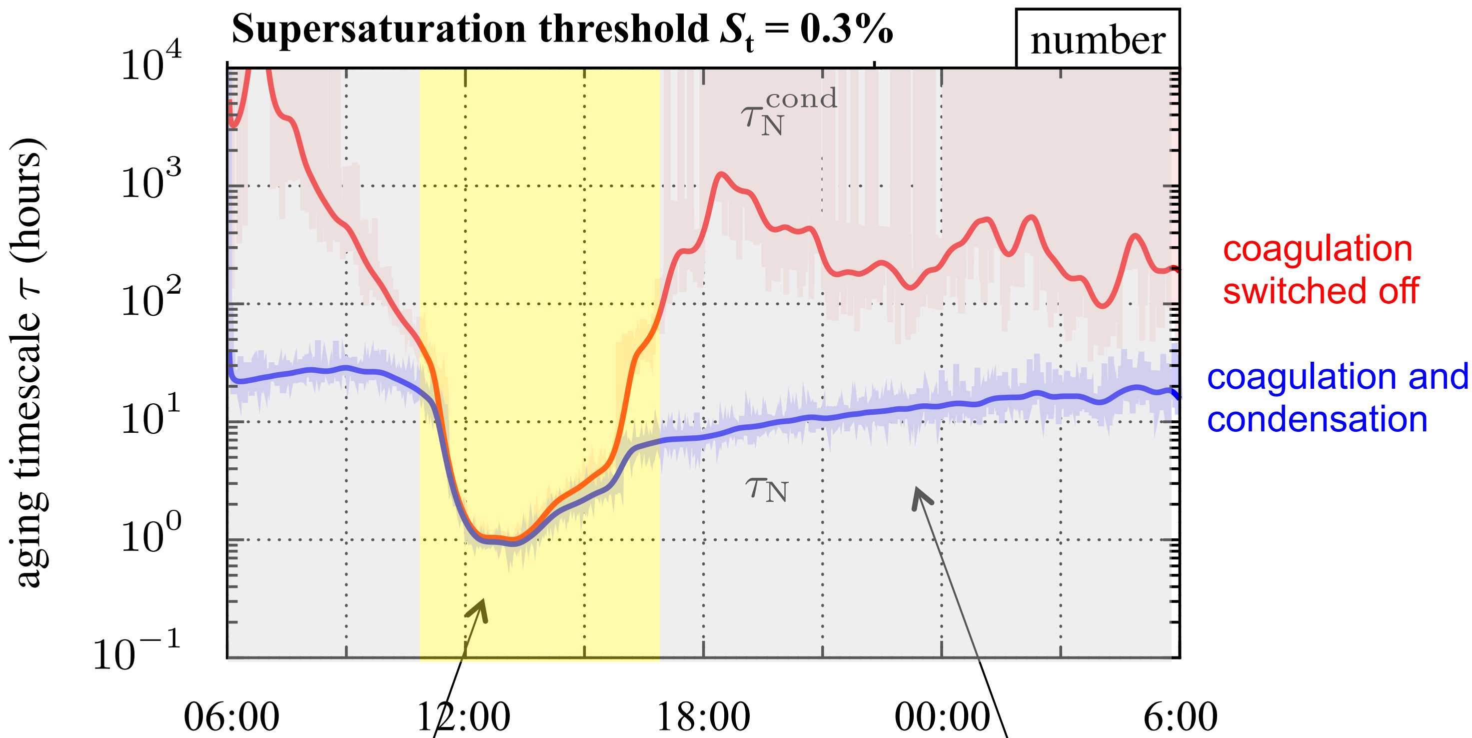
# Population Aging Timescales

(Riemer et al., *J. Aerosol Sci.*, 2010)




Day:
- Fast aging
- Condensation dominated

Night:
- Slower aging
- Coagulation dominated

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# Hypotheses

Based on a very limited number of model simulations:

- Hypothesis 1:
  - First-order aging time scales, used in global models, are too long.
- Hypothesis 2:
  - SSA is underestimated when using internal mixture assumption.
- Hypothesis 3:
  - CCN number is overestimated when using internal mixture assumption.
- Hypothesis 4:
  - Semivolatile aerosol species can cause reverse aging.

