Investigation of spatial and temporal variations in aerosol mixing state using a particle-resolved regional aerosol model

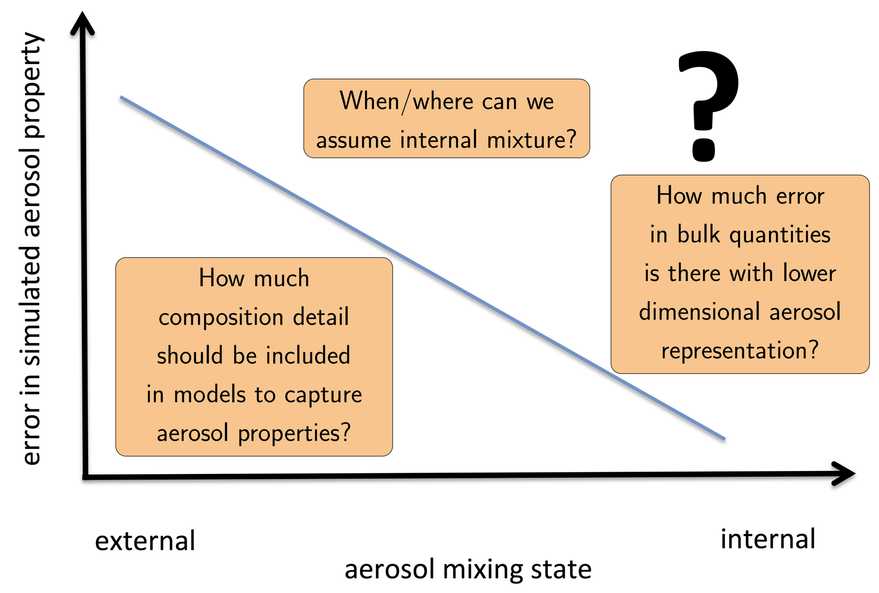
Jeffrey H Curtis
Department of Atmospheric Sciences
University of Illinois at Urbana-Champaign
jcurtis2@illinois.edu

Nicole Riemer
Department of Atmospheric Sciences
University of Illinois at Urbana-Champaign
nriemer@illinois.edu

Matthew West
Department of Mechanical Science and Engineering
University of Illinois at Urbana-Champaign
mwest@illinois.edu

How important is aerosol mixing state?

Particle-level understanding is essential to accurately predict aerosol properties such as optical properties and cloud condensation nuclei activity. Our objective is to develop a 3D particle-resolved model that can capture the complex aerosol mixing states that exist in the atmosphere. Here we present the transformation of WRF-PartMC-MOSAIC from idealized plume scenarios to realistic regional-scale simulations.



Aerosol representation methods

We coupled the Weather Research and Forecast (WRF) model with the PartMC-MOSAIC model. This resulted in the first spatially-resolved, particle-resolving model, which allows for unprecedented levels of detail regarding the simulation of aerosol composition at the regional scale.

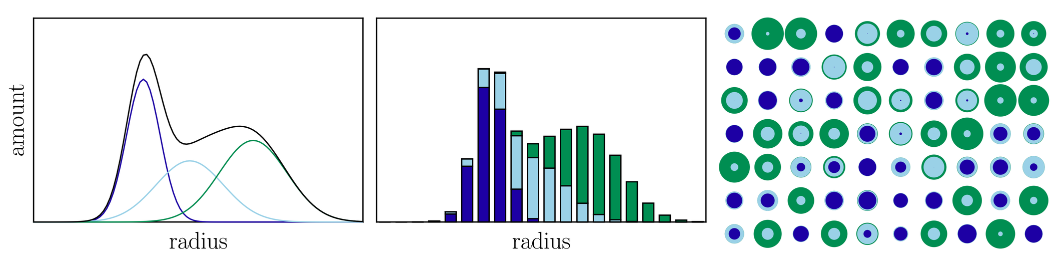


Figure 1: Left: Modal aerosol models represent the aerosol size distribution as a sum of modes. Center: Sectional models store the number or mass of aerosol per bin. Right: By contrast, a particle-resolved model such as PartMC can track complex mixing states.

Source-oriented emissions

Particle-resolved allows for complex representation of aerosol composition. We developed a methodology to take emissions from a mass-based 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.

