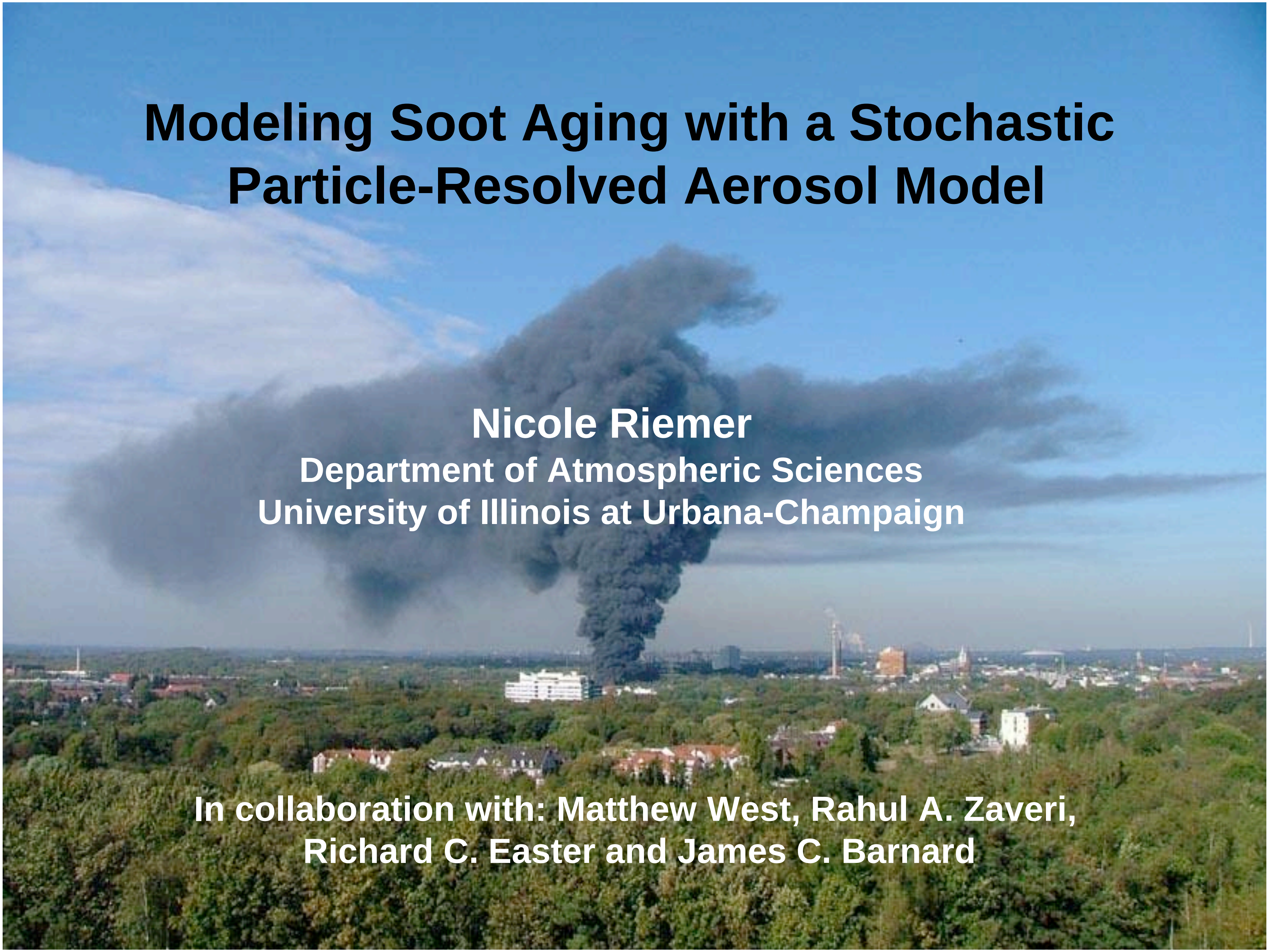
Modeling Soot Aging with a Stochastic Particle-Resolved Aerosol Model



