Challenges from the aerosol modeling community

Matt West and Nicole Riemer

University of Illinois at Urbana-Champaign

March 21, 2018

With apologies to Tim Onasch, Art Sedlacek, and Ernie Lewis

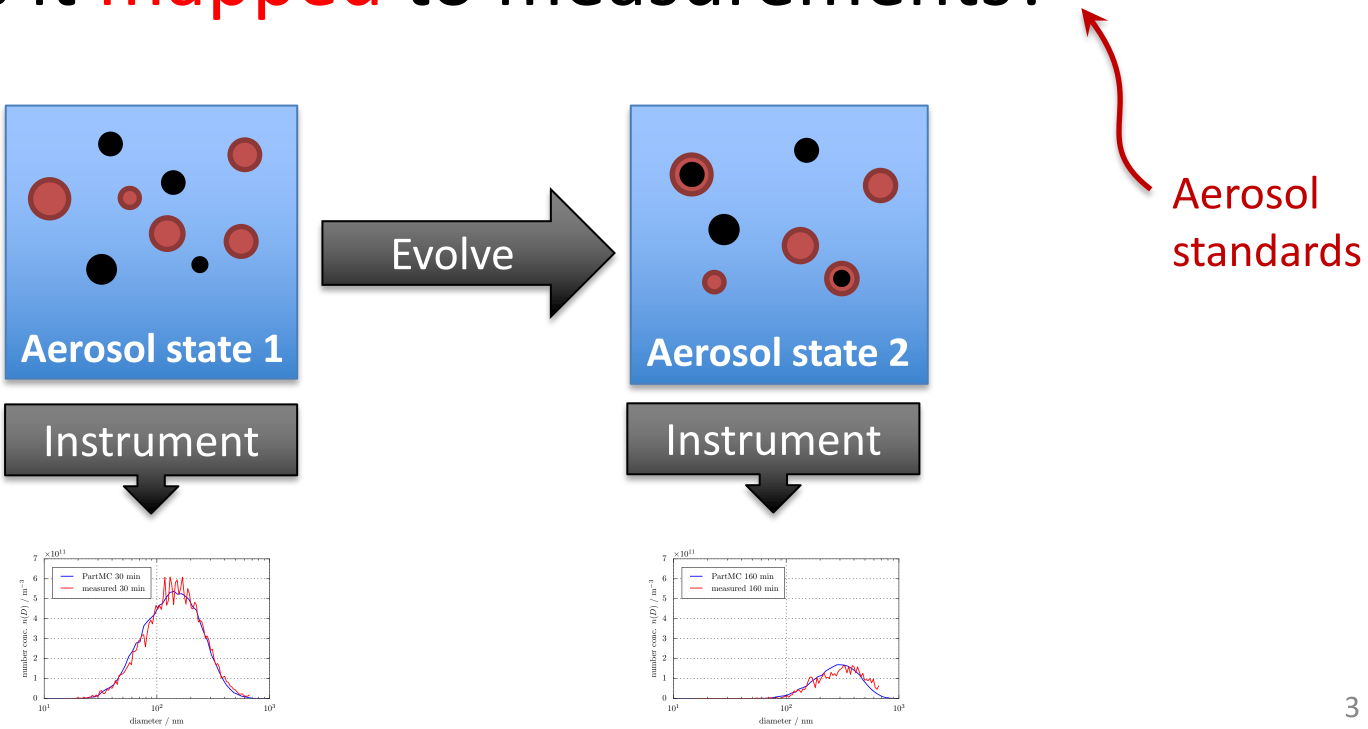
Process Readiness Level	Requirements for a process to be at a certain PRL
PRL 5:	**Extensive validation** performed against observational data, **uncertainty quantified**
PRL 4:	**Process implemented** in a regional or global model; Process-level verification
PRL 3:	**Quantitative process model:** Set of ODEs with known rate functions
PRL 2:	**Qualitative description of process:** When and where does it occur? What are the reactants and the products?
PRL 1:	**Phenomenon observed in the field**

Example of a process at a certain PRL

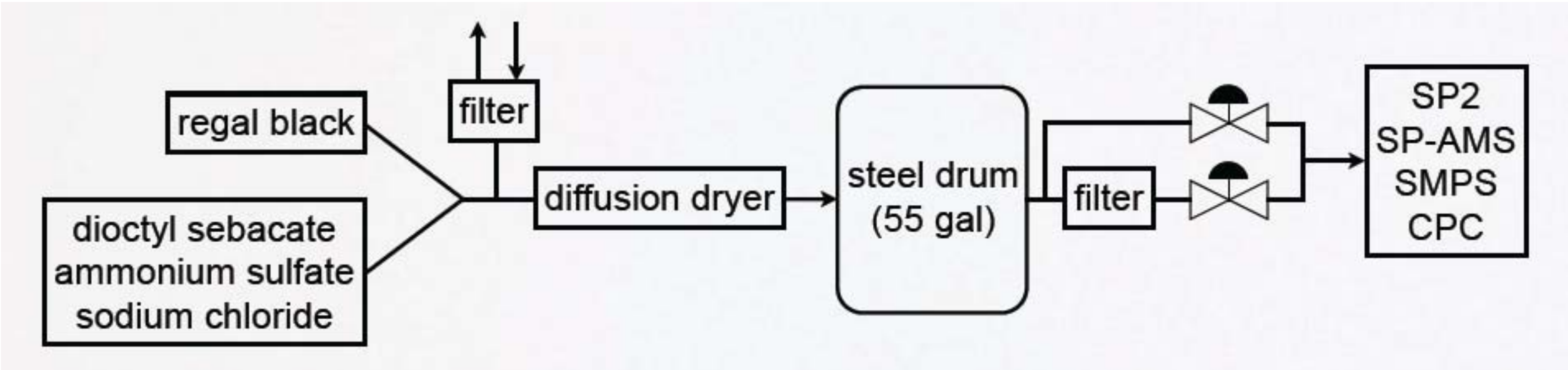
- inorganic gas
- CCN activation
- BC aging
- SOA formation
- Ice nucleation

