A 3D Particle-resolved Model to Quantify Spatial and Temporal Variations in Aerosol Mixing State

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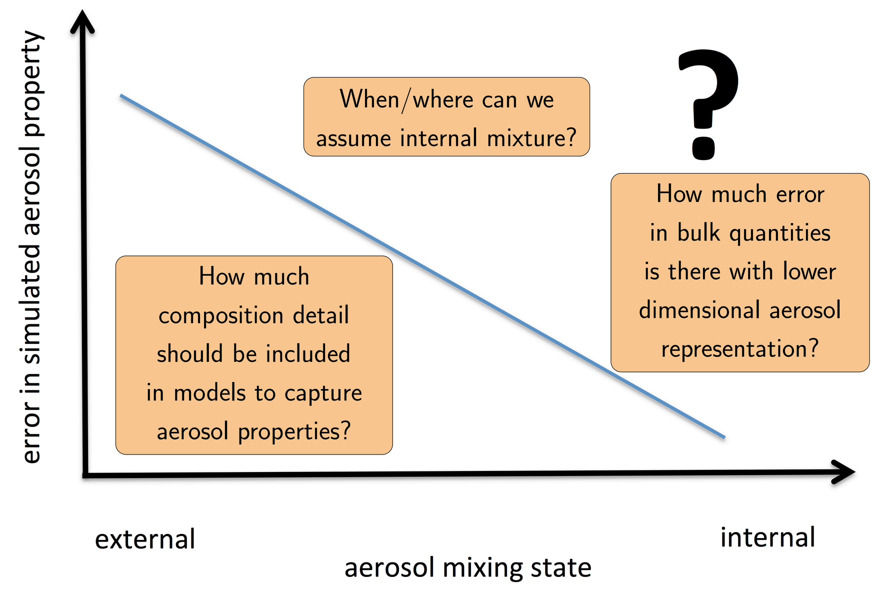
How important is aerosol mixing state?

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.



PartMC-MOSAIC: Arrived at the regional scale

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.

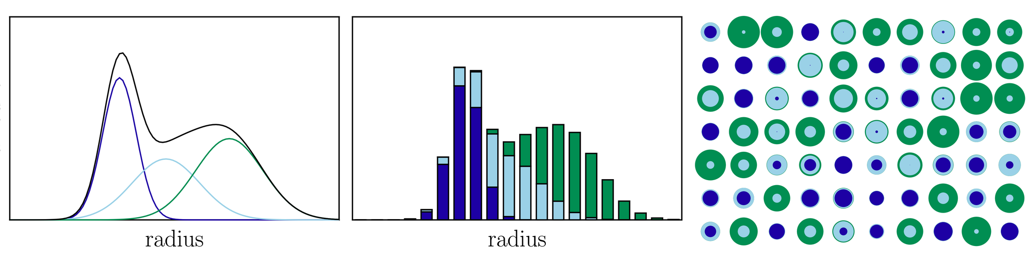


Figure 1: Left: Modal aerosol models represent the aerosol size distribution as a sum of modes. Center: Sectional models store the number or mass of aerosol per bin. Right: By contrast, a particle-resolved model such as PartMC can track complex mixing states.

Stochastic aerosol transport

Aerosol particles are transported using multinomial sampling. The probabilities for moving between grid cells are determined from the discretization of the advection-diffusion equation in space, time and particle number. This approach, in contrast to tracking and updating every particle position each time step, is computationally efficient.

