An Efficient Representation of Aerosol Mixing State for Atmospheric Models

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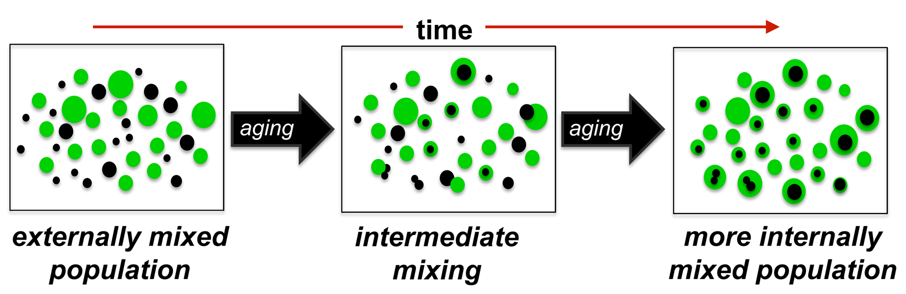
1. Introduction

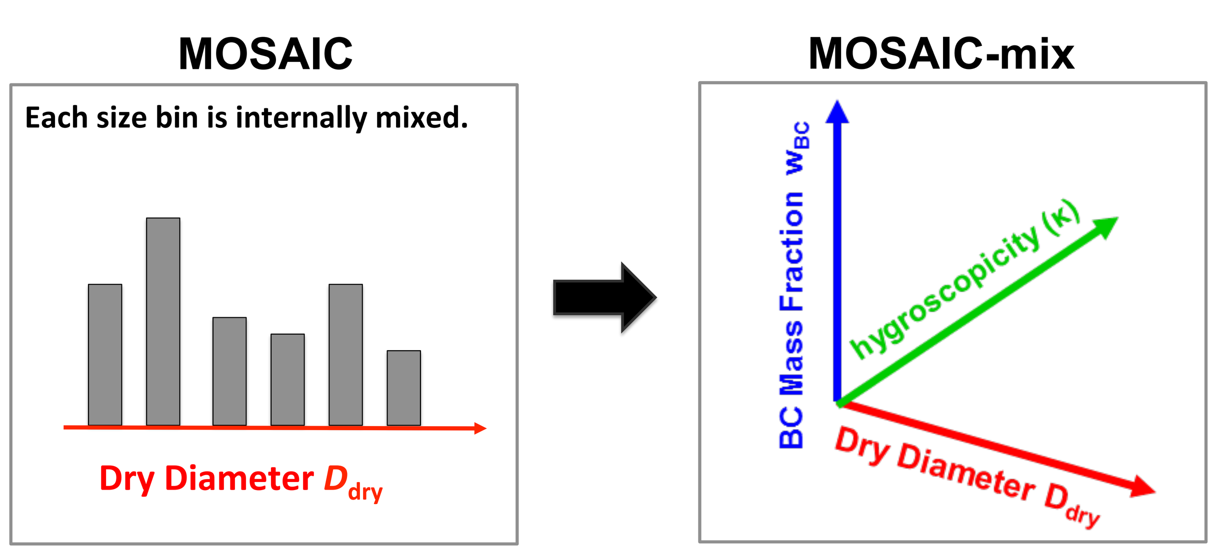
Atmospheric aerosol particles influence the Earth's radiative balance directly by absorbing and scattering the incoming solar radiation and indirectly by serving as cloud condensation nuclei (CCN). Aerosol optical and CCN properties strongly depend on particle size and mixing state (chemical composition), which evolve due to condensation and coagulation processes.



Here we describe and evaluate a novel sectional framework in the box-model version of the **Model for Simulating Aerosol Interactions and Chemistry**, referred to as **MOSAIC-mix**, that efficiently represents both size and mixing state of aerosols.