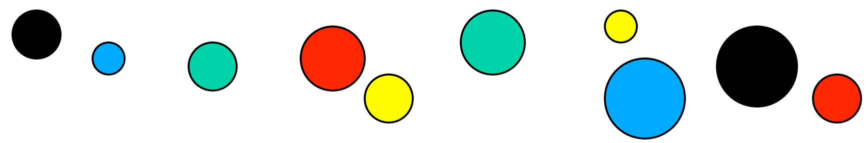
Nicole Riemer

Department of Atmospheric Sciences

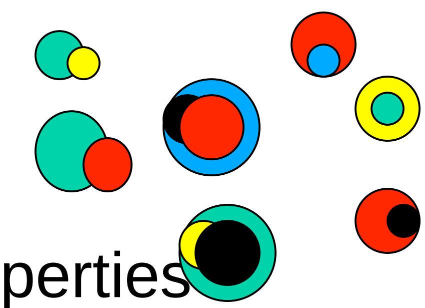
University of Illinois at Urbana-Champaign

In collaboration with: Matthew West, Rahul A. Zaveri, Richard C. Easter and James C. Barnard

Aging and the Aerosol Mixing State

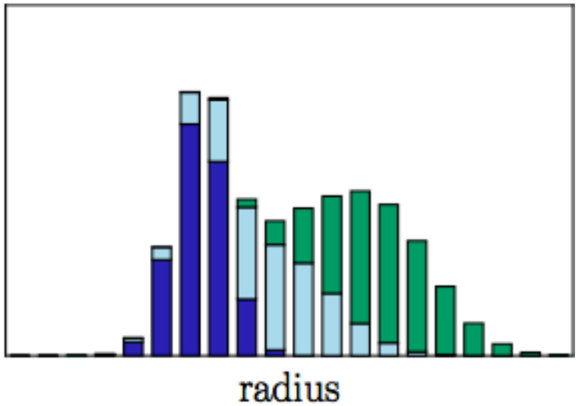


- Mixing states of primary aerosols change over time by coagulation and condensation (and photochemical processes)
- Understanding of aging process means understanding of mixing state evolution
- Aerosol mixing state affects
  - Optical properties
  - Cloud condensation nuclei activation properties
  - Ice nucleating properties



Treatment of Mixing State

- **Evolution of mixing state is presently poorly treated in aerosol models**
  - Each size bin is treated as internally mixed
  - This mixes “instantly” and unrealistically particles of the same size but different compositions
  - Rigorous multicomponent sectional approach is infeasible (e.g., 10 components and 30 fixed sizes → 30^{10} sections!!)
  - Arbitrary choice of particle types with ad hoc transferal rules