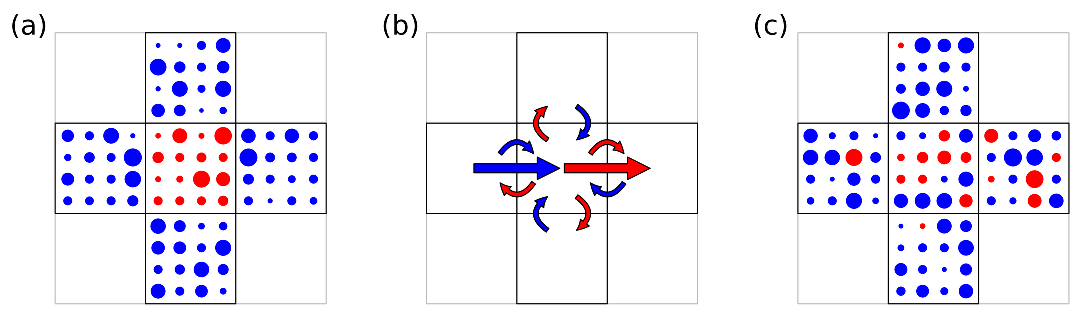


Figure 2: Example of stochastic particle transport from central grid cell to all neighboring grid cells from time t (a) to time $t + \Delta t$ (c). Red particles were sampled with advection probabilities of $\frac{1}{4}$ in the positive x direction and isotropic diffusion of $\frac{1}{16}$. In WRF-PartMC-MOSAIC the propabilities for moving between grid cells are determined from the simulated wind field and turbulent diffusion coefficient.

Source-oriented particle-resolved emissions

Particle-resolved modeling allows for the complex representation of aerosol composition. We developed a framework that takes mass-based aerosol emission fluxes as it is typical for traditional WRF-Chem simulations, and converts them to number-based emissions fluxes, consistent with the PartMC framework. With this approach the source information of particles can be easily tracked.

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 flux, 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.

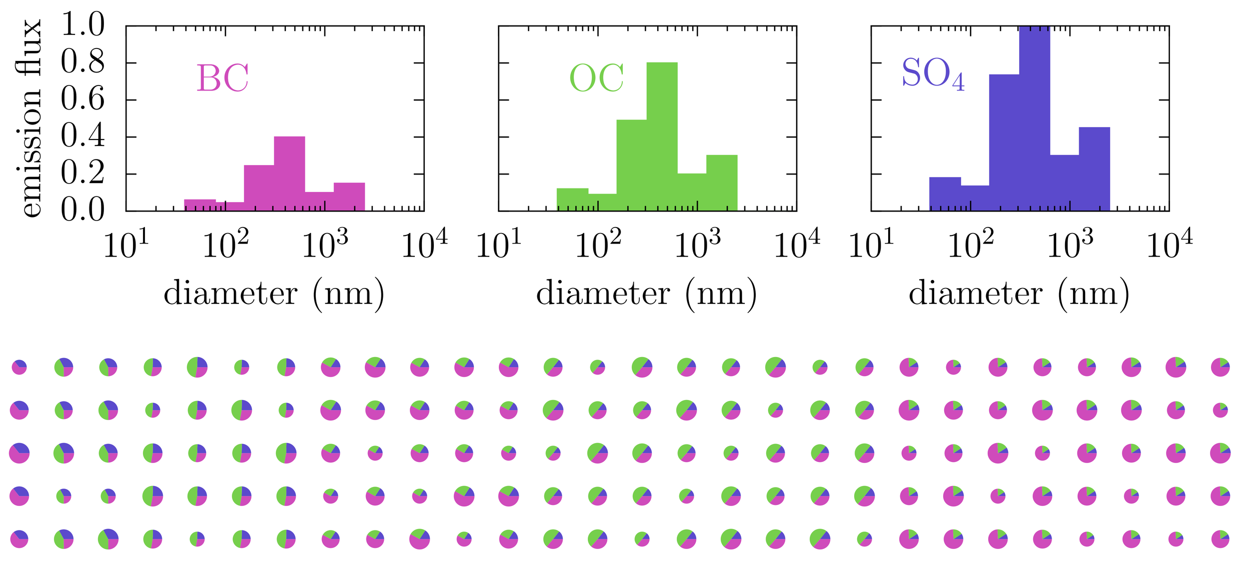


Figure 3: Traditional, size-resolved representation of emissions (top) contrasted with the true, underlying particle-resolved emissions (bottom). Both representations have the same bulk mass emission fluxes, where the ratio $\mathrm{BC} : \mathrm{OC} : \mathrm{SO_4}$ is 1:2:3. The particle-resolved emissions originate from six different sources with differ in their compositions and emission fluxes.