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# Approach

- Task A:

  Construct a suite of case studies for PartMC-MOSAIC, representative of different geographical locations and environments.
- Task B:

  Validate PartMC-MOSAIC for selected scenarios against experimental data from recent field campaigns.
- Task C:

  Compute key quantities in each case for black carbon impact.
- Task D:

  Quantify errors in these key quantities due to simplifying assumptions in traditional aerosol models
- Task E:

  Formulate numerical parameter estimates and usage recommendations for global and regional climate models.

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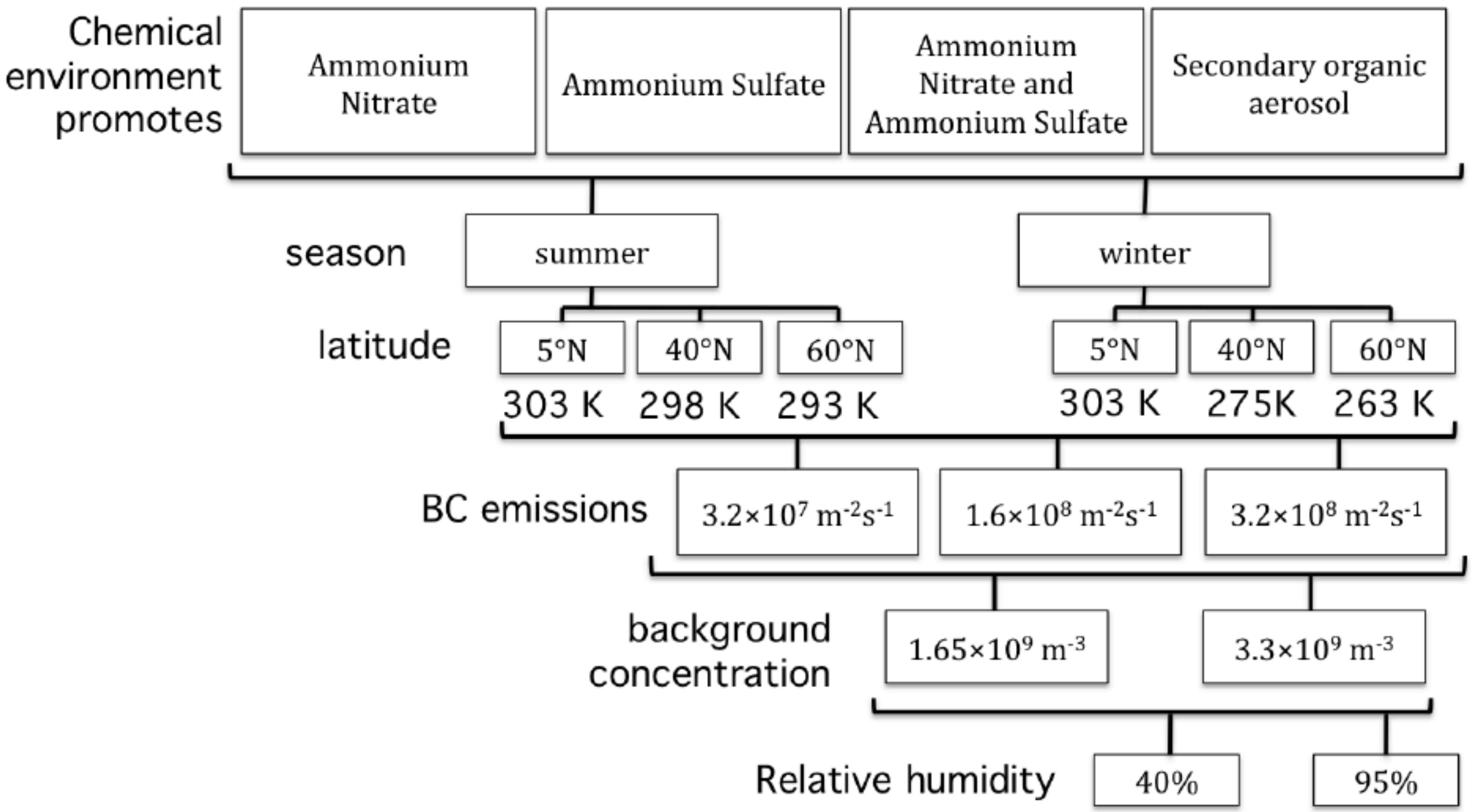


