Towards a Particle-Resolved Aerosol Representation for the Simulation of the Aerosol Impact on Regional Scales

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Why Particle-Resolved Aerosol Models?

Individual aerosol particles are a complex mixture of a wide variety of species, such as soluble inorganic salts and acids, insoluble crustal materials, trace metals, and carbonaceous materials, requiring a high-dimensional representation. The capabilities of traditional models to treat this high dimensionality are currently limited, and this introduces shortcomings in our understanding of the impact of aerosol particles on climate. To improve this we have recently introduced the particle-resolved model PartMC-MOSAIC [Riemer et al., 2009, Zaveri et al., 2008], a new approach to examining the evolution of aerosol properties without making a priori assumptions about the evolution of particle composition.

Capabilities of PartMC-MOSAIC

The stochastic particle-resolved aerosol model PartMC tracks the chemical composition of individual aerosol particles in a Lagrangian box. Particle emissions, dilution with the background, and Brownian coagulation are simulated stochastically by generating a realization of a Poisson process. Gas- and aerosol-phase chemistry is implemented deterministically by coupling with the MOSAIC chemistry code. PartMC-MOSAIC accurately predicts both number and mass size distributions and is therefore suited for applications where either quantity is required. Figure 1 shows an example of the model output of black carbon mixing state and the corresponding critical supersaturation as two-dimensional projections of the 20-dimensional number distribution.

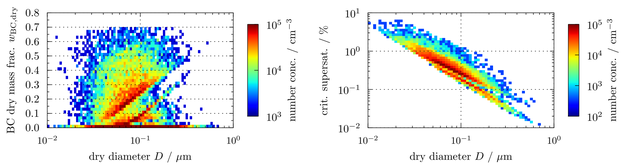

Figure 1: Two-dimensional projections of the 20-dimensional number distribution after 7 hours of simulation. Left: Black carbon mixing state. A continuum of mixing states formed between the extreme mixing states of $w_{\mathrm{BC,dry}} = 0\%$ (particles that do not contain BC) and $w_{\mathrm{BC,dry}} = 70\%$ (freshly emitted diesel particles) as a result of coagulation and condensation processes. At a given size, a wide range of particles exists with different mixing states. Right: critical supersaturation. Given a certain particle size, the critical supersaturation ranges over about one order of magnitude.

The Path For Going Beyond The Box

While particle-resolved aerosol simulations offer unprecedented resolution of the aerosol mixing state, they are significantly more expensive than conventional models. Here we explore strategies to improve the efficiency of PartMC-MOSAIC to enable its extension from a Lagrangian box model to a model that is coupled to a 1D (and eventually 3D) transport code. We focus on two specific aspects: (1) a new algorithm for parallel simulation, and (2) "superparticles" as a coarse-graining method, with each superparticle representing a collection of physical particles.

Parallel Simulation via Mixing

We have developed a new parallel simulation algorithm for the box-model PartMC method. This consists of having several processor cores timestep independent parallel simulations (denoted $\Phi_{\Delta t}^{\mathrm{para}}$), combined with mixing particles randomly between the different processors at each timestep (denoted Φ_p^{mix}), where, with probability p, each particle is transferred to a different randomly chosen processor at the end of a timestep. Using Φ^{proj} for the projection of all particles to a single core, and $\Phi_{\Delta t}^{\mathrm{cent}}$ for the centralized (single core) simulation algorithm, we prove that this method is weakly convergent at rate $\mathcal{O}(\sqrt{\Delta t})$:

$$\Phi^{\mathrm{proj}} \circ \left(\Phi_p^{\mathrm{mix}} \circ \Phi_{\Delta t}^{\mathrm{para}}\right)^{T/\Delta t} \rightharpoonup \Phi_T^{\mathrm{cent}} \circ \Phi^{\mathrm{proj}} \text{ as } \Delta t \to 0 \text{ for } p > 0.$$

As we see from the numerical results in Figure 2, when the mixing rate is zero ($p = 0$), the simulation displays some inaccuracies. Even small mixing probabilities ($p = 0.01$), however, accurately capture the system evolution. This is quantified in Figure 3, where we see that as the number of processor cores increases the zero mixing solution fails to converge correctly, while even small mixing rates ensure the expected $N^{-1/2}$ convergence rate. Here the total number of particles N is proportional to the number of cores, as we hold constant the particles per core. Regarding scalability, we see that we are about 25% slower than ideal up to hundreds of cores.