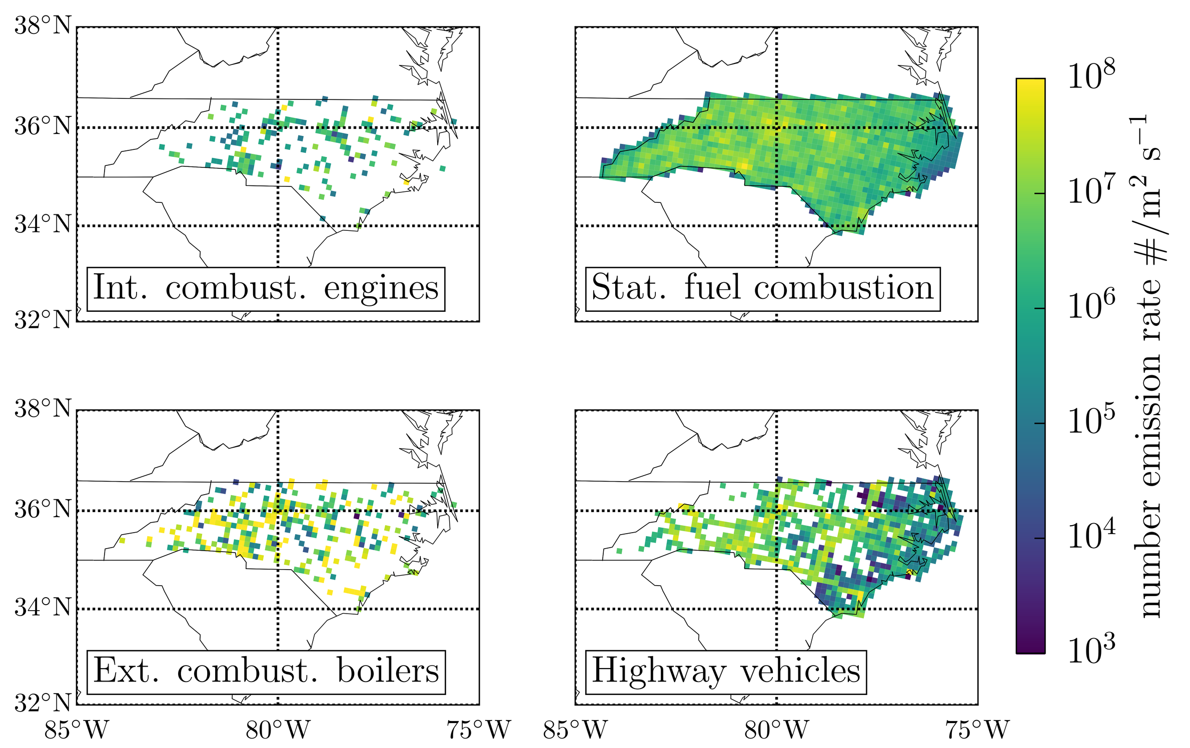


Figure 4: Emission number flux for four different sources: Internal combustion boilers, off-highway vehicles, external combustion boilers and highway vehicles. Each emission source has its unique composition profile.