Scenario library of idealized urban plume scenarios

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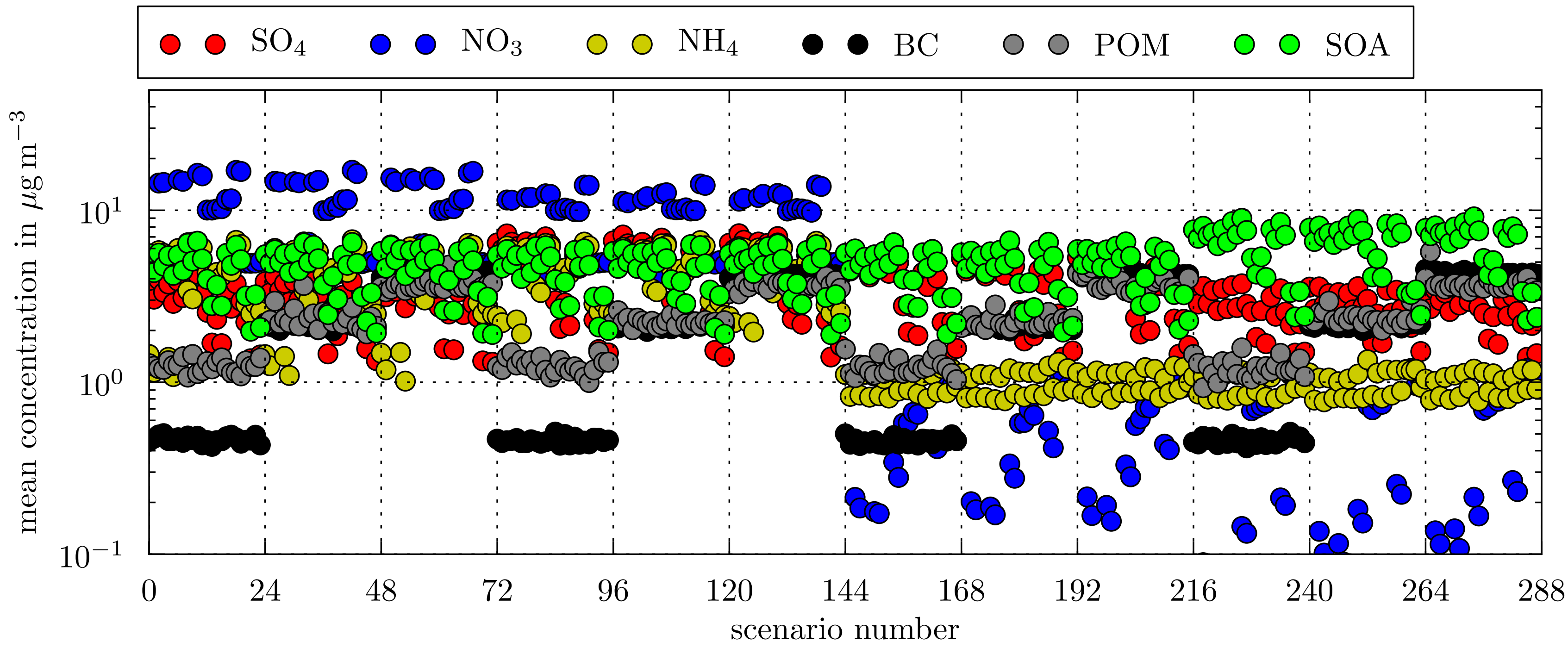