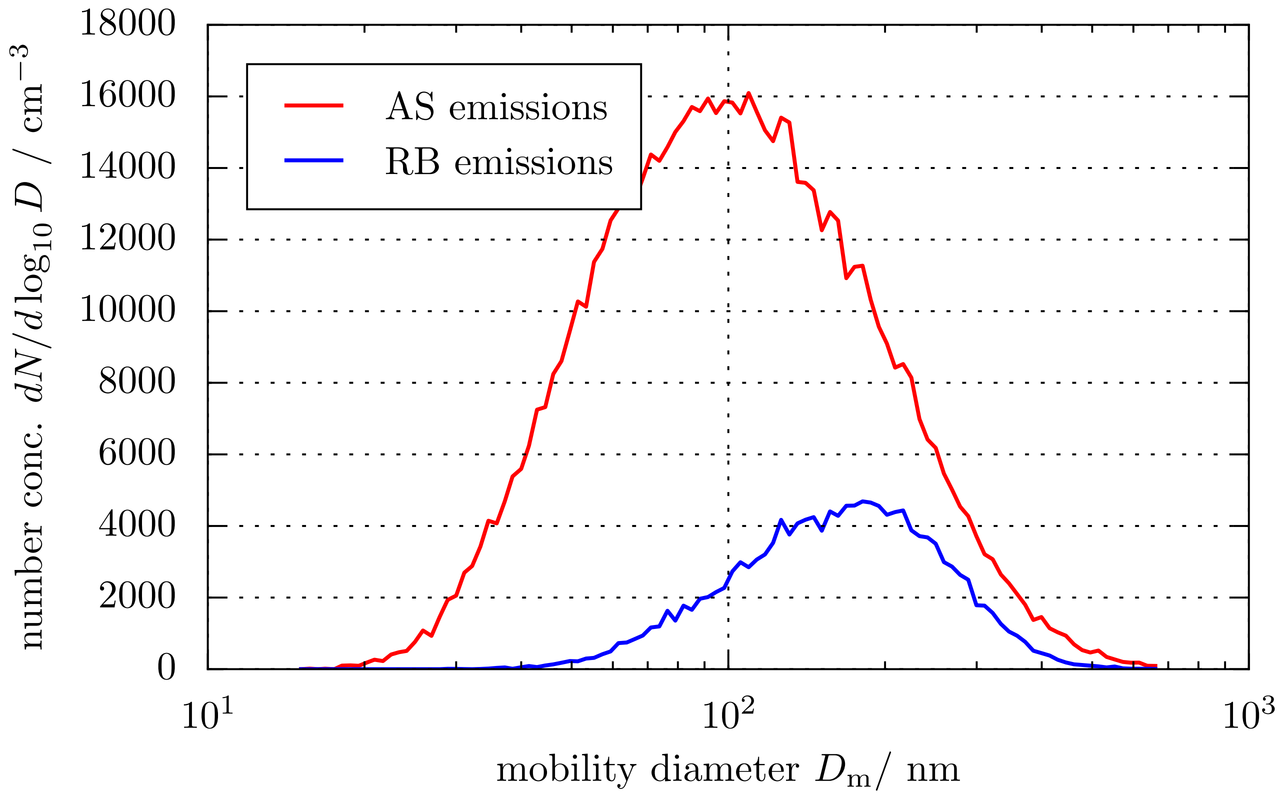
Model-measurement challenges

1. What is the aerosol state?
2. How does it evolve?
3. How is it mapped to measurements?

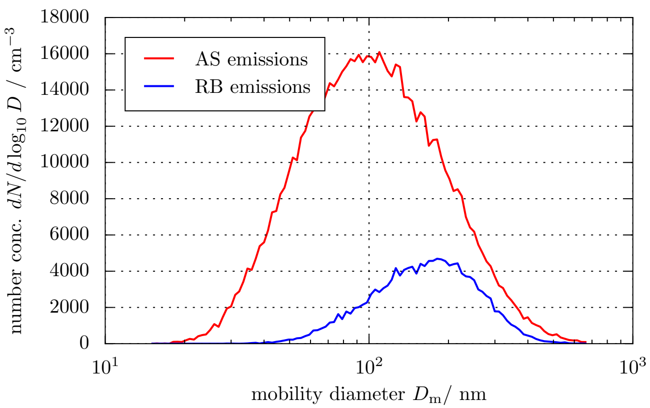


Dilution rate is about 2.6 L/min.

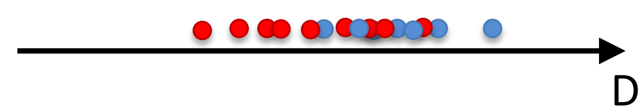




How does PartMC work?