New Approach

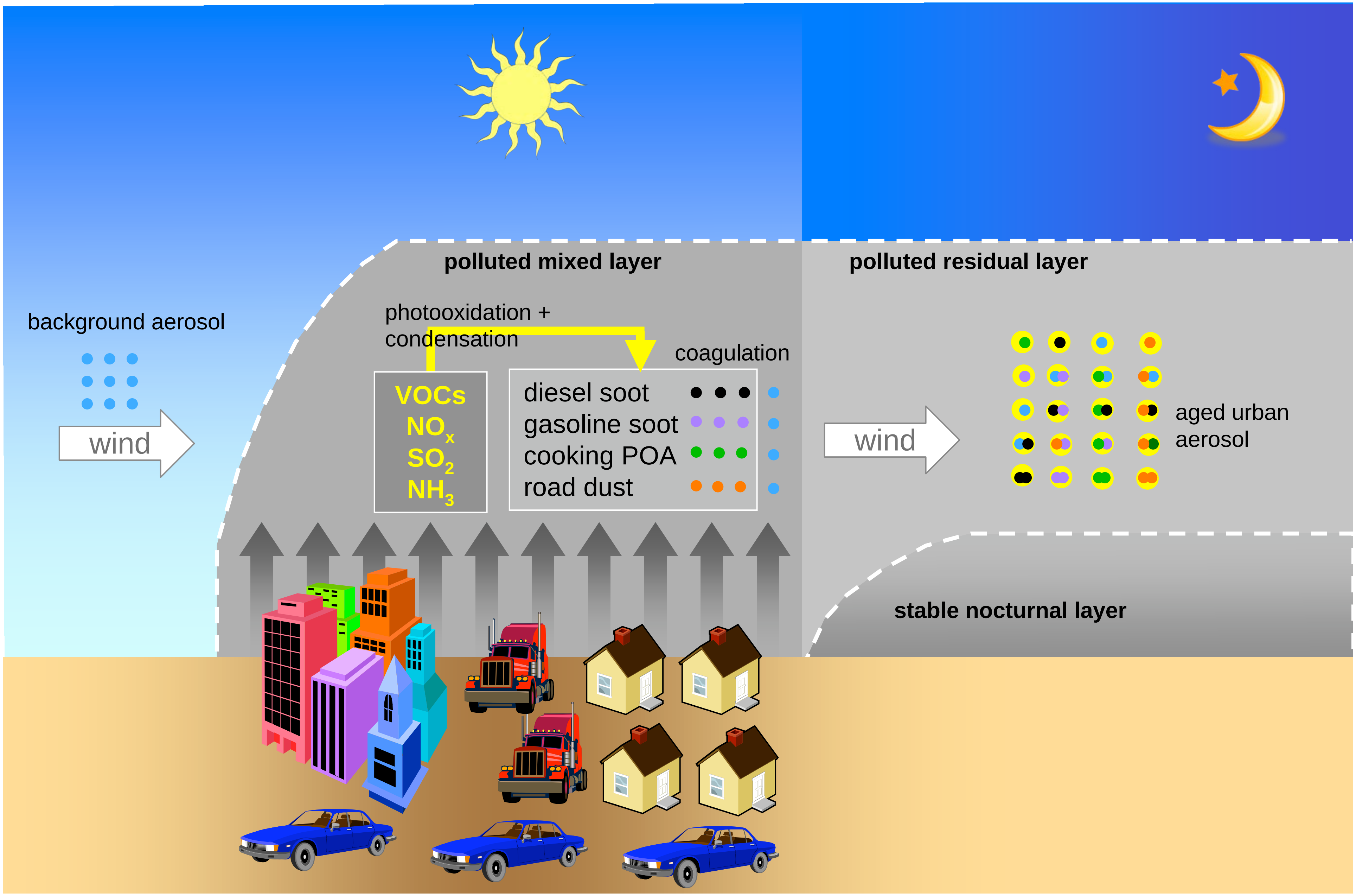
- Develop a unique particle-resolved aerosol model
  - Explicitly resolve individual particles in a complex aerosol within a small volume of air that represents a much larger well-mixed air mass of interest
  - Use deterministic numerical solutions for trace gas emission, dilution, photochemistry, aerosol thermodynamics, and gas-particle mass transfer
  - Use stochastic Monte Carlo solution for particle coagulation, emission, and dilution
- **Result: Accurately simulate evolution of aerosol number, size, mass, composition, and mixing state**
- **Two main applications:**
  - **Perform detailed simulations directly on the particle scale**
  - **Benchmark model for higher levels of the model hierarchy** (e.g. McGraw et al., submitted to Journal of Physics Conf. Proceedings 2008)

Aerosol Coagulation Module: PartMC
Aerosol and Gas Chemistry Module: MOSAIC

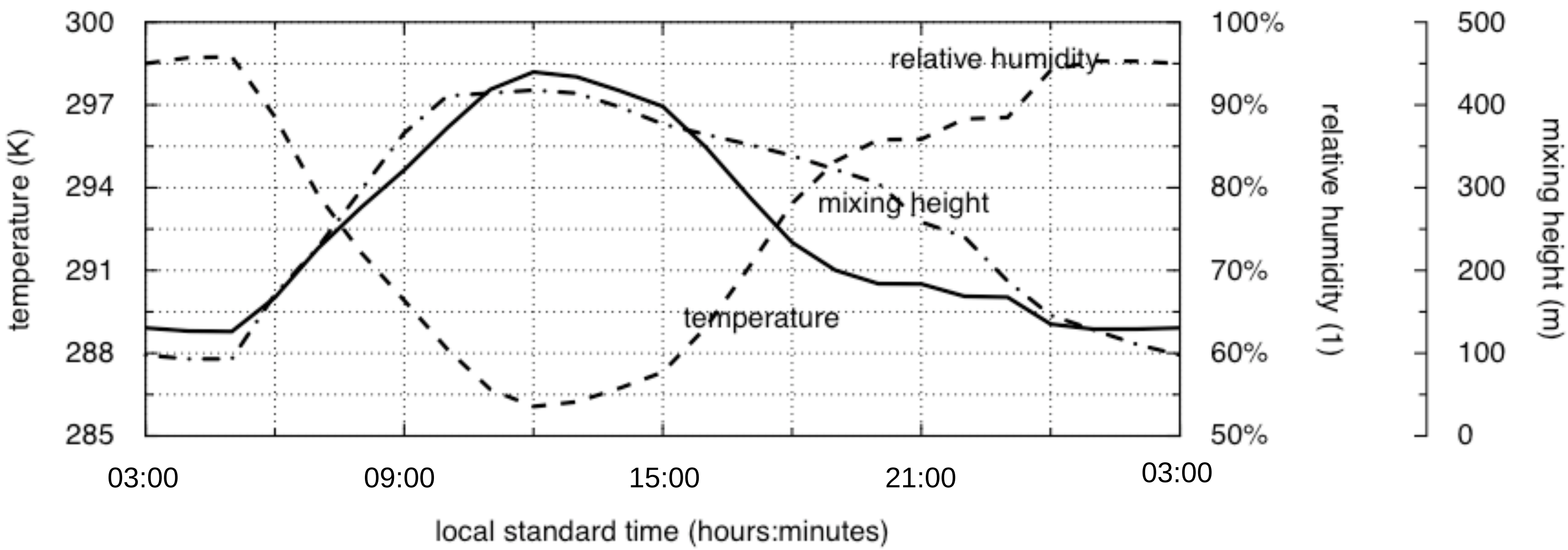
- **PartMC**
  - Directly simulates coagulation of individual particles
  - Avoids numerical diffusion present in sectional representations
  - Computationally efficient hierarchical accept-reject algorithm