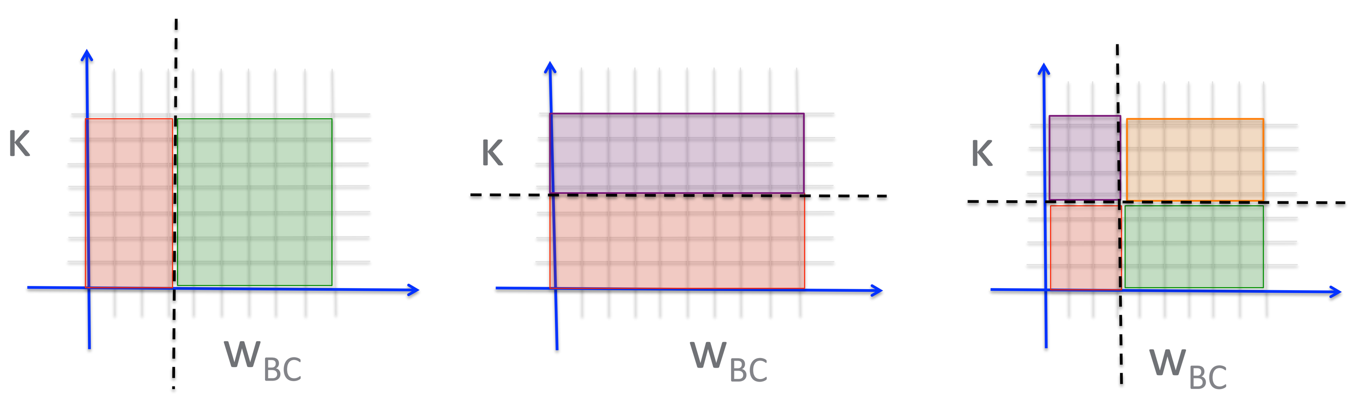
2. MOSAIC-mix Framework

- Light absorption by black carbon (BC) particles depends on their size and how thickly they are coated with non-refractory species such as ammonium, sulfate, nitrate, organics, and water.
- The CCN activation behavior of a particle depends on its dry size and the hygroscopicities of all the individual species mixed together.
- MOSAIC-mix represents the mixing state by resolving aerosol dry size (D_{dry}), BC dry mass fraction (w_{BC}), and overall particle hygroscopicity (k).