Sample particle diameters



Particles are vectors:

[3, 0]
[7, 0]
[0, 2]
[0, 5]

AS RB

Evolve

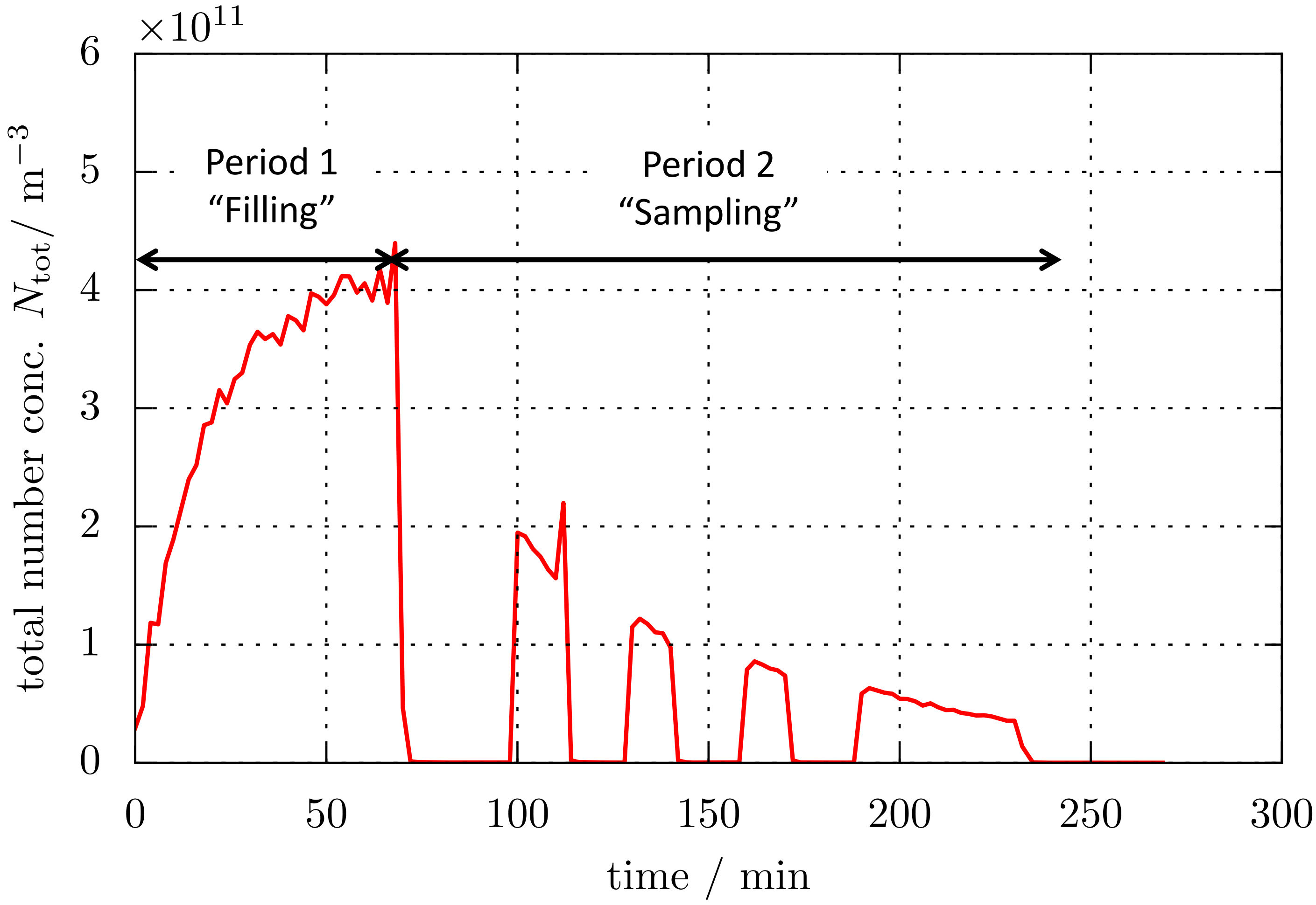
[3, 0]
[7, 2]
[0, 5]

AS RB

1. What is the aerosol state?

- Per-particle vectors
  - particle = [m_{BC}, m_{SO4}, m_{H20}, …, D_{core}, d_f, …]
    - Mass of each species
    - But what is a “species”? Organics?
    - Also morphology (core diameter, inclusions, fractal dim, charge, …)
- Even for non-particle-resolved models
  - Even when a model can’t resolve some details, measurements of these are still important
  - Important for later re-modeling or re-processing