- **MOSAIC** (Model for Simulating Aerosol Interactions and Chemistry)
  - Treats 20 species in the particle phase and 67 species in the gas phase
  - Accurate particle thermodynamics for inorganics (Zaveri et al., JGR, 2005a,b)
  - Dynamic gas-particle mass transfer (Zaveri et al., JGR, 2008)
  - Semi-empirical scheme for SOA (Schell et al., JGR, 2001)
  - CBM-Z gas-phase photochemical mechanism (Zaveri and Peters, JGR, 1999)
  - Robust and computationally efficient

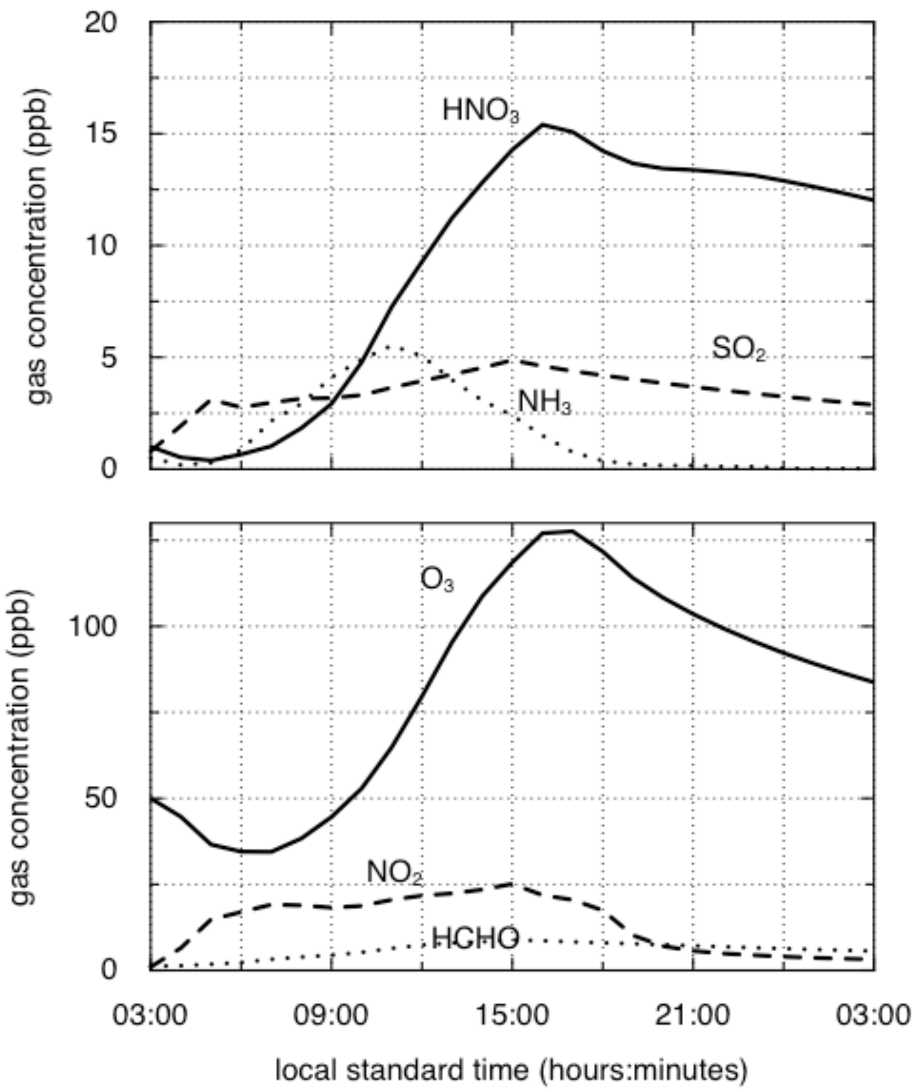
Idealized Urban Plume Scenario



Meteorological Parameters



Gas Phase



20
15
10
5
0
gas concentration (ppb)
HNO3
SO2
NH3
O3
NO2
HCHO
100
50
0
gas concentration (ppb)
03:00
09:00
15:00
21:00
03:00
local standard time (hours:minutes)