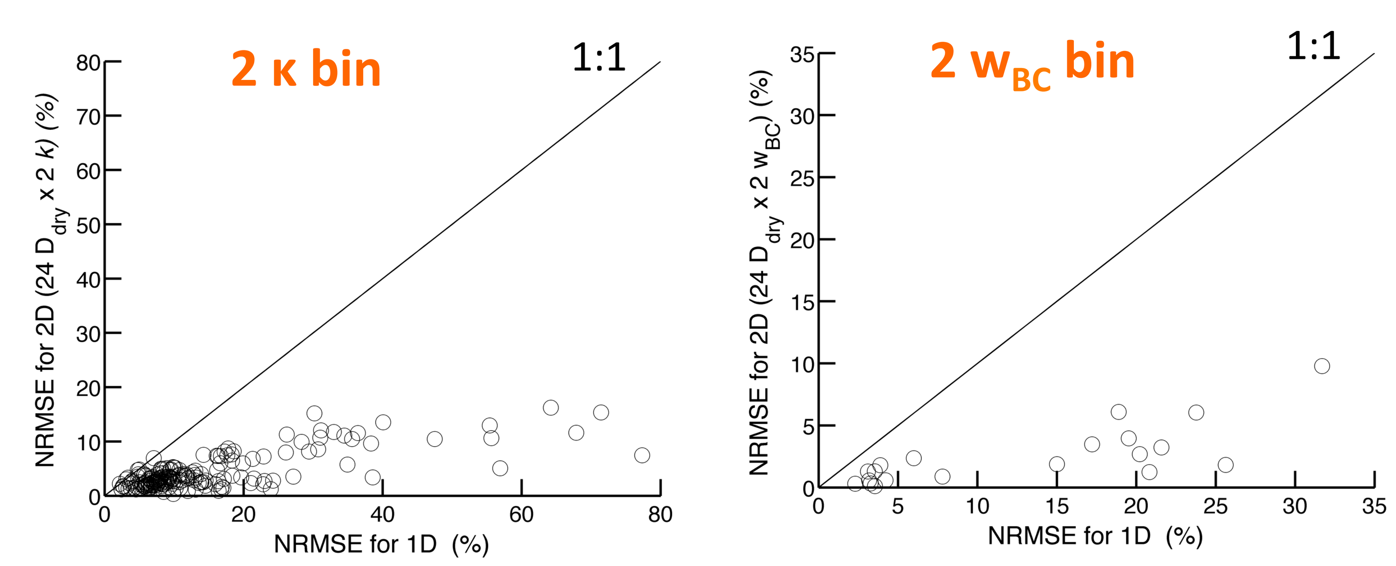


- A high-resolution MOSAIC-mix configuration with $24D_{dry}$ x 35 w_{BC} x 30 κ was used as a benchmark to determine a low-resolution configuration. High-resolution MOSAIC-mix agreed with particle-resolved PartMC-MOSAIC within 1.3% w.r.t. CCN concentrations and absorption coefficient.
- About 20,000 low-resolution MOSAIC-mix configurations in 1D, 2D, or 3D (24Ddry x 1-3 wBC x 1-3 κ) with different choices of bin boundaries were evaluated against the high-resolution configuration.

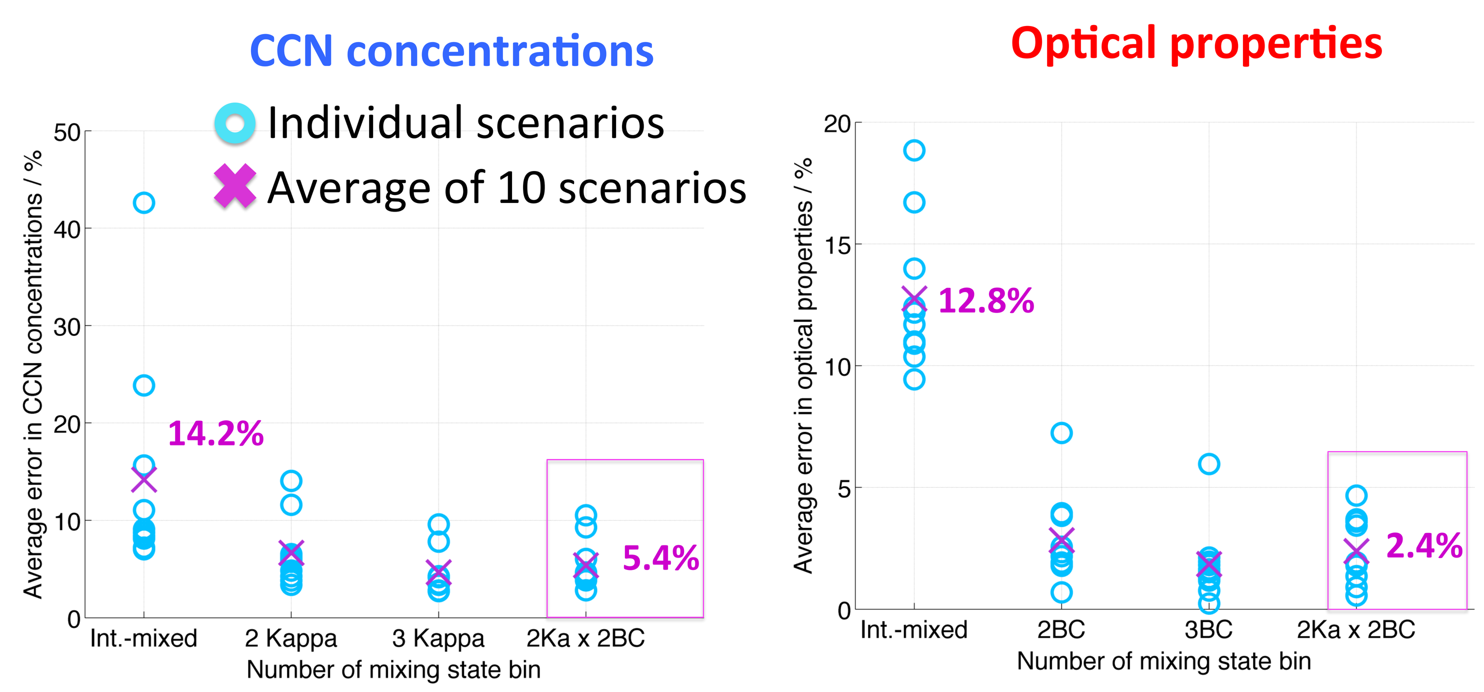
Some examples:



- Using 10 idealized urban plume scenarios in which different types of aerosols evolve over 24 h under a range of atmospherically relevant conditions, errors in CCN concentrations and optical properties were examined with respect to the level of detail of the aerosol mixing state representation.
- Errors in CCN concentrations and optical properties for mixing state-resolved simulations are smaller than those for internal-mixture-assumed simulations.