Importantly, one aerosol species (e.g. BC) can be emitted by different sources within a grid cell. For traditional sectional models these individual emission fluxes are combined to one total species emission, hence the source attribution is lost. Another consequence of the sectional approach is that within a given size bin, the particles all have identical composition (fully internally mixed). In contrast, particle-resolved emissions allow sources with different composition to be represented.

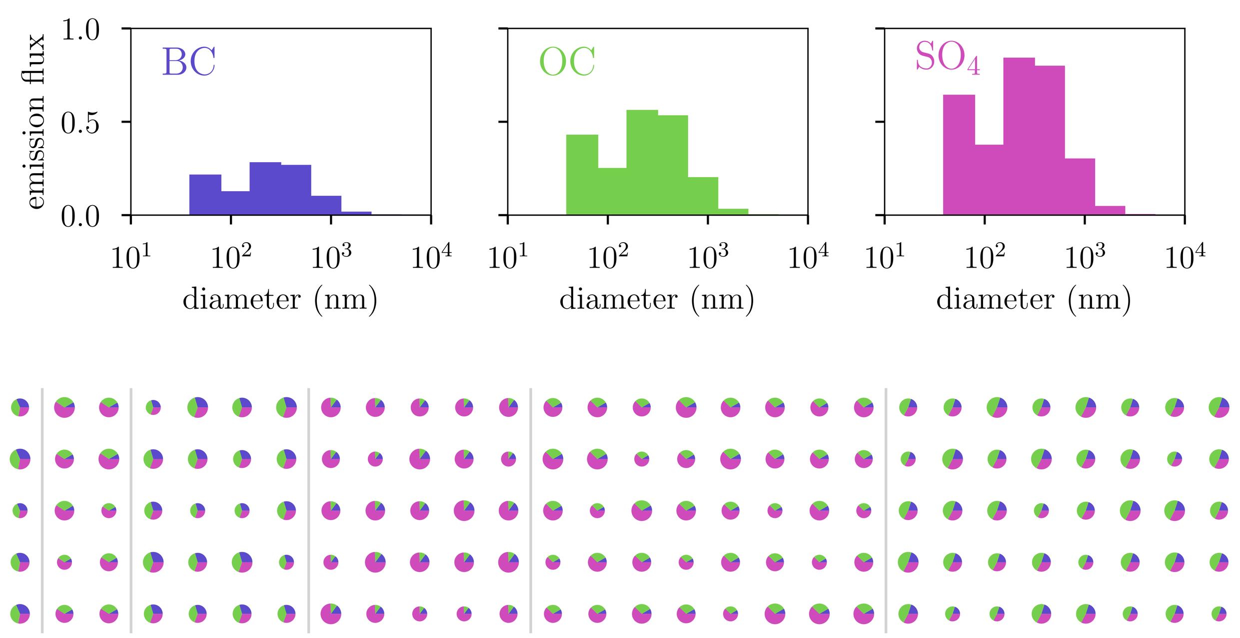


Figure 2: Traditional representation of emissions (top) contrasted with particle-resolved emissions (bottom). Total summed species emissions are equal between the two representations. Within sectional models, emission sources within a grid cell are combined as a total emission. Within a given size bin, the particles all have identical composition (fully internally mixed). In this schematic, the mass fractions for all particles are $BC = \frac{1}{6}$, $OC = \frac{2}{6}$ and $SO_4 = \frac{3}{6}$ (all particles look like ●). With particle-resolved emissions, many sources with different composition can be represented. This feature allows for a diverse and complex mixing state. Here there are six different emission types with varying mass fractions and total number fluxes.