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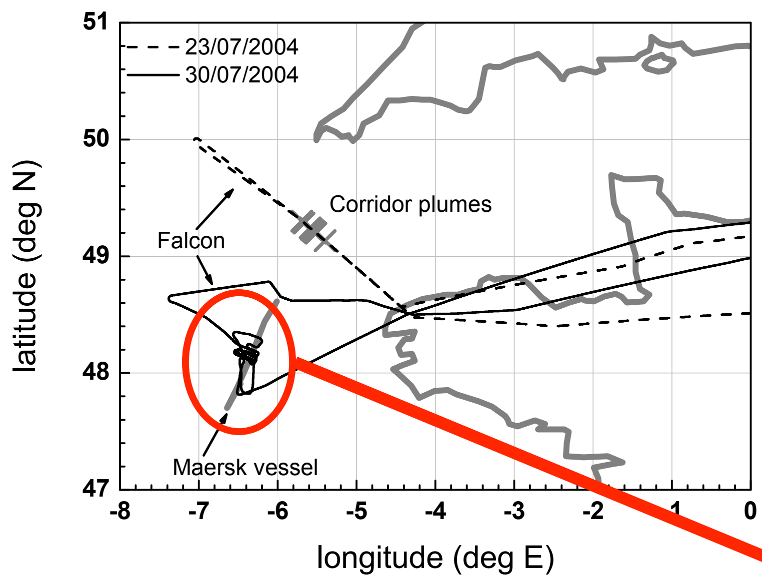
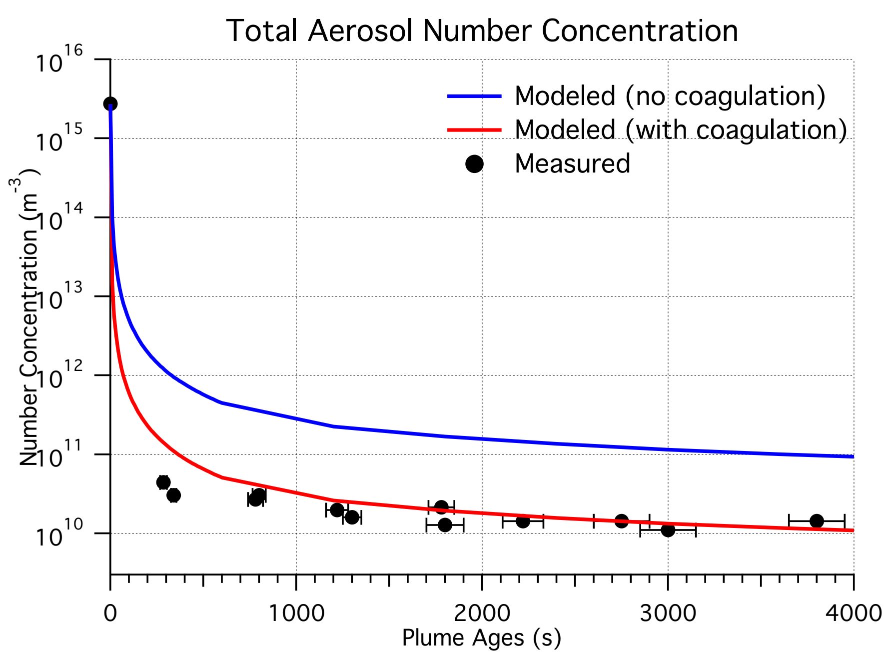
# Task B: Validate PartMC-MOSAIC for selected scenarios against experimental data from recent field campaigns

Particle-resolved modeling of an evolving ship plume in the English Channel (QUANTIFY study)