- Small error reductions per added bin after 2 or 3 bins. However, **resolving both w_{BC} and κ (with 2 w_{BC} x 2 κ) led to smaller total error (CCN + optical)** than resolving *either* w_{BC} or κ.



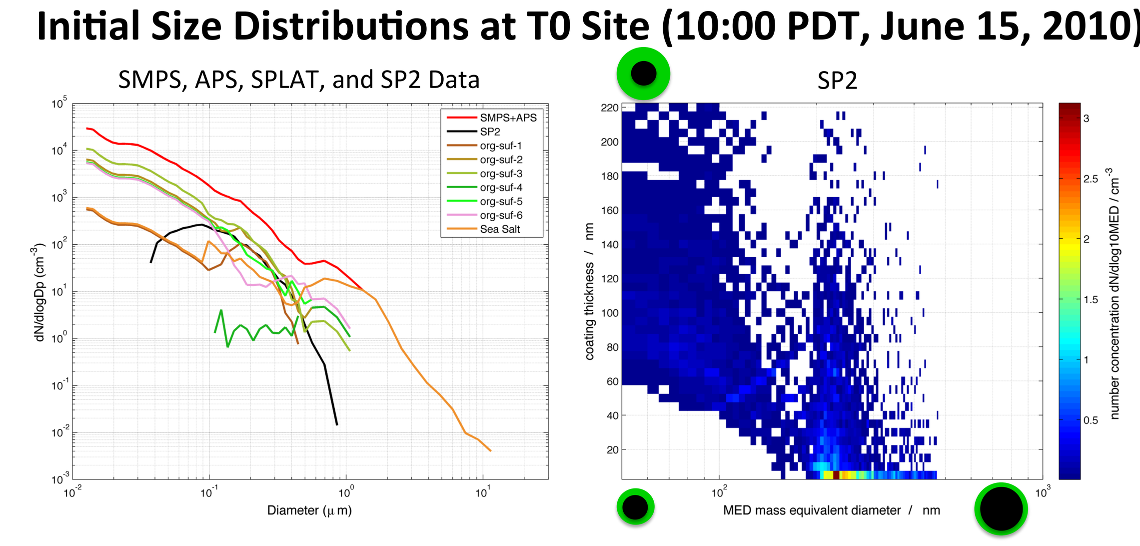
Overall Performance (Ching et al., 2016)

Number of mixing state bin	Optimization criteria	Optimum kappa	Optimum BC fraction	Average error in CCN (%)	Average error in Optical (%)	(Error in CCN + Optical) (%)	Error in B_{abs} (%)	
Internal mixture-assumed simulation	-	-	-	14.2	12.8	27.0	21.4	Worst
2(kappa) x 1(BC)	CCN conc.	0.1995	-	6.7	7.3	14.0	12.3	
3(kappa) x 1(BC)	CCN conc.	(0.0631, 0.1995)	-	4.7	6.9	11.6	11.6	
1(kappa) x 2(BC)	Optical properties	-	0.26	9.7	2.8	12.6	4.3	
1(kappa) x 3(BC)	Optical properties	-	(0.1, 0.36)	7.0	1.9	8.9	2.8	
2(kappa) x 2(BC)	Equal weighting	0.1	0.30	5.4	2.4	7.8	3.7	Best

3. Application to CARES Field Campaign

- The model was applied to simulate BC aging observed during the 2010 CARES field campaign (Zaveri et al., 2012).
- Five forward trajectories originating from T0 site on June 15 were selected based on FLEXPART-WRF simulations (Fast et al., 2012).
- PartMC-MOSAIC and MOSAIC-mix were initialized using single-particle observations of SP2 and SPLAT along with trace gas data at the T0 site.
- Predicted BC mixing state results were evaluated using G-1 aircraft observations.