Time series of measured total number concentration on 8/28



All the specifications that are needed for the model

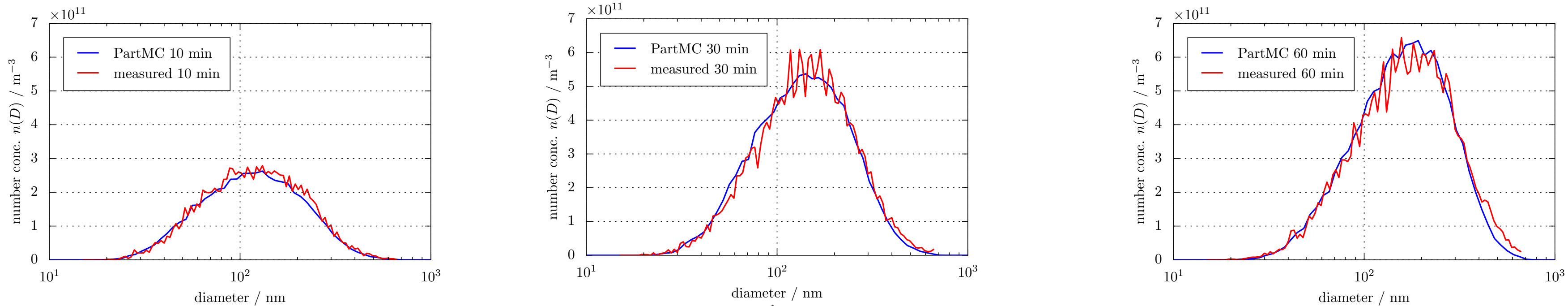
Quantity	Variable name	Value	Source
Barrel height	h_{B}	0.8954 m	Aerodyne
Barrel inner diameter	D_{B}	0.5715 m	Aerodyne
Barrel sedimentation area	A_{S}	0.2565 m^2	calculated
Barrel wall area	A_{D}	2.1206 m^2	calculated
Barrel volume	V_{B}	0.2297 m^3	calculated
Filling inflow for AS particles	R_{AS}	3 $\ell\,\mathrm{min}^{-1}$	Aerodyne
Filling inflow for RB particles	R_{RB}	3 $\ell\,\mathrm{min}^{-1}$	Aerodyne
Dilution outflow during Period 1	R_{dil_1}	6 $\ell\,\mathrm{min}^{-1}$	$R_{\mathrm{AS}} + R_{\mathrm{RB}}$
Dilution outflow during Period 2	R_{dil_2}	2.5 $\ell\,\mathrm{min}^{-1}$	Aerodyne
Relative humidity	RH	10%	Aerodyne
Temperature	T	293 K	Aerodyne
Pressure	p	10^5 Pa	Aerodyne
Fractal dimension	d_{f}	2.3	Tian et al. [3]
Wall loss parameter	k_{D}	0.06 m	Tian et al. [3]
Wall loss parameter	a	0.25	Theoretical, Bunz and Dlugi [1], Fuchs [2]
Radius of primary particles	R_0	10 nm	assumed
Volume filling factor	f	1.43	Tian et al. [3]
Total number conc.	N_{tot}	dynamic	
Number conc. of AS particles	N_{AS}	11,075 cm^{-1}	Eq. (1)
Number conc. of RB particles	N_{RB}	2,312 cm^{-1}	Eq. (1)
Filling rate for AS particles	λ_{AS}	2.177×10^{-4} s^{-1}	Eqn. (2)
Filling rate for RB particles	λ_{RB}	2.177×10^{-4} s^{-1}	Eqn. (2)
Dilution rate during Period 1	λ_{dil_1}	4.354×10^{-4} s^{-1}	Eqn. (2)
Dilution rate during Period 2	λ_{dil_2}	1.814×10^{-4} s^{-1}	Eqn. (2)
Wall loss	L_{wall}	dynamic	Eqn. (4) in Tian et al. [3]
Coagulation loss	L_{coag}	dynamic	Eqn. (1) Tian et al. [3]

