Final Report: DOE DE-SC0011771 — "Quantifying and Accounting for the Importance of Aerosol Mixing State"

Overview

The objective of this project was to develop a comprehensive framework of modeling and theory to rigorously investigate the impacts of aerosol mixing state on climate-relevant aerosol properties. The project led to several breakthroughs in the development of new methods as well as new science results. The significant accomplishments of this project are as follows:

1. We developed a conceptual framework to quantify mixing state based on rigorous information-theoretic entropy concepts, and used this to quantify climate-relevant aerosol impacts. **Our new *mixing state index* has seen significant adoption by experimentalists and modelers.** *Publications:* Ching et al. (2017, 2016a, 2018, 2016b); O'Brien et al. (2015).

2. We developed new numerical algorithms to enable the efficient simulation of particle-resolved aerosol microphysics within 3D atmospheric models, and implemented these in our coupled WRF-PartMC model. **This makes WRF-PartMC the world's most accurate model of aerosol microphysics coupled to atmospheric dynamics, and we performed the first ever 3D regional-scale simulations that fully resolve aerosol mixing state (see Figure 1).** *Publications:* Curtis et al. (2016, 2017); Maginnis et al. (2016).

3. **We integrated our modeling and conceptual framework with observations including ASR field campaigns (CARES, see Figure 1) and ASR-supported laboratory campaigns (Boston College BC-3).** *Publications:* Shou et al. (2019); Tian et al. (2017); Willis et al. (2016).

4. We extensively participated in ASR program activities, including **leading the Aerosol Mixing State Focus Group, co-chairing the Absorbing Aerosol Workshop, and co-chairing the Aerosol Processes Working Group.**

Significance: Tackling unknown unknowns

This project has been motivated and guided by the fact that the macroscale climate impacts of atmospheric aerosol fundamentally depend on the microscale properties of individual particles. Even though we have a good understanding of many aerosol processes on a process level, it is challenging to represent these in a global modeling framework, since global models by necessity require simplified aerosol representations due to computational constraints. Many global climate model studies have demonstrated that assumptions regarding aerosol microphysics significantly impact model results (Lee et al., 2011). While these studies are useful for quantifying sensitivities to "known unknowns" (i.e., within-framework assumptions), they cannot address "unknown unknowns" (i.e., structural uncertainties) of current aerosol models. Our highly detailed particle-resolved aerosol model enables us to close this gap.

Background: Why particle-resolved techniques?

Simulating all particles explicitly in a population resolves the mixing state rigorously and completely eliminates any errors associated with numerical diffusion. This makes PartMC-MOSAIC suitable for use as: (1) a numerical benchmark of mixing state for more approximate models; (2) an ideal platform for accurate process studies down to the particle scale; and (3) a rigorous foundation