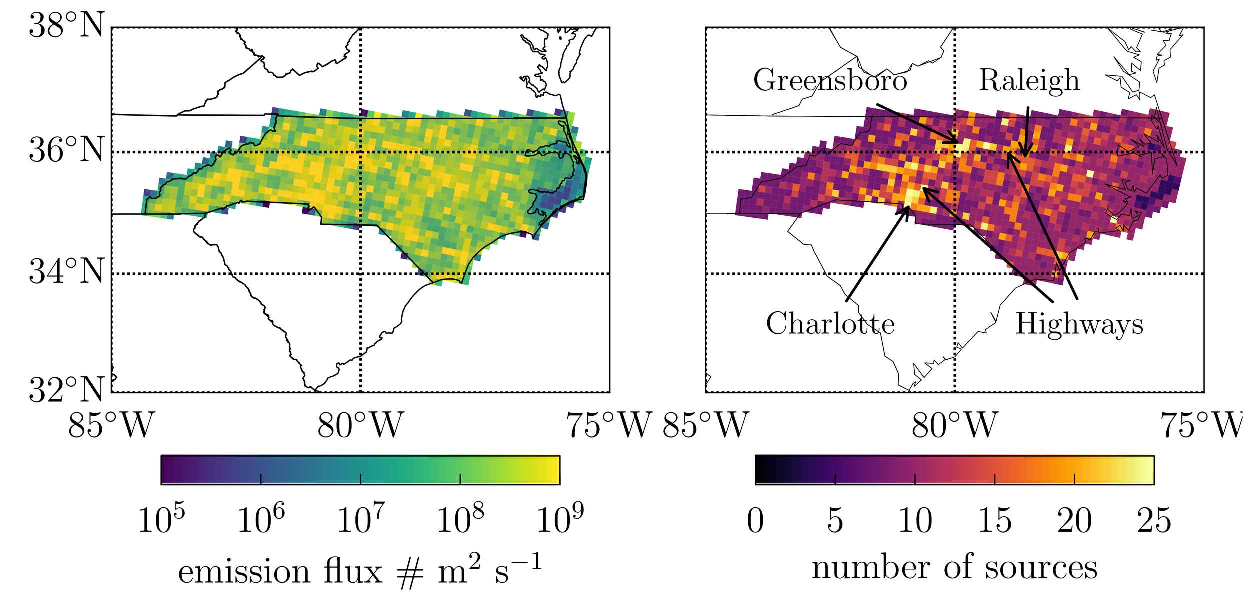


Figure 5: Total particle number emission flux (left) and total number of tracked source modes (right). The urban areas show a larger particle number emission flux as well as a larger number of particle sources per grid cell.

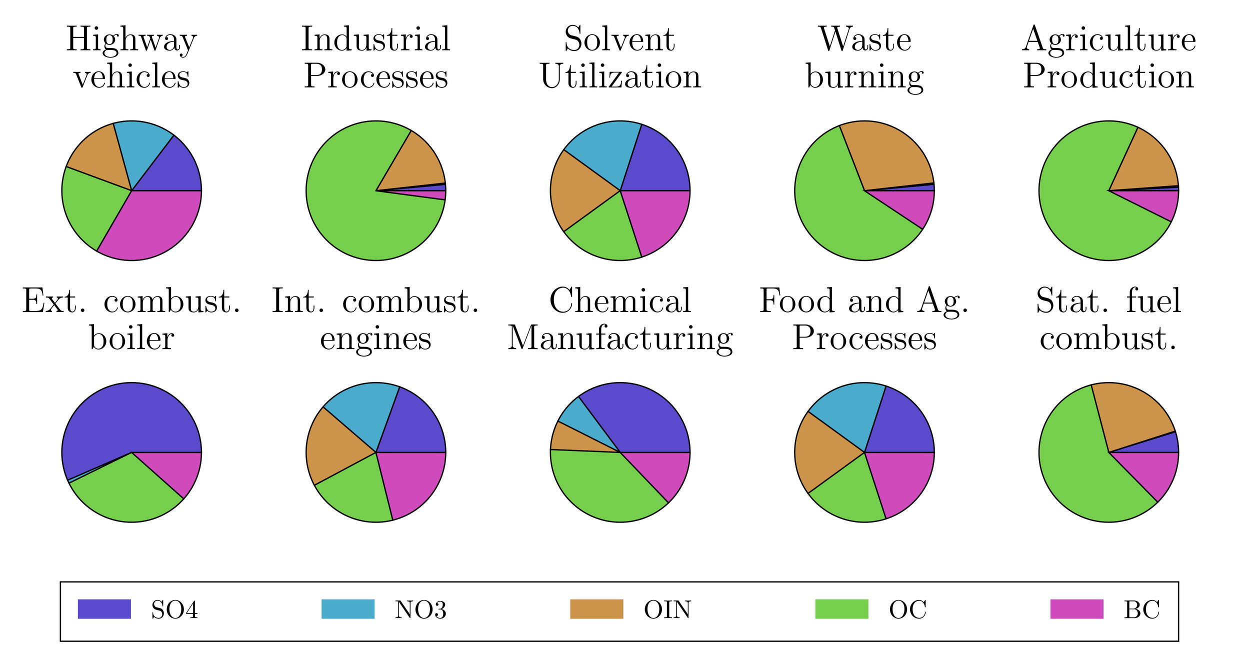


Figure 6: Composition profile of selected particle emission sources. Every tracked emission source is sampled from a prescribed lognormal size distribution (specified by total number concentration, geometric mean diameter and geometric standard deviation) and particle composition. For the simulation presented here we included 30 different emission sources.