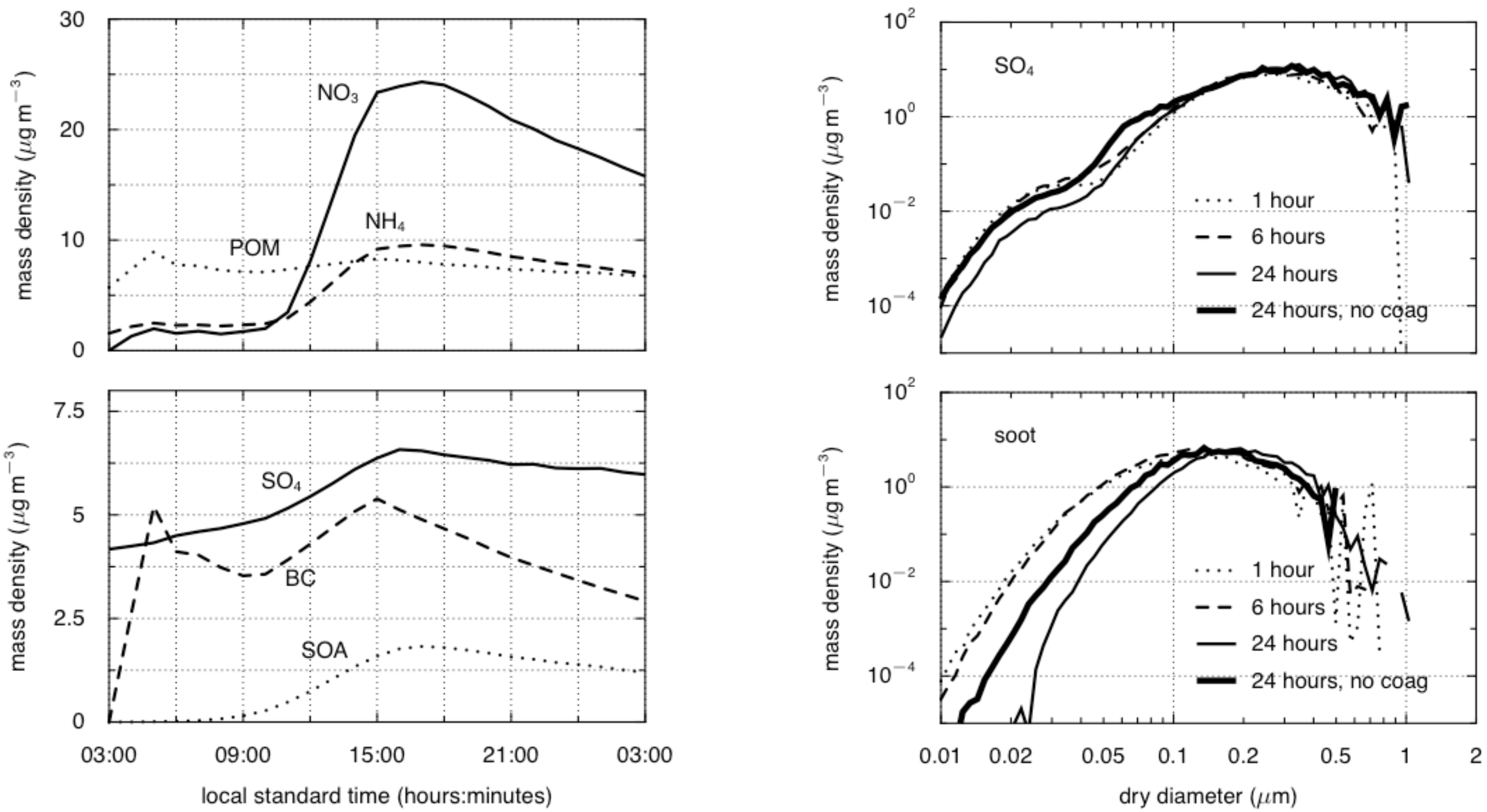
Initial and Emitted Aerosol Compositions