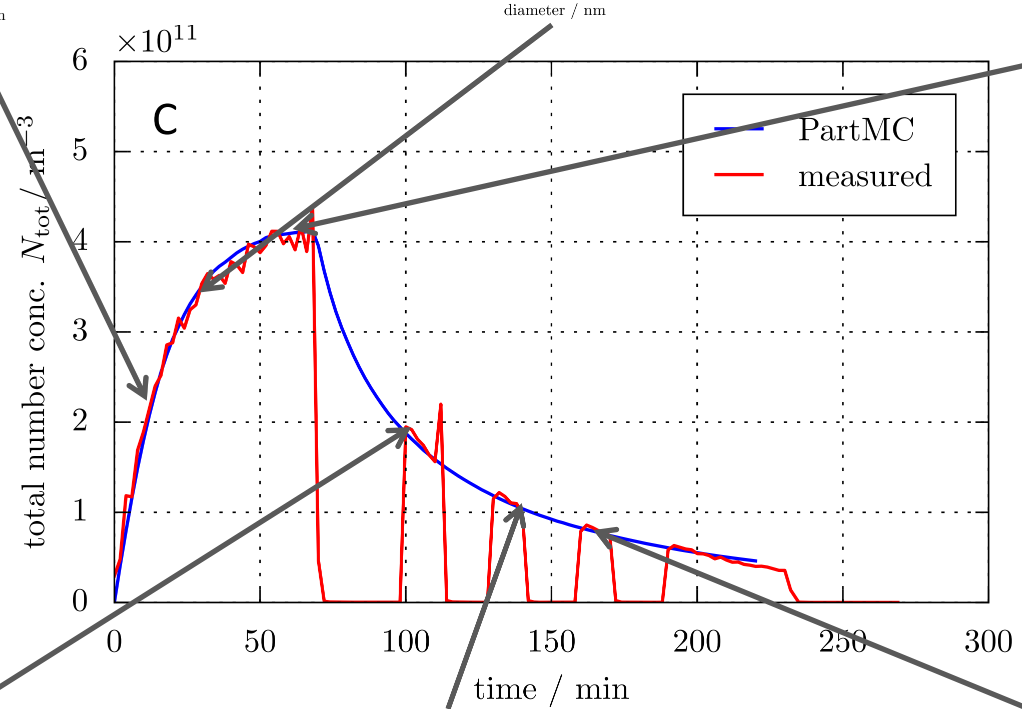
Fitted or guessed

Uncertain

2. How does the state evolve?

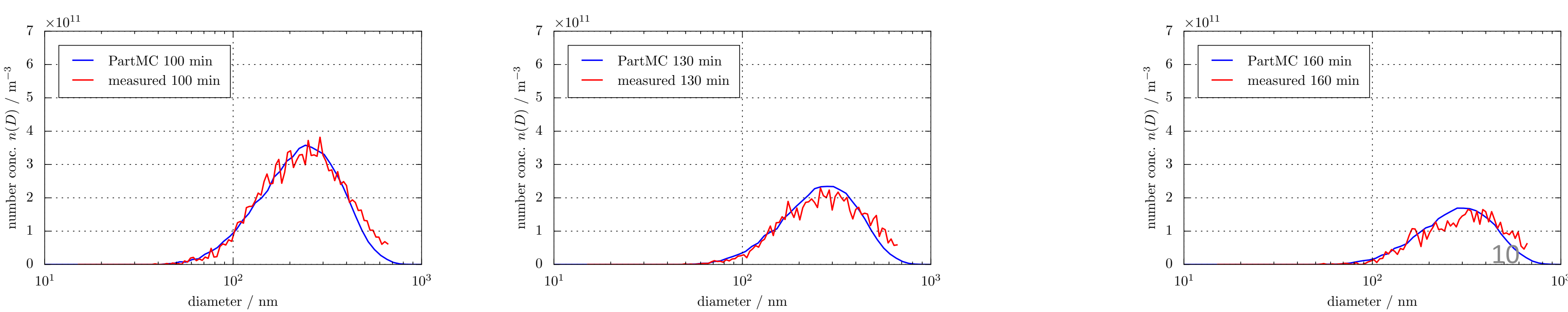
- Well-characterized inputs
  - Having to fit parameters is possible but painful
- All parameters along the way measured
  - Gas, environment, walls, fluxes
  - Unmeasured time-varying parameters are a nightmare (e.g., variable dilution rates)
- State measured periodically

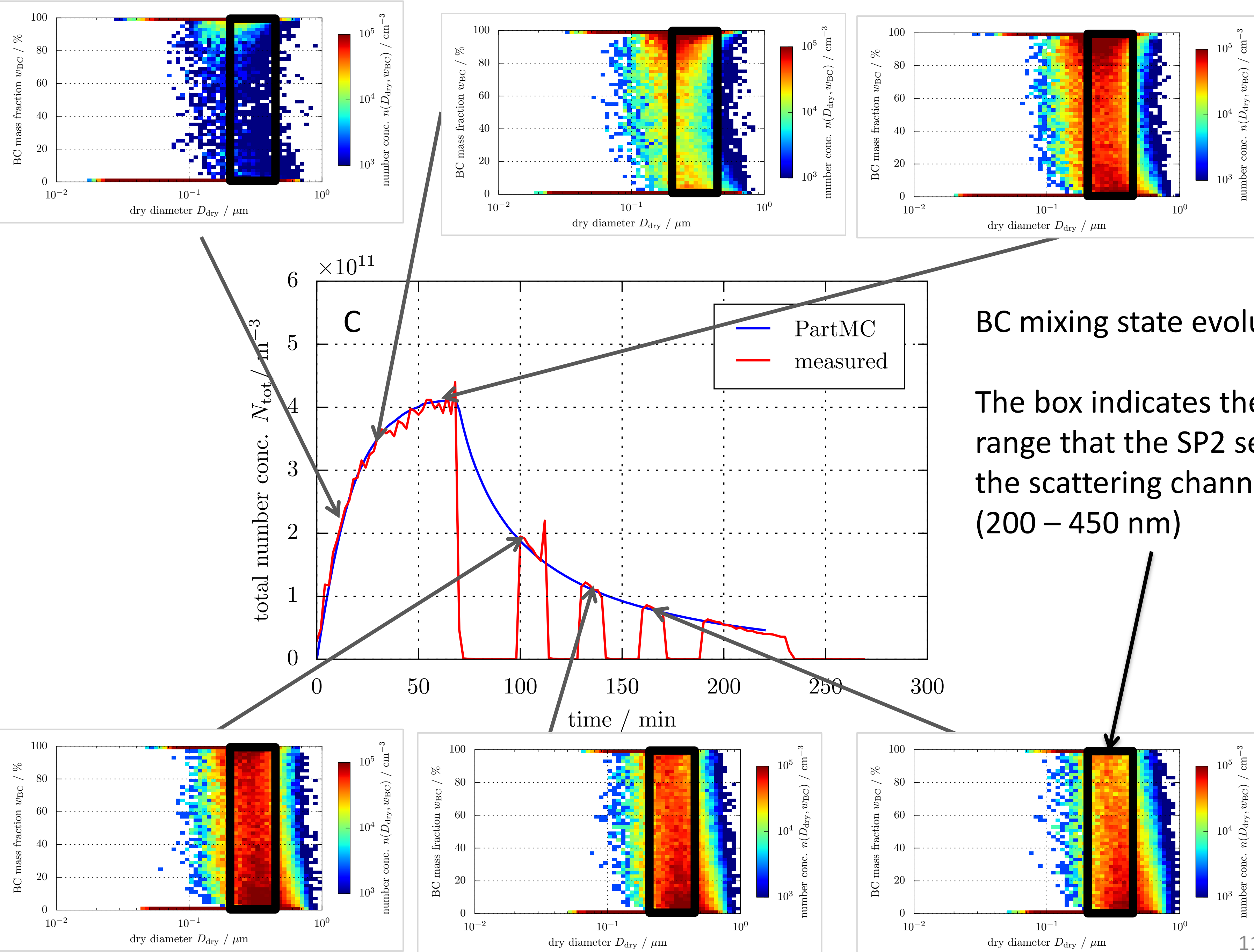




Comparison of size distributions

PartMC underpredicts the right side of the distribution somewhat, but overall this is not bad.