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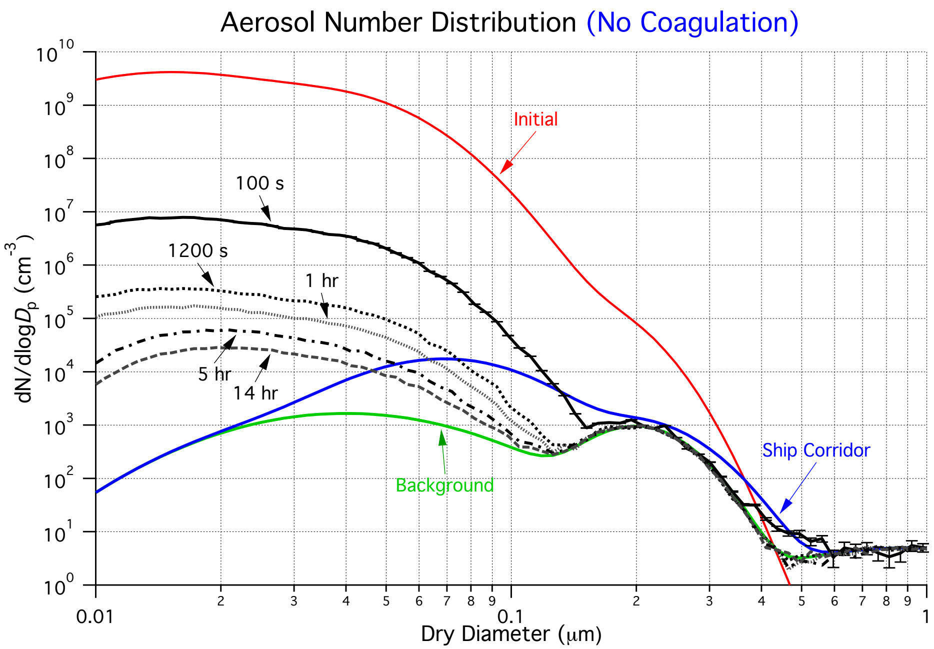
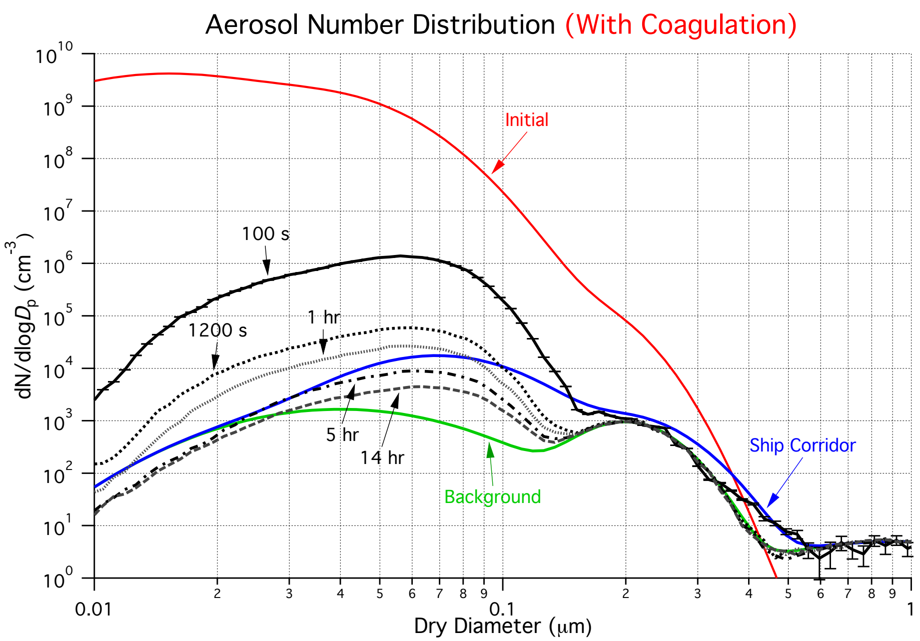
# Task B: Validate PartMC-MOSAIC for selected scenarios against experimental data from recent field campaigns