Initial & Bkg PM	*N* (cm^{-3})	r_{pg} (μm)	σ
50% $(NH_4)_2SO_4$ + 50% POM	3200	0.01	1.45
50% $(NH_4)_2SO_4$ + 50% POM	2900	0.058	1.65

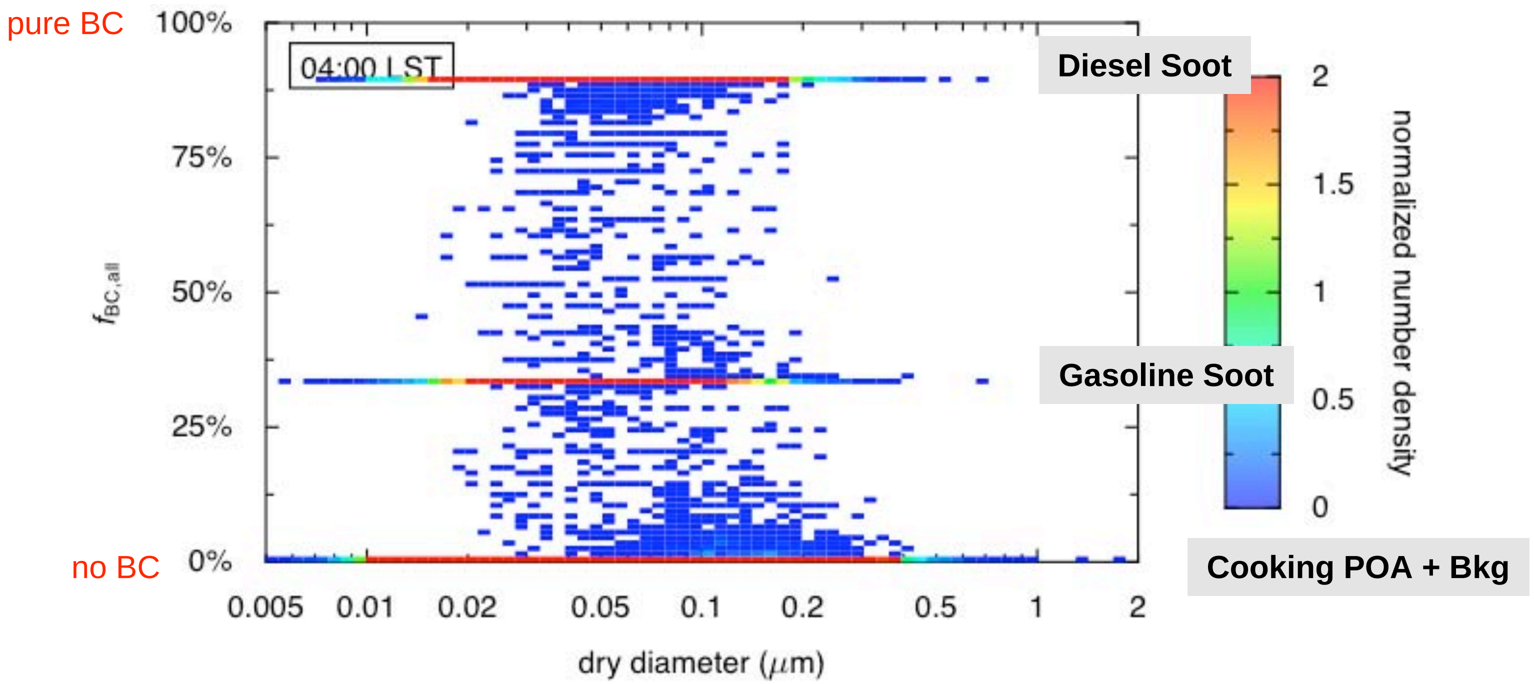
Emitted PM type	*E* ($kg\ day^{-1}\ km^{-2}$)	r_{pg} (μm)	σ
Cooking POA (100% POM)	2.2	0.043	1.91
Diesel soot (90% BC + 10% POM)	1.8	0.025	1.74
Gasoline soot (34% BC + 66% POM)	1.4	0.025	1.74

Emission rates typically found in a large urban area, based on Los Angeles 2006 data