BC mixing state evolution

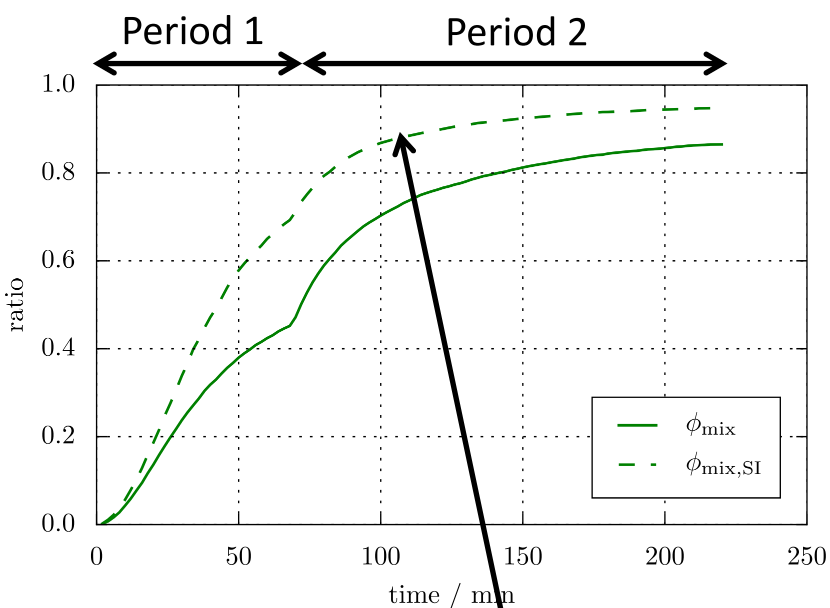
The box indicates the range that the SP2 sees in the scattering channel (200 – 450 nm)

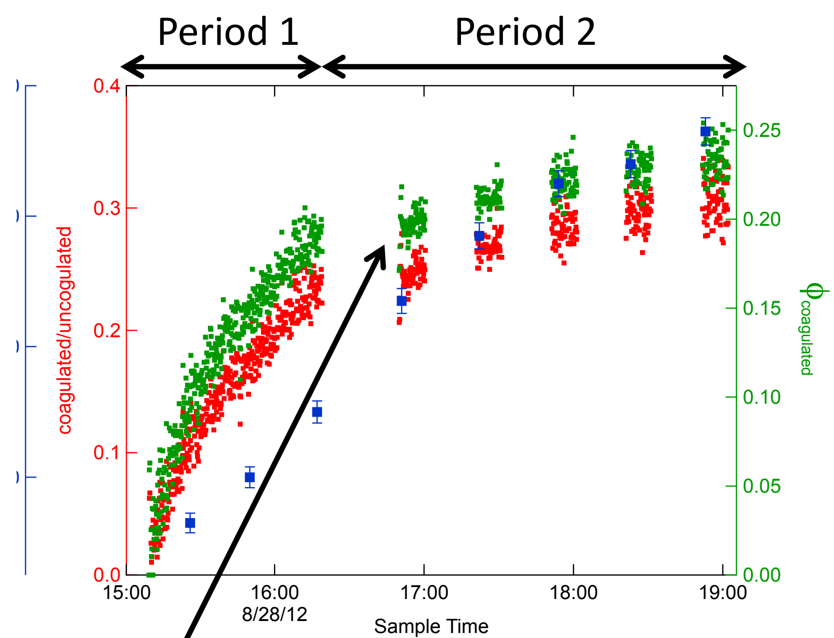
$$\phi_{\text{mix}} = \frac{N_{\text{mix}}}{N_{\text{BC}}+N_{\text{mix}}}$$

Fraction of mixed particles

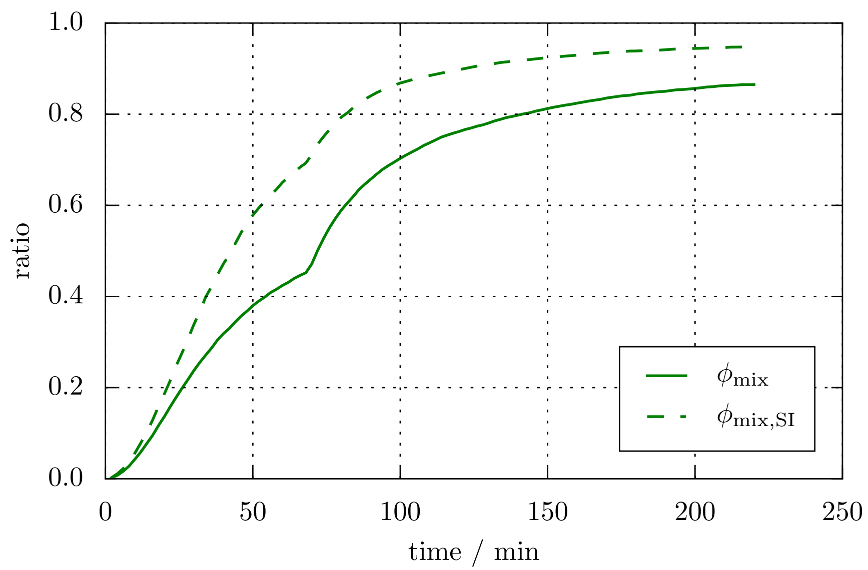
$$\phi_{\text{mix,SI}} = \frac{N_{\text{mix,SI}}}{N_{\text{BC,SI}}+N_{\text{mix,SI}}}$$