Comparison to size distribution from shipping corridor



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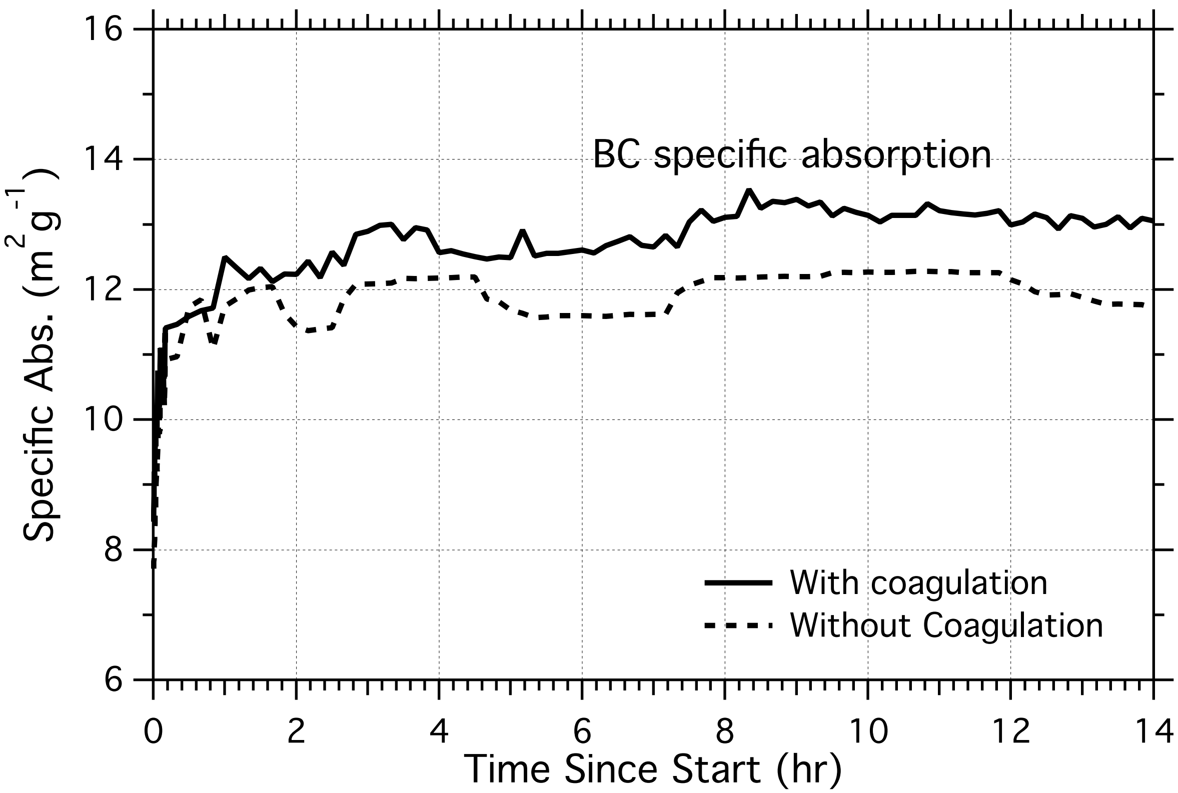
# Task B: Validate PartMC-MOSAIC for selected scenarios against experimental data from recent field campaigns