Particle-resolved modeling results

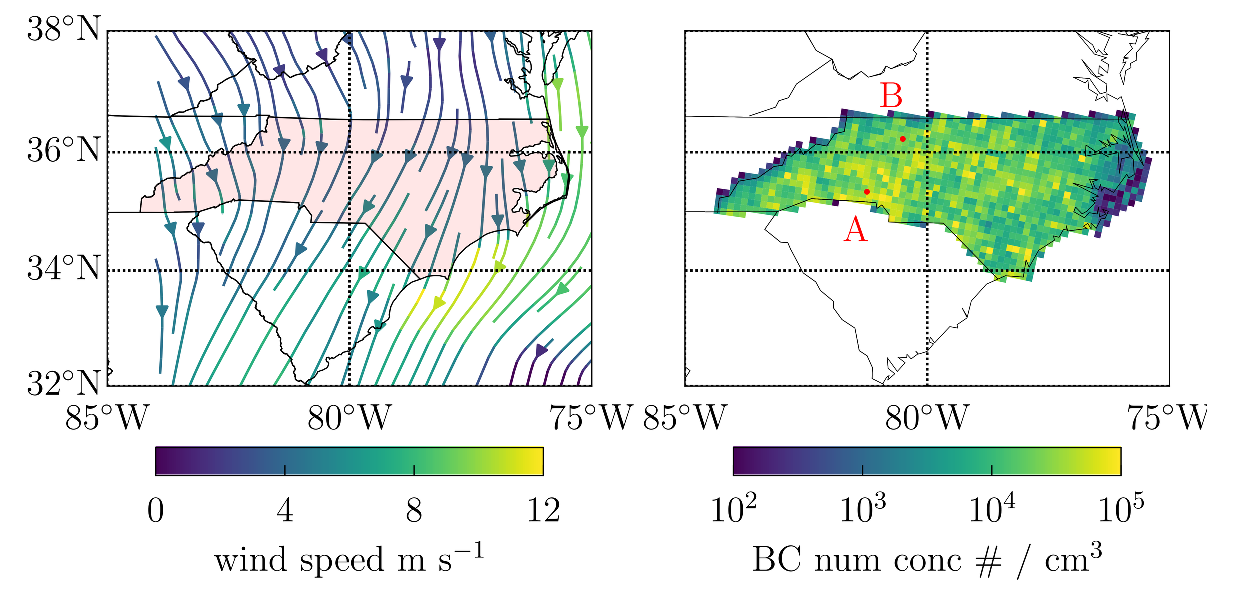


Figure 7: Wind field (left) and simulated number concentration of black-carbon-containing particles (right).

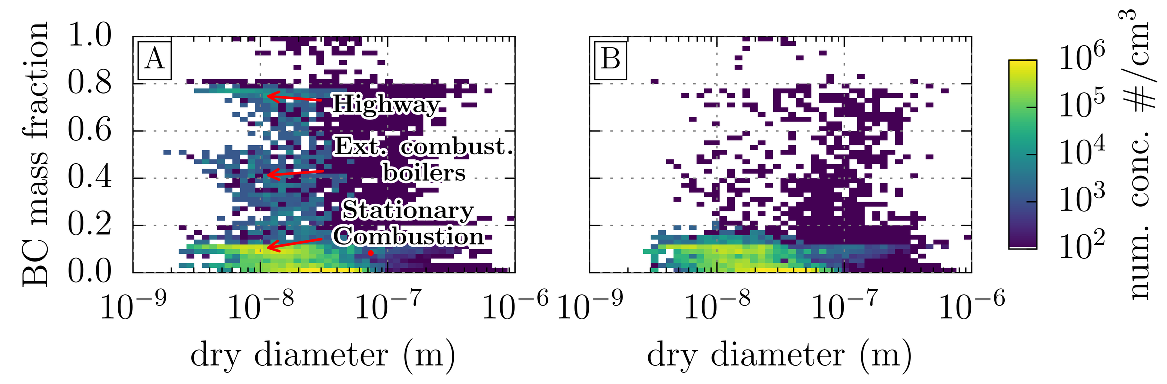


Figure 8: 2D histograms of black carbon mixing state at points A and B.