Emissions over California

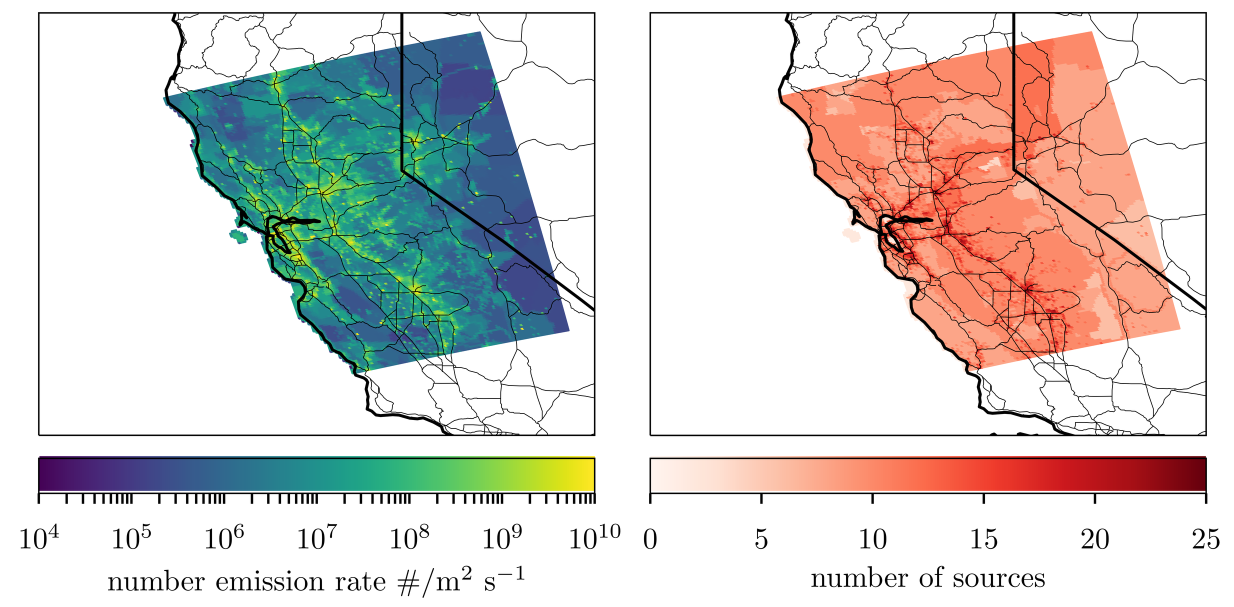


Figure 3: Total particle number emission flux (left) and total number of tracked source modes (right). The urban areas show a larger particle number emission flux as well as a larger number of particle sources per grid cell.