Traditional Representation of Aerosol Concentrations



Evolution of Black Carbon Mixing State



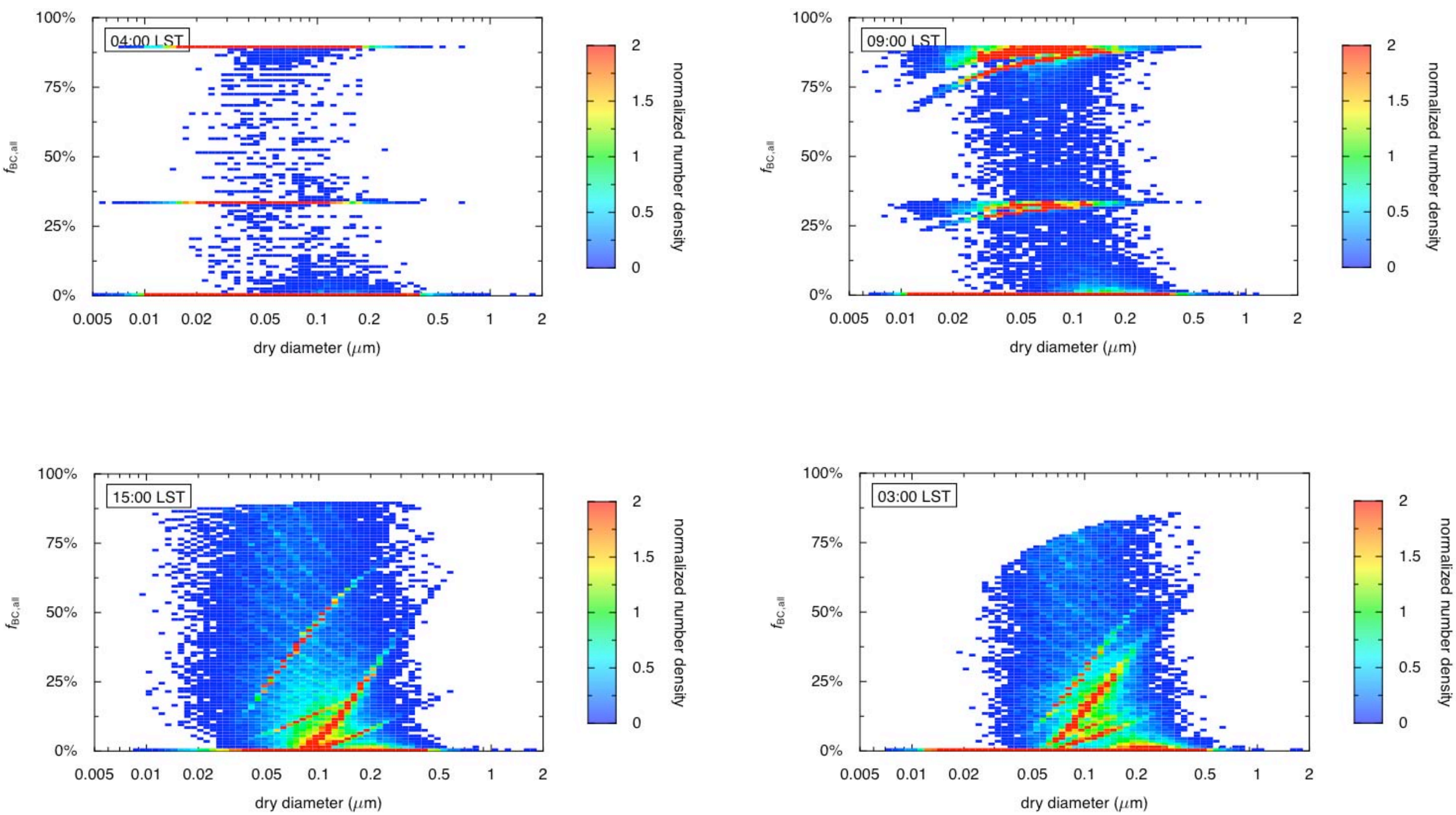
BC mass fraction:

$$f_{\mathrm{BC,all}} = \frac{\mu_{\mathrm{BC}}}{\mu_{\mathrm{all}}}$$

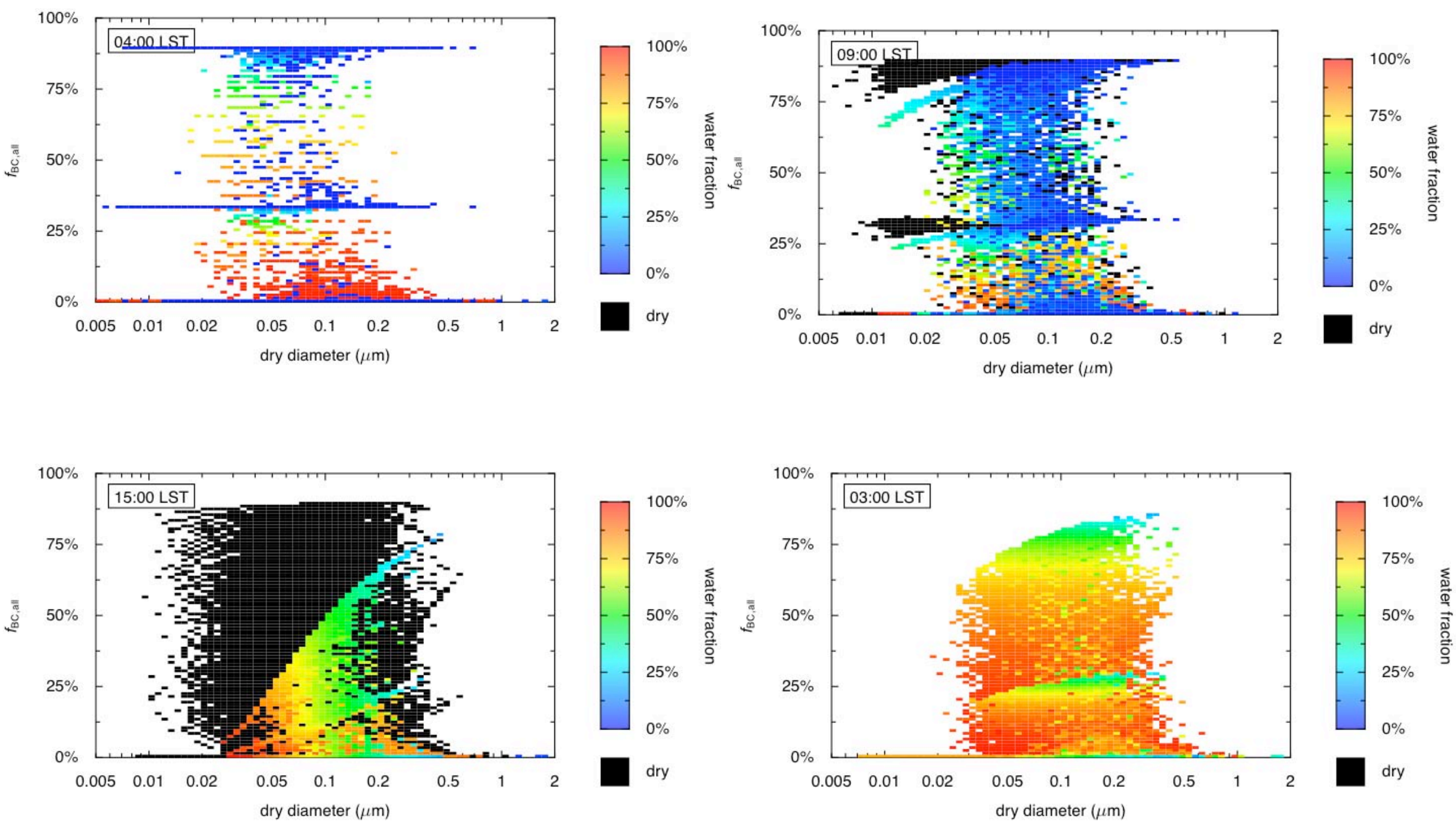
2D normalized number density distribution:

$$n(f, D) = \frac{1}{N_{\mathrm{tot}}} \frac{\partial^2 N(f, D)}{\partial f \, \partial \log_{10}(D)}$$

Evolution of Black Carbon Mixing State



Aerosol Water Content



Summary

- PartMC-MOSAIC: Unique particle-resolved aerosol chemistry and dynamics box-model
- Simulates the evolution of aerosol number, size, mass, composition, and mixing state
- Idealized urban plume scenario shows:
  - Same-size particles have very different mixing states.
  - Internally mixed sectional models do not resolve the composition-axis; instead just provide a one-dimensional size distribution.
  - Due to diurnal cycle of RH and temperature:
    - Co-existence of wet and dry particles
    - Important for ammonium nitrate formation
  - Coagulation shifts black carbon mass in diameter-composition space:
    - Mass of small particles ($D < 0.05$ um) with high BC content (above 80%) decreases by 89% due to coagulation;
    - Mass of large particles ($D > 0.1$ um) with low BC content (2%-10%) increases by 2300% due to coagulation