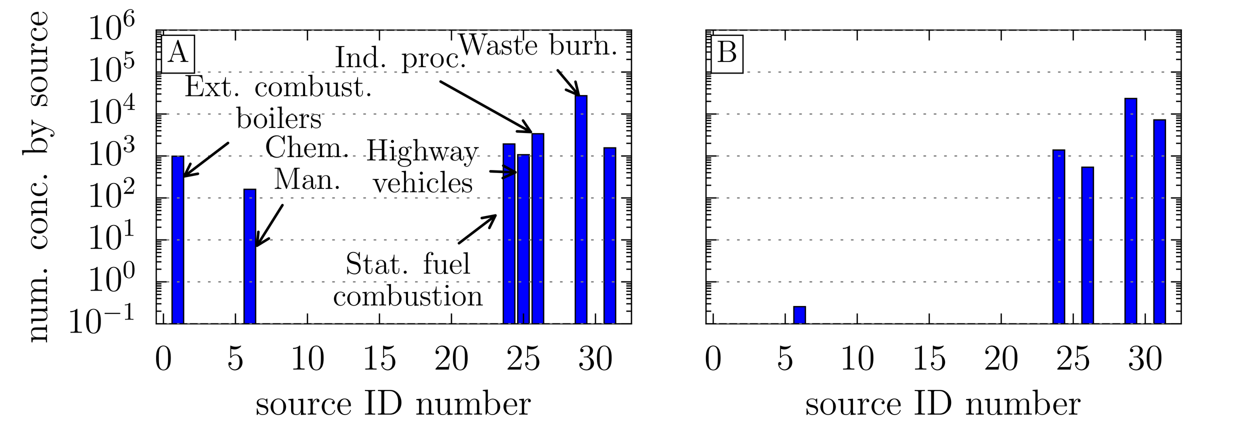


Figure 9: Particle source information aerosol populations at points A and B. The originating source of all particles is tracked throughout a simulation.

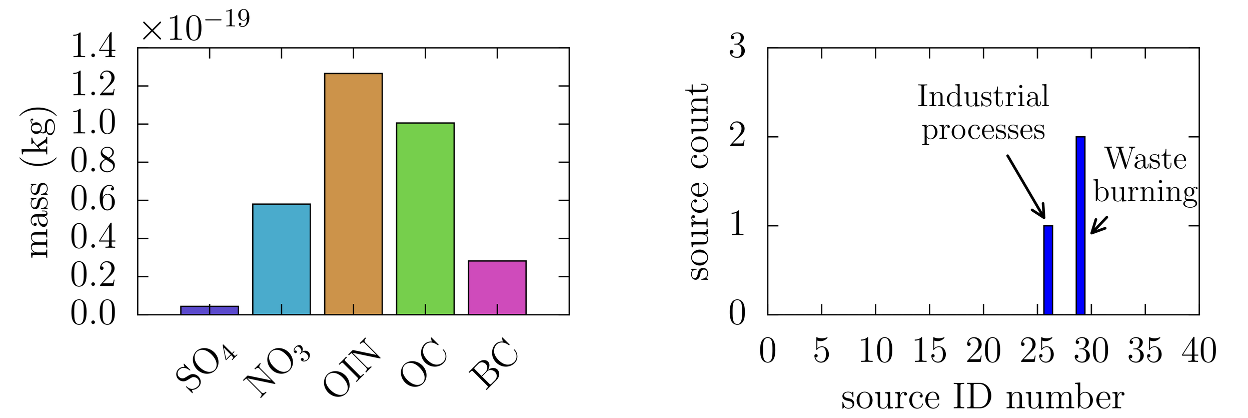
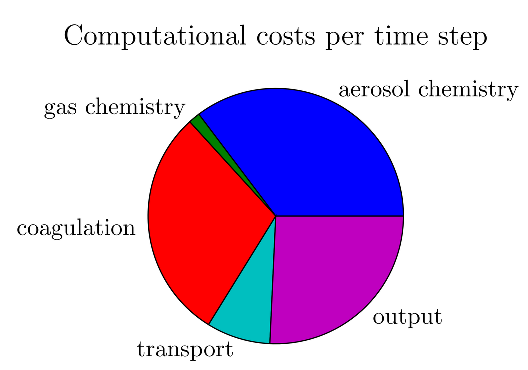


Figure 10: Composition information and source information of a single particle at point A.

Computational demands



domain of interest	required storage
North Carolina (1 day)	5 TB
CARES (1 day)	230 TB
CARES (3 days)	690 TB

Figure 11: Left: Summary of computational time spent on various processes in WRF-PartMC-MOSAIC. Right: Storage requirements to allow for hourly output of full particle states.

Conclusions and future work

- WRF-PartMC-MOSAIC was applied to a realistic scenario using WRF simulated meteorology fields and detailed source-oriented particle-resolved aerosol emissions.
- A 12-hour simulation for the North Carolina domain on Blue Waters takes 9 hours using 3,604 cores. The model shows excellent scaling behavior to large number of cores.
- WRF-PartMC-MOSAIC will next be applied to a northern California domain to model CARES.