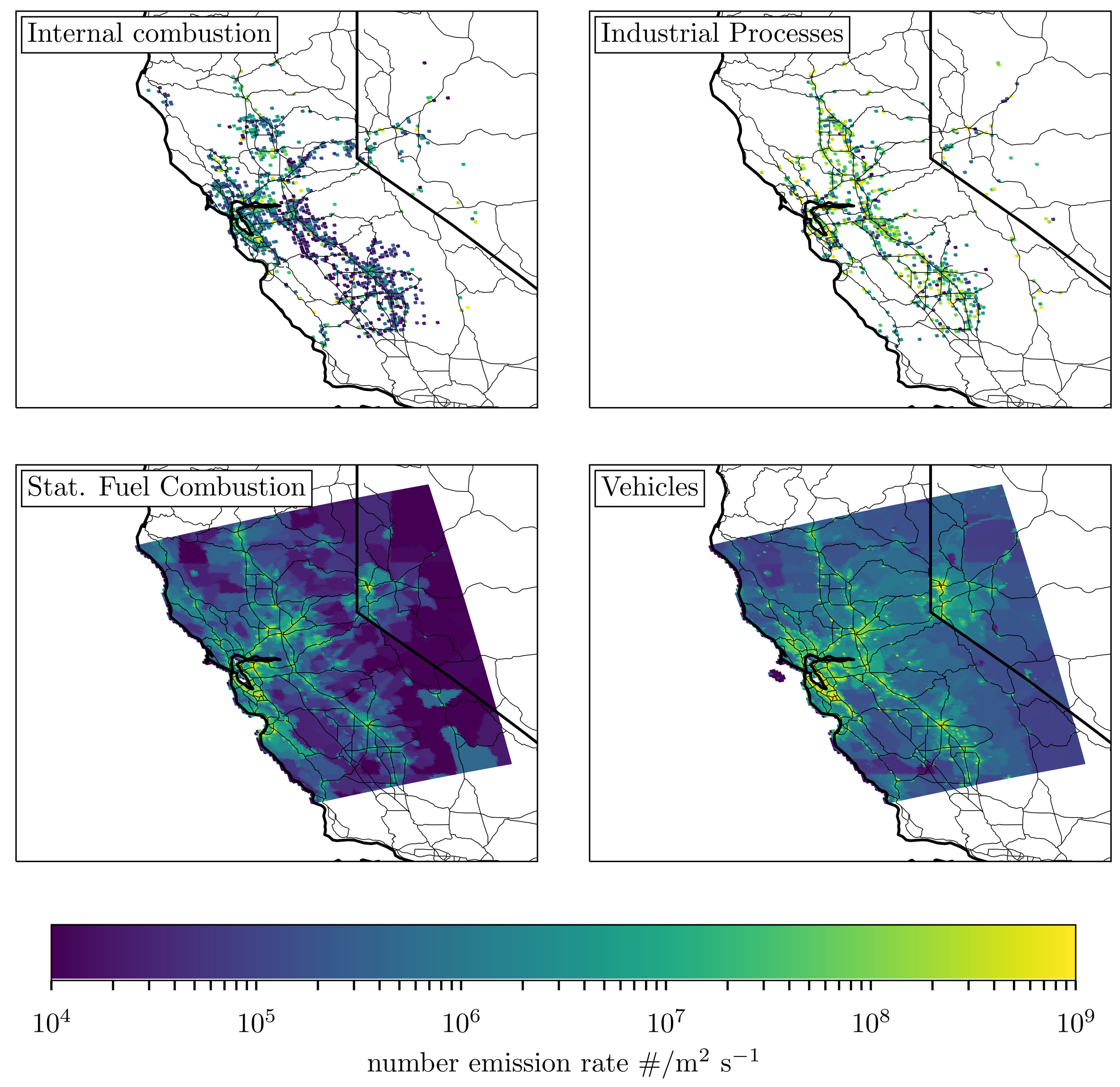


Figure 4: Emission number flux for four different source sectors: Internal Combustion Engines, Industrial Processes (Non-Combustion), Stationary Source Fuel Combustion and Vehicles. Each emission source has a unique composition profile. We track 37 different emission source categories in the presented scenario.

Particle-resolved aerosol results

We simulated the CARES domain of 170 × 160 with 4 km horizontal resolution and 65 vertical levels. Each grid cell contains 10 000 computational particles to model the aerosol population.

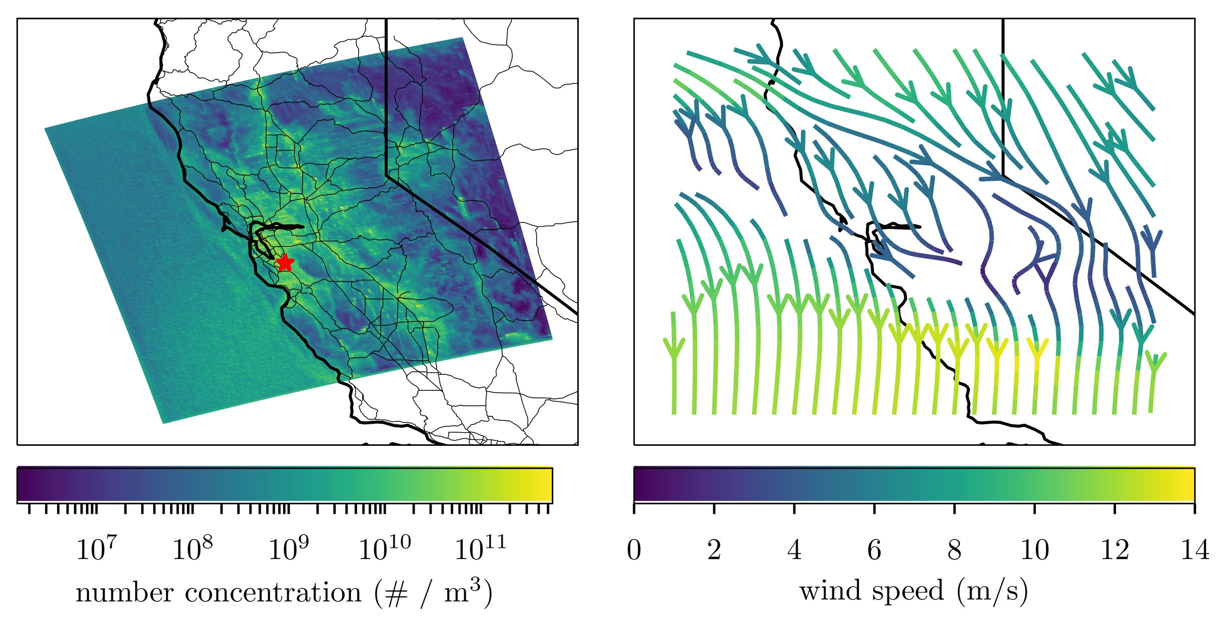


Figure 5: Total aerosol number concentration (left) and wind field (right).