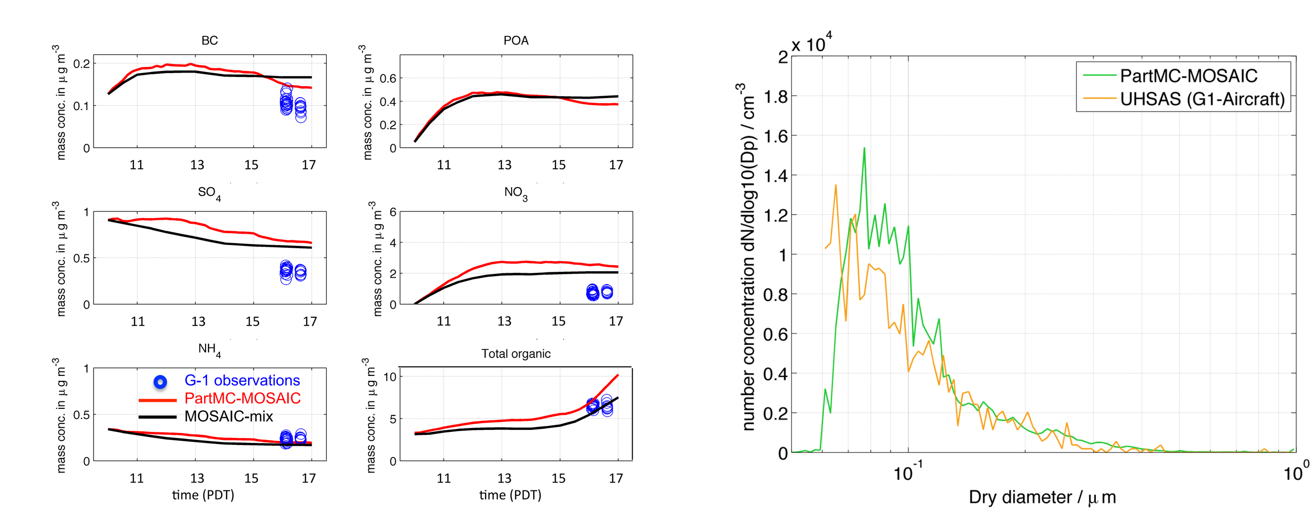
Initial Size Distributions at T0 Site (10:00 PDT, June 15, 2010)

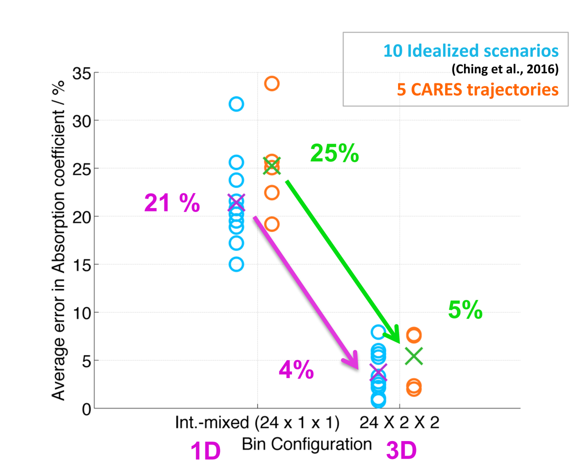


17 particle types of different compositions were used to initialize PartMC-MOSAIC.

MOSAIC-mix with 24 D_{dry} x 2 w_{BC} x 2 k was used to resolve these 17 particle types.

Model Evaluation Using G-1 Data (16:00 PDT, June 15, 2010)





Error in predicted absorption coefficient by MOSAIC-mix is calculated using PartMC-MOSAIC results as benchmark.

Internal mixture assumption overestimates absorption by BC by 15-35%. In contrast, the low-resolution MOSAIC-mix configuration (24 D_{dry} x 2 w_{BC} x 2 k) was able to reduce the average error to 4-5%.

4. Conclusions

- Internal mixture assumption in aerosol models leads to large errors in the predicted optical properties and CCN concentrations.
- The novel mixing state representation in MOSAIC-mix efficiently reduces these errors to sufficiently low values.
- MOSAIC-mix is being implemented in WRF-Chem to assess the impacts of aerosol mixing state on direct and indirect radiative forcing.

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