## Calculation of optical properties



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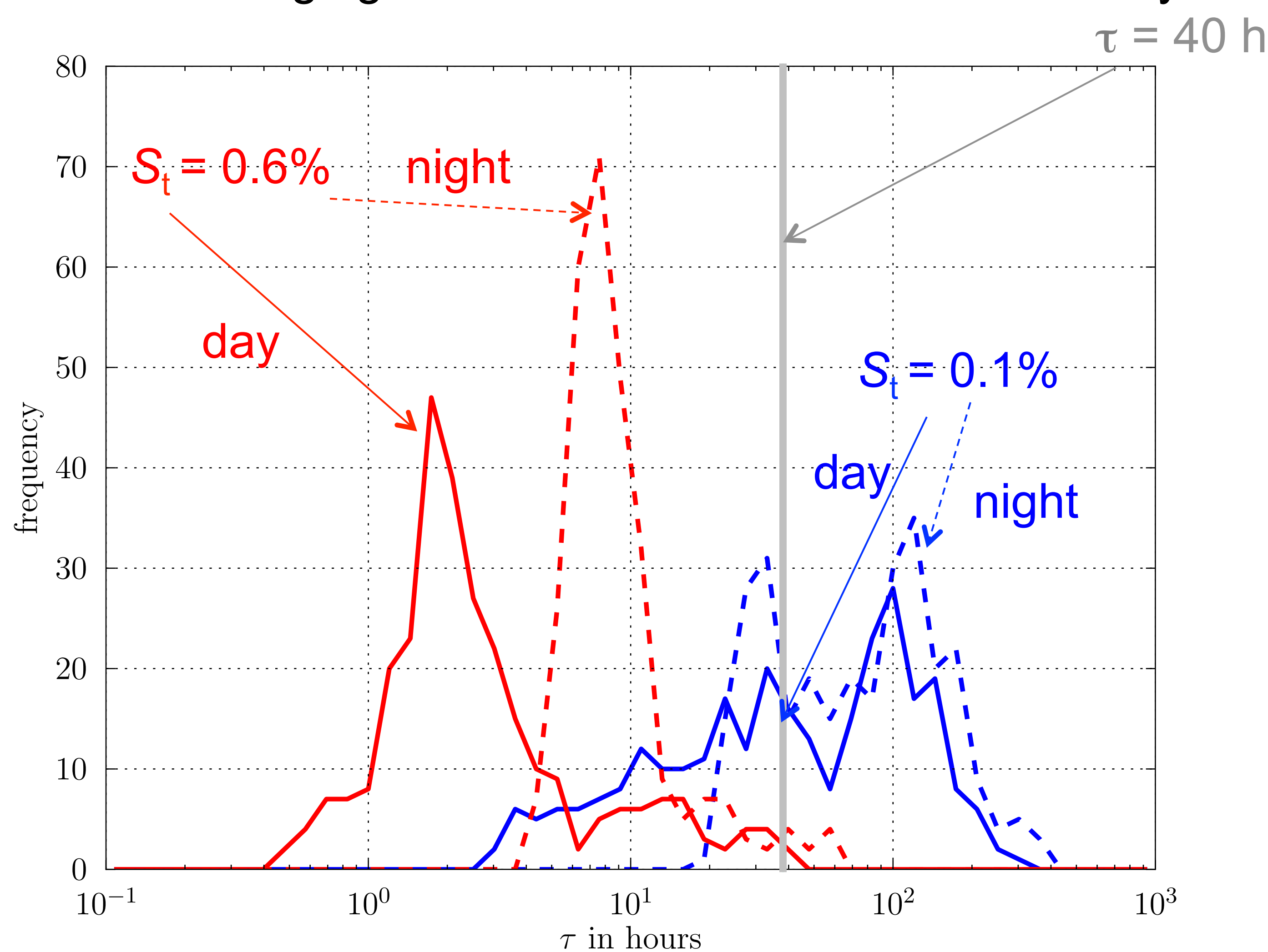
# Task C: Compute key quantities for black carbon impact

Distribution of aging timescales based on scenario library results



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# Ongoing work

- Task A:
  - Developing more tailored scenarios: Mumbai and New Delhi
- Task B:
  - Validating PartMC-MOSAIC with data from other field campaigns: CARES
- Task C:
  - Moving to online calculation of key quantities instead of post-processing
- Task D:
  - Developing sectional model version to enable error quantification
- Main challenge:
  - Data availability (per-particle data)

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# Outcomes

- Develop a suite of case studies, some of which validated with observations from recent field studies, that provide a high-detail picture of the processing of black carbon aerosol in the atmosphere in different environments.
- Calculate the evolution of optical properties and CCN properties, and deliver parameters for use in global model parameterizations.
- Provide validation of PartMC-MOSAIC.
- Estimate errors made by current aerosol models of different types due to simplified representations of black carbon mixing state.