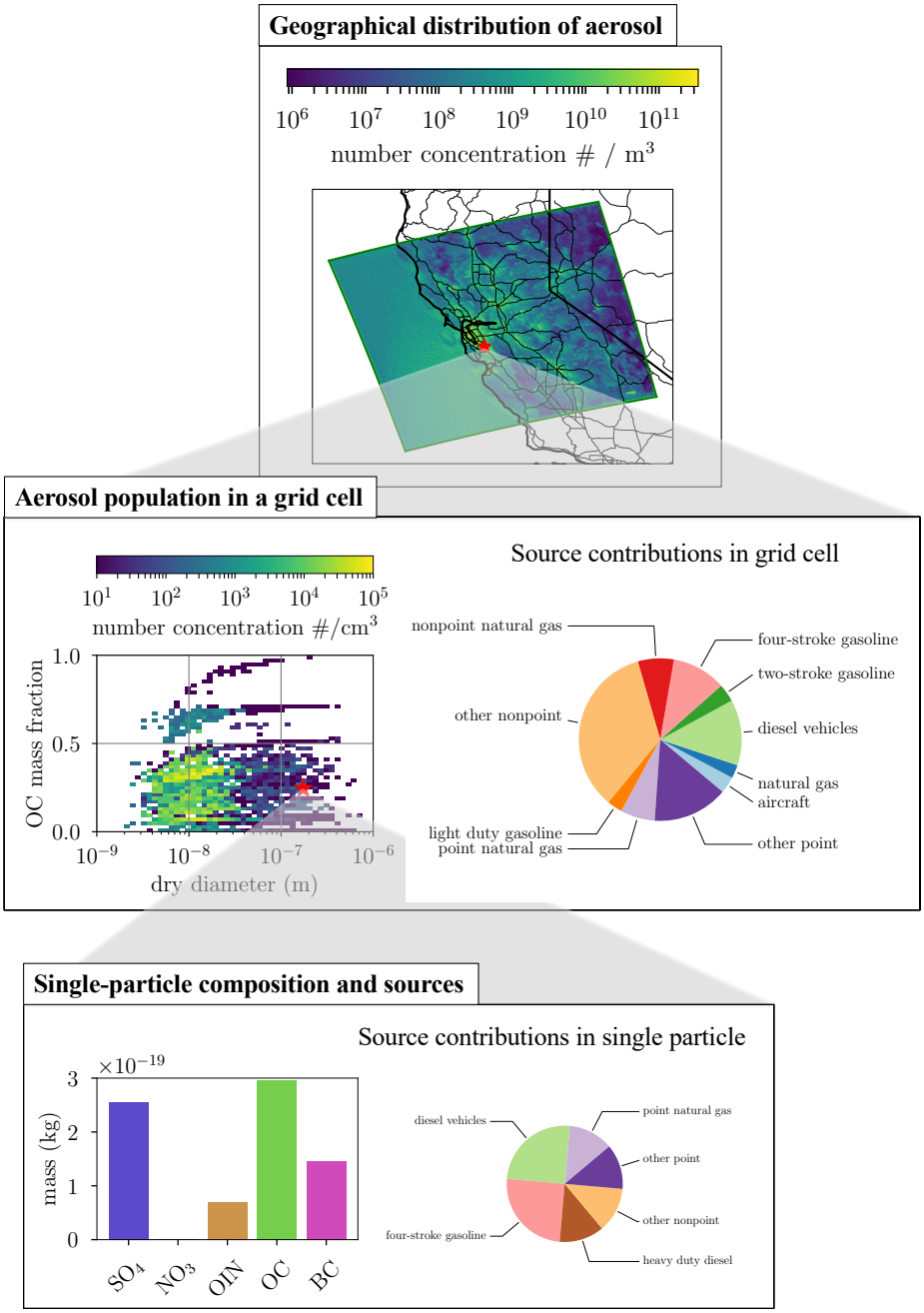

Figure 1: This figure illustrates the level of detail that can be obtained by WRF-PartMC for a simulation for California (the DOE CARES campaign domain). This is the first-ever regional simulation that is able to directly resolve aerosol mixing state on a particle level. In such a model run, we are simulating the complex aerosol dynamics and chemistry for about 5 billion individual particles. For any given particle in the simulation, we store its chemical composition and the sources of the constituent particles that it is composed of (particles aggregate because they coagulate with each other during transport). For each grid cell in the model domain, we can then construct the full mixing state of the aerosol population, as well as the full source apportionment profile.

for the development of coarse-scale parameterizations. The main challenge with particle-resolved methods is that they are computationally expensive. However, with our advances in algorithm development we were able to push the envelope of aerosol simulation. While we know from our previous studies that population mixing state matters for climate-relevant properties (i.e. CCN concentrations, optical properties) on the particle scale (Ching et al., 2012; Fierce et al., 2016, 2013; Zaveri et al., 2010), we have not yet had the appropriate tools for quantifying these effects on the regional to global scale. The research under Grant DE-SC0011771 has filled this gap as it has allowed us to develop the first 3D regional-scale particle-resolved aerosol model (WRF-PartMC).

Project outcomes

As part of this project, we have coupled our pre-existing particle-resolved aerosol box model (PartMC) to the community Weather and Research Forecast model (WRF). This new model capability (WRF-PartMC) not only resolves the particle mixing state on a per-particle level but also the spatial distribution of aerosol and trace gases of the atmosphere, fully coupled to meteorological processes. This involved substantial model development on stochastic algorithms and on preparing the aerosol emissions, going from mass-based aerosol emission fluxes as it is typical for traditional WRF-Chem simulations, to number-based source-oriented emissions, consistent with the PartMC framework. Figure 1 illustrates the level of detail that can be obtained by WRF-PartMC for a simulation for the Carbonaceous Aerosol and Radiative Effects Study (CARES) domain in California. In such a model run, we are simulating the complex aerosol dynamics and chemistry for about 5 billion individual particles. For any given particle in the simulation, we store its chemical composition and the sources of the constituent particles that it is composed of (particles aggregate because they coagulate with each other during transport). For each grid cell in the model domain, we can then construct the full mixing state of the aerosol population, as well as the full source apportionment profile.