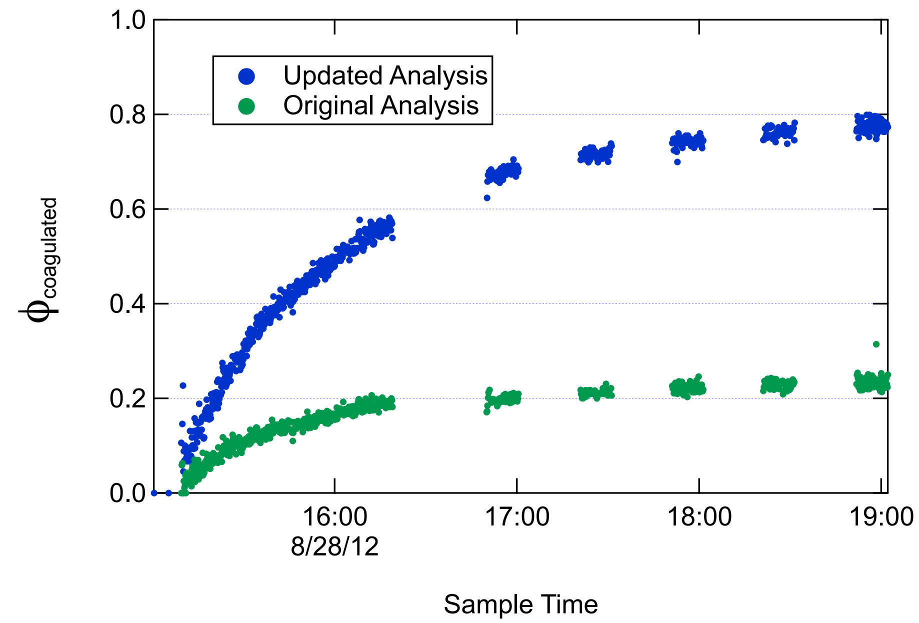
Fraction of mixed particles in the size range $200 \text{ nm} < D_{\text{opt}} < 450 \text{ nm}$





The green dashed line is supposed to be comparable with the green dots.

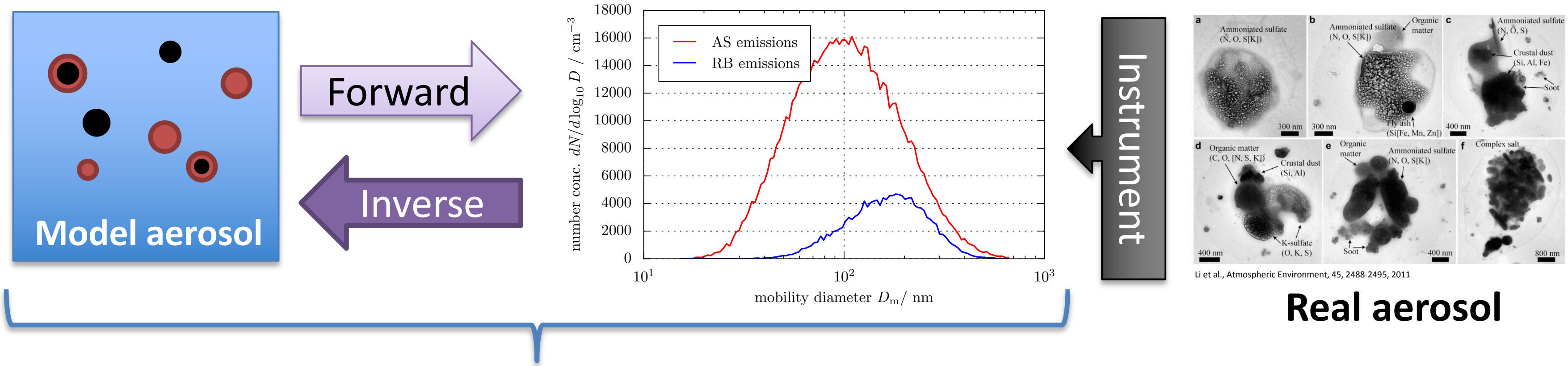




- Updated analysis: uncoagulated lag times -0.4 µs to +0.4 µs
- Original analysis: uncoagulated lag times -0.4 µs to +1.6 µs