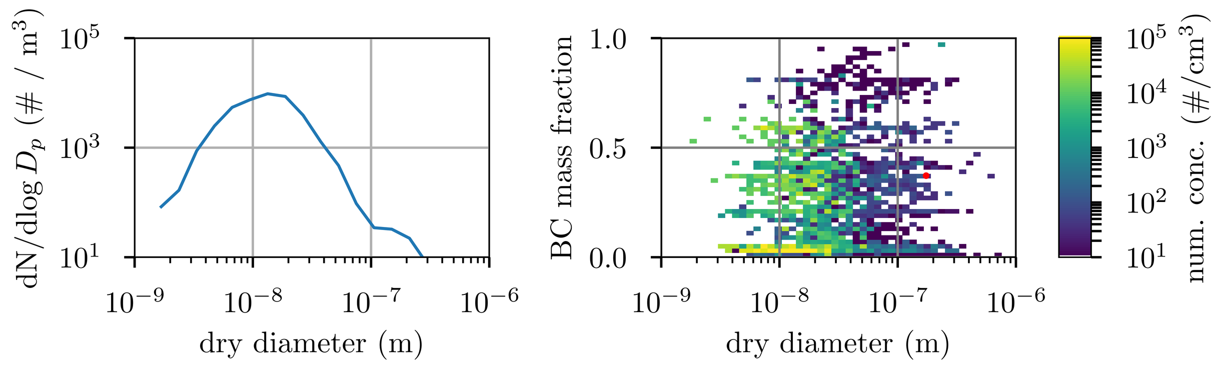


Figure 6: Aerosol size distribution (left) and two-dimensional histogram of black carbon mixing state (right) for location denoted by a star in Fig. 5. WRF-PartMC allows for a continuum of composition.

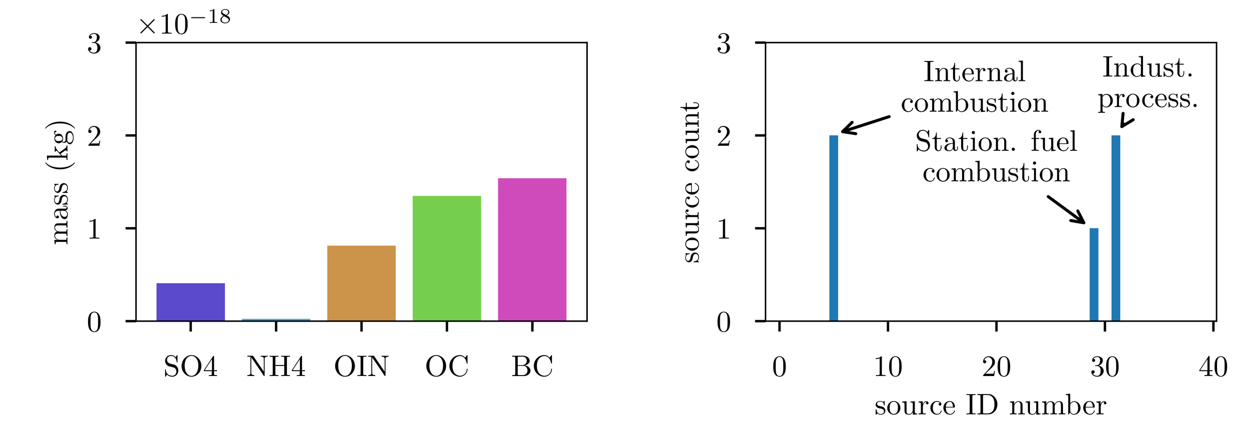


Figure 7: Composition information and source information of a single particle sampled at red point in Fig. 6. WRF-PartMC tracks the composition of individual particles and their source contributions.