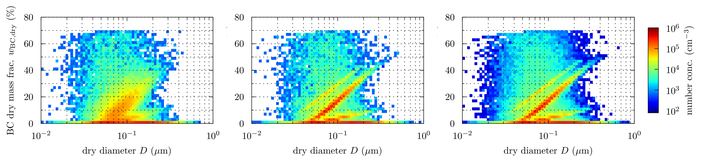

Figure 2: Mixing-based parallel particle code results. Left: parallel simulation with 512 processor cores and 32 particles per core (16 384 particles total), and no mixing ($p = 0$). Center: the same parallel simulation but with a small amount of mixing ($p = 0.01$). Right: A benchmark comparison calculation with 10^5 particles on a single processor core. We observe that even small amounts of mixing are enough for the parallel simulation to accurately resolve the particle population evolution.

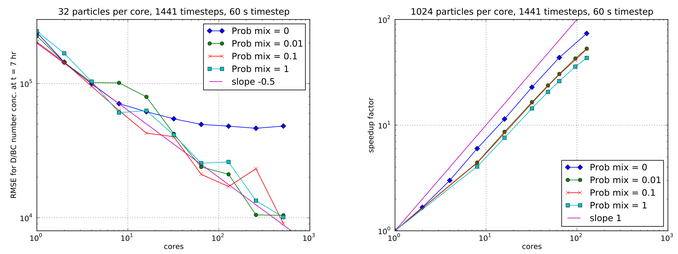

Figure 3: Convergence and speedup for parallel code. Left: simulation error versus number of cores. Right: simulation speedup factor versus number of cores. In both cases a constant number of particles per core is used (32 on the left, 1024 on the right). We see that small mixing rates ($p = 0.01$) are sufficient to achieve good error convergence (left) while having speedup only about 25% less than ideal (right).

Superparticles

A direct particle-resolved method in which one represents each "physical" particle by a single "computational" particle tends to have the following difficulty: either the population of large particles is under-resolved and the accuracy of the simulation is significantly compromised, or the total number of particles in the entire simulation must be very large. The superparticle method allows coarse-graining where each "superparticle" in our new scheme represents a family of identical particles. This means that each computational particle is "weighted" by an appropriate factor so that the true number distribution $n(D)$ is given by $n(D) = w(D)c(D)$, where $w(D)$ is a fixed weighting function, $c(D)$ is the computational (simulated) number distribution, and D is the diameter. Such an efficiency increase moves certain problems into the realm of tractability, as demonstrated with the example of new particle formation by nucleation (Figure 4).

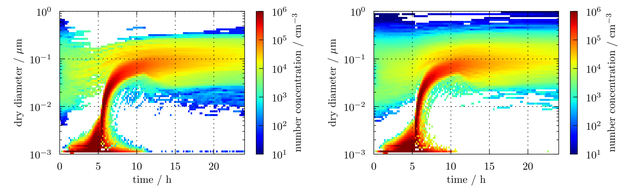

Figure 4: Temporal development of an aerosol particle size distribution after a nucleation burst. Left: weighting factor $w(D) = 1$. Right: weighting factor $w(D) = D^{-1}$. The superparticle method ensures that the whole range of sizes—from a few nanometers for the freshly nucleated particles to micrometers for the background particles—can be resolved with practical levels of computation.

Conclusion

Particle-resolved aerosol models represent a new way of simulating multidimensional aerosol size distributions. They do not rely on ad hoc assumptions regarding the aerosol mixing state. These capabilities come with high computational costs. Here we show two paths for improving the efficiency of such models:

1. Parallel Simulation via Mixing: The particle population is distributed over several processors. At each timestep processors simulate independently, then exchange particles with a given probability. We showed that this algorithm converges and scales near-linearly up to hundreds of cores.
2. Superparticle Method: This allows coarse-graining where each "superparticle" in our new scheme represents a family of identical physical particles. This method is useful for applications where different types of particles need to be tracked that have very disparate concentrations.

References

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