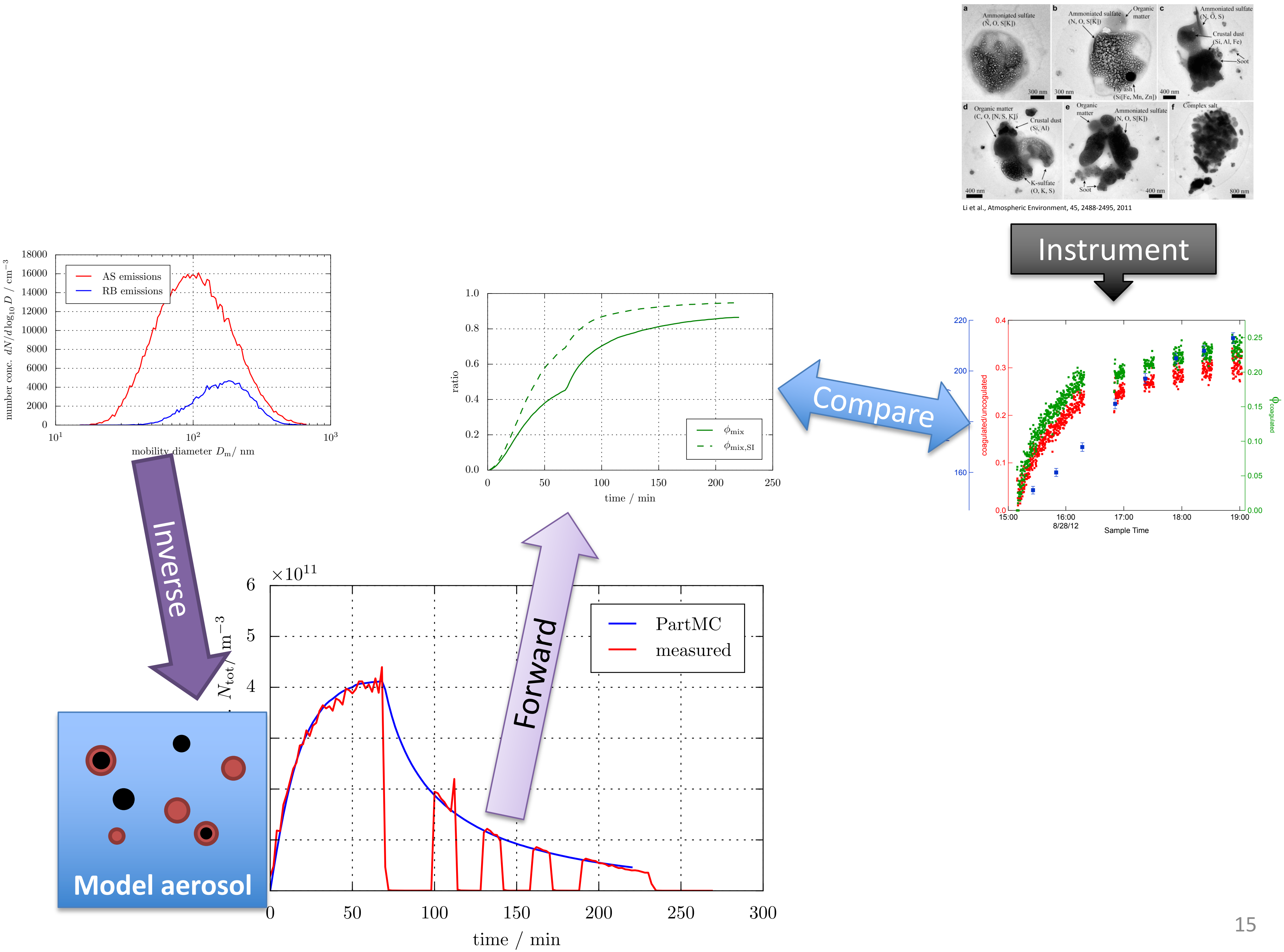
- More data in the SP2 signal (bimodal scattering peaks) could better resolve this

3. How does aerosol state map to measurements?



This is what we have to work with

- Inverse: measurement $\Longrightarrow$ state
  - Needed for initial condition
  - Key question: Can we recover a list of particle vectors from the measurements?
- Forward: state $\Longrightarrow$ measurement



Li et al., Atmospheric Environment, 45, 2488-2495, 2011
Instrument
Compare
Inverse
Forward
Model aerosol
AS emissions
RB emissions
number conc. $dN/d\log_{10} D$ / cm^{-3}
mobility diameter D_m/ nm
ratio
time / min
ϕ_{mix}
$\phi_{mix,SI}$
PartMC
measured
N_{tot}/ m^{-3}
$\times 10^{11}$
time / min
coagulated/uncoagulated
Sample Time
8/28/12

Why aerosol standards?

- Solve the mapping problems
  - Inverse: we measured *y*, what is really there?
  - Forward: we have *x*, what should we measure?
- Well understood mappings:
  - Mobility diameter ⟺ mass-equiv diameter
- Poorly understood mappings:
  - SP2 lag times
  - Single particle mass specs ("qualitative")

Mapping to aerosol state

- How do we reconcile different instruments?
  - Important to get complete state
  - Given SP2, AMS, SPLAT in CARES — how do we initialize a model? What are the particles?
- We want full state: per-particle mass fractions
  - With error bars!

Mixing State FG: Connections

	Theory/ Metrics	PRM	SP2	Micros-copy	SP mass spectro-metry	Bulk measure-ments	Remote sensing	RM/ GCM
Theory/ Metrics		high	medium	medium	low	low	low	low
PRM	high		medium	medium	medium	high	low	low
SP2	medium	medium		medium	medium	high	low	low
Micros-copy	medium	medium	medium		medium	medium	low	low
SP mass spetro-metry	low	medium	medium	medium		medium	low	low
Bulk measure-ments	low	high	high	medium	medium		high	medium
Remote sensing	low	low	low	low	low	high		high
RM/ GCM	low	low	low	low	low	medium	high	