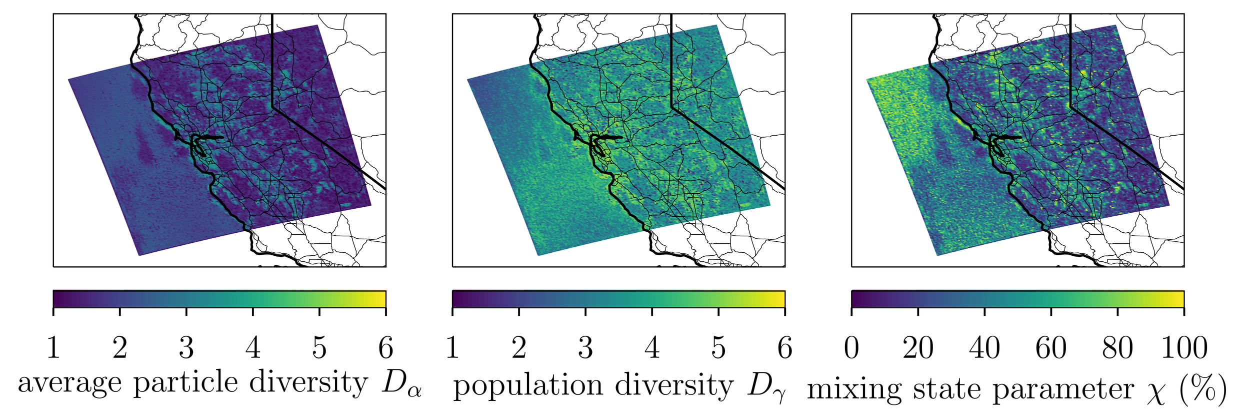


Figure 8: Spatial distribution of mixing state metrics. (left) Average particle diversity D_α is the average effective number of species in each particle in a population, (center) population diversity D_γ is the effective number of species in the aerosol population, and (right) the mixing state index χ is the extent that a population is internally mixed, where $\chi = 0\%$ for a population that is fully externally mixed (i.e., each particle contains a single species) and $\chi = 100\%$ for a fully internally mixed population (i.e., all particles look the same).

Computational demands

CARES simulations are conducted using Blue Waters. The computing capacity of Blue Waters allows for cores to each simulate 1 × 1 tiles of the domain, utilizing 26871 cores for this scenario. A complete 2 day scenario requires 1.3 million core hours.

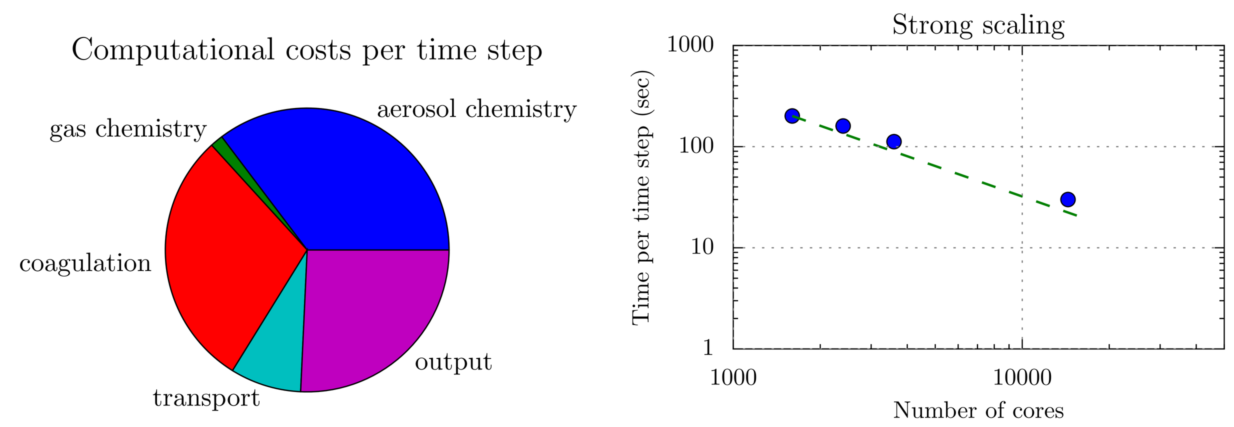


Figure 9: Computational cost (left) is dominated by per-particle chemistry and coagulation. Scaling (right) shows how WRF-PartMC scales nearly linearly as greater number of cores are used.

Conclusions and future work

- WRF-PartMC-MOSAIC was applied to a Northern California scenario using WRF simulated meteorological fields and detailed source-oriented particle-resolved aerosol emissions.
- Particle-resolved simulations allow for examination on a per-particle level of both complex aerosol composition and contributing sources.
- Future work: Quantify errors in optical and CCN properties comparing particle-resolved simulations against of less-detailed models i.e., sectional/bin models (MOSAIC) and modal models (MAM3/MAM4).

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