Our new *mixing state index* χ quantifies the extent to which the particle population is internally mixed by examining how similar the particles are to each other within the population. The values of χ range from 0—reflecting a fully externally mixed particle population—to 100%, reflecting a fully internally mixed particle population. The mixing state index is useful for error quantification studies as shown in Figure 2. Here, the error in cloud condensation nuclei (CCN) concentration is graphed for each grid cell in the modeling domain shown in Figure 2 assuming an internal mixture for the particle population. Positive values indicate that composition-averaging resulted in an overestimation in CCN concentrations. The solid black line indicates the median value within each χ bin (dashed lines marking the 10th and 90th percentile). Relative errors are highest for more externally mixed populations (lower values of χ) but may remain small due to cancellation effects.

For the first time, the particle-resolved model PartMC was validated with experimental data where the aerosol mixing state evolved as two initially externally mixed particle populations underwent agglomeration in an aerosol chamber as part of the Boston College-Aerodyne Research, Inc. 2012 Black Carbon BC3 study. The evolution of the particle population was monitored with an SMPS and an SP2 instrument over the course of several hours. The model simulations included agglomeration, dilution and wall losses, and accounted for the non-spherical shape of the aggregates. We applied an efficient optimization algorithm to determine several unconstrained parameters using the total size distributions and the size distributions of the black carbon particle components. This work provides the foundation for more sophisticated chamber studies that might include additional aerosol processes, such as gas-particle partitioning or particle restructuring, or studies that quantify mixing state impacts on population-integrated quantities, such as total absorption.

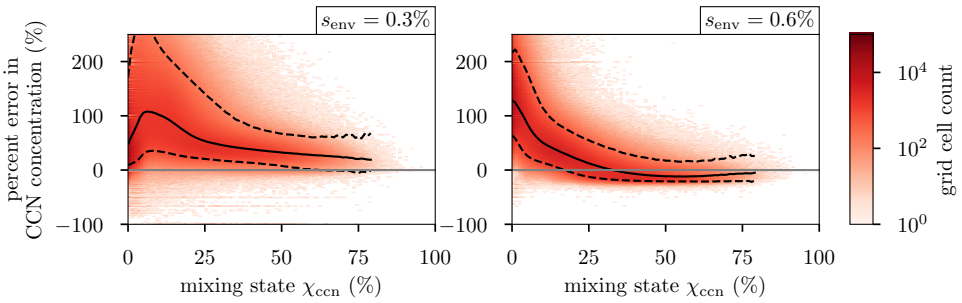

Figure 2: This figure shows the first-ever direct evaluation of the modeling error in cloud condensation nuclei (CCN) concentration when assuming internal mixture. This uses data from the simulations shown in Figure 1 and is based on both our new modeling capability (WRF-PartMC) and our new theoretical quantification of aerosol mixing state, χ. The plot shows the error in CCN number concentrations as a function of mixing state parameter χ for environmental supersaturations of 0.3% and 0.6%. Positive values indicate that assuming internal mixture resulted in an overestimation in CCN concentrations. Solid black lines indicate the median values within each χ bin (dashed lines marking the 10th and 90th percentile). Relative errors are highest for more externally mixed populations (lower values of χ) but may remain small due to cancellation effects.

Involvement in ASR Program Activities

Co-PI Riemer led the effort of a focus group on aerosol mixing state within the Aerosol Processes Working group (2014–2016). The aim of this focus group was to study the sensitivity of aerosol physical and optical properties to the aerosol mixing state, and work to determine the extent to which models need to include all of this complexity. The lasting success of this focus group consists of the development of a rigorously defined mixing state metric by the PIs (Riemer and West, 2013), which was readily adopted by the measurement community. Co-PI Riemer served as co-chair and writing team member of the Absorbing Aerosol Workshop in January 2016 (Cappa et al., 2016). Since 2017 Co-PI Riemer is serving as the Co-Chair of the DOE ASR Aerosol Processes Working Group. Together with Co-Chair Jim Smith, she is taking leadership by coordinating PI activities within the Working Group and by coordinating the agenda of the DOE ASR PI meeting in March.

Publications resulting from this grant

1. J. Ching, N. Riemer, M. West [2016], Black carbon mixing state impacts on cloud microphysical properties: effects of aerosol plume and environmental conditions, J. Geophys. Res. **121**, DOI: 10.1002/2016JD024851.

2. M. D. Willis, R. M. Healy, N. Riemer, M. West, J. M. Wang, C.-H. Jeong, J. C. Wenger, G. J. Evans, J. P. D. Abbatt, and A. K. Y. Lee [2016], Quantification of black carbon mixing state from traffic: implications for aerosol optical properties, Atmos. Chem. Phys. **16**, 4693-4706, DOI: 10.5194/acp-16-4693-2016.

3. R. E. O'Brien, B. Wang, A. Laskin, N. Riemer, M. West, Q. Zhang, Y. Sun, X.-Y. Yu, P. Alpert, D. A. Knopf, M. K. Gilles, R. C. Moffet [2015], Chemical imaging of ambient aerosol

particles: observational constraints on mixing state parameterization, J. Geophys. Res. **120**, DOI: 10.1002/2015JD023480.

4. J. Ching, R. A. Zaveri, R. C. Easter, N. Riemer, J. D. Fast [2016], A three-dimensional sectional representation of aerosol mixing state for simulating optical properties and cloud condensation nuclei, J. Geophys. Res. **121**, DOI: 10.1002/2015JD024323.

5. J. H. Curtis, M. D. Michelotti, N. Riemer, M. T. Heath, M. West [2016], Accelerated simulation of stochastic particle removal processes in particle-resolved aerosol models, J. Computational Phys. **322**, 21-32, DOI: 10.1016/j.jcp.2016.06.029.

6. J. Tian, B. Brem, M. West, T. C. Bond, M. J. Rood, N. Riemer [2017], Simulating aerosol chamber experiments with the particle-resolved aerosol model PartMC, Aerosol Sci. Technol. **51**, 856-867, DOI: 10.1080/02786826.2017.1311988.

7. P. Maginnis, M. West, and G. Dullerud [2016], Variance-reduced simulation of lattice discrete-time Markov chains with applications in reaction networks. J. Computational Phys. **322**, 400–414, DOI: 10.1016/j.jcp.2016.06.019.

8. J. Ching, J. Fast, M. West, N. Riemer [2017], Metrics to quantify the importance of mixing state for CCN activity, Atmos. Chem. Phys. **17**, 7445-7458, DOI: 10.5194/acp-17-7445-2017.

9. J. H. Curtis, N. Riemer, M. West [2017], A single-column particle-resolved model for simulating the vertical distribution of aerosol mixing state: WRF-PartMC-MOSAIC-SCM v1.0, Geosci. Model Dev. **10**, 4057-4079, DOI: 10.5194/gmd-10-4057-2017.

10. J. Ching, M. West, N. Riemer [2018], Quantifying impacts of aerosol mixing state on nucleation-scavenging of black carbon aerosol particles, Atmosphere, 9, **17**, DOI: 10.3390/atmos9010017.

11. C. Shou, N. Riemer, T. B. Onasch, A. J. Sedlacek, A. T. Lambe, E. R. Lewis, P. Davidovits, M. West [2019], Mixing state evolution of agglomerating particles in an aerosol chamber: Comparison of measurements and particle-resolved simulations, Aerosol Sci. Technol. **53**, 1229–1243, DOI: 10.1080/02786826.2019.1661959.

Student theses resulting from this grant

We graduated four Ph.D. students and one M.S. student.

1. P. Gudla [2016], High-order multistep asynchronous splitting methods (MASM) for the numerical solution of multiple-time-scale ordinary differential equations, `http://hdl.handle.net/2142/90556`, Ph.D. thesis.

2. P. Maginnis [2018], Variance-reduced simulation of lattice Markov chains, `http://hdl.handle.net/2142/101032`, Ph.D. thesis.

3. C. Shou [2018], Learning-accelerated algorithms for simulation and optimization, `http://hdl.handle.net/2142/102453`, Ph.D. thesis.

4. J. H. Curtis [2019], Particle-resolved aerosol modeling on the regional scale – insights into importance of capturing aerosol mixing state, `http://hdl.handle.net/2142/105004`, Ph.D. thesis.

5. N. Nuwal [2016], Explicit stochastic schemes for transport in particle-resolved simulations, `http://hdl.handle.net/2142/92971`, M.S. thesis.

Personnel funded under this project

Matthew West (PI), Nicole Riemer (co-PI), Jeffrey Curtis (Ph.D. student), Jessica Gasparik (M.S. student), Pradeep Gudla (Ph.D. student), Michael Hughes (M.S. student), Nakul Nuwal (M.S. student), Chenchao Shou (Ph.D. student), Yu Yao (Ph.D. student), Scott Van Hoy (Undergraduate student).

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