Idealized Particle-Resolved Large-Eddy Simulations to Evaluate the Impact of Emissions Spatial Heterogeneity on CCN Activity

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Abstract. Aerosol-cloud interactions remain a large source of uncertainty in global climate models (GCMs) due to complex, nonlinear processes that alter aerosol properties and the inability to represent the full compositional complexity of aerosol populations within large-scale modeling frameworks. The spatial resolution of GCMs is often coarser than the scale of the spatially varying emissions in the modeled geographic region. This results in diffuse, uniform concentration fields of primary aerosol and gas-phase species instead of spatially heterogeneous concentrations. Aerosol processes such as gas-particle partitioning and coagulation are concentration-dependent in a non-linear manner, and thus the representation of spatially heterogeneous emissions impacts aerosol aging and properties. This includes climate-relevant quantities key to aerosol-cloud interactions including particle hygroscopicity and cloud condensation nuclei (CCN) activity. We investigate the impact of emissions spatial heterogeneity on aerosol properties including CCN activity via a series of first-of-a-kind particle-resolved large-eddy simulations with the modeling framework WRF-PartMC-MOSAIC-LES. CCN concentrations within the planetary boundary layer (PBL) are compared across numerous scenarios ranging in emissions spatial heterogeneity. We find that CCN concentrations at low supersaturations ($S = 0.1$–0.3%) increase in the upper PBL by up to 25% for emissions scenarios with high spatial heterogeneity when compared to a uniform emissions base case.

1 Introduction

- Impact of aerosols on climate, focus on aerosol-cloud interactions
  - Aerosols alter Earth's radiative budget both directly through scattering and absorption of radiation and indirectly through aerosol-cloud interactions.
  - Hygroscopic aerosol particles act as cloud condensation nuclei (CCN), allowing water vapor to condense onto their surface at ambient supersaturations S typical of the troposphere.

  - Abundance of CCN affects cloud properties (morphology, lifetime, indirect radiative effects), thus important to constrain estimates of CCN concentrations due to impact on ACI.

- GCMs do a poor job of predicting CCN activity

  - Inability to represent the scales of transport
  - Do not resolve scale of emissions, result in diffuse and uniform concentrations that lead to different conclusions than what actually results from spatially heterogeneous distributions of gases and aerosols that vary in concentration (this matters because coagulation and gas particle partitioning are concentration dependent processes)
  - Numerous studies have investigated the sub-grid variability present in domain scales typical of GCM grids, found that climate relevant properties like aerosol optical properties and CCN activity differ from GCM-resolution modeled values. Note that the scale of these studies often does not extend down to the sub-kilometer range (scale of emissions heterogeneity and turbulent transport)

- Relevance of the aerosol representation

  - Structural uncertainty resulting from the use of coarse aerosol treatments (i.e., modal or sectional) impact aerosol properties like optical properties (Crippa 2017) and CCN activity (Zaveri 2010, Fierce 2024).
  - Particle resolved treatment, allows per particle aging and process level analysis of changes to aerosol properties such as CCN activity

- State of aerosol-transport models

  - Coarse-scale models: RANS for transport and sectional or modal treatments for aerosols
  - Recent developments in turbulence-resolving aerosol aware models (UCLALES-SALSA, DALES, others?)
  - Development of regional scale particle-resolved simulations with WRF-PartMC
  - No modeling framework has yet to leverage particle resolved aerosol treatment alongside turbulence resolving transport models such as LES

- Objectives of this paper

  - Present a first of a kind particle-resolved large-eddy simulation model, WRF-PartMC-MOSAIC-LES
  - Investigate impacts of emissions spatial heterogeneity on aerosol state including CCN activity via numerous idealized simulations

2 Methods

2.1 WRF-PartMC-MOSAIC-LES

- Extension of Jeff's work in developing WRF-PartMC (Curtis et al. 2024), briefly describe stochastic advection algorithm work to couple WRF and PartMC
- Discuss the LES model (using Deardorff's TKE scheme for sub-grid scale parameterization of eddy diffusivity, spatial resolution of the domain etc.)
- MOSAIC: handles gas phase chemistry, gas-particle partitioning and aerosol thermodynamics, and simplified radiation for photolysis reactions

2.2 Computational domain setup

- Idealized configuration: Domain is homogeneous with no topography or variations in surface characteristics (i.e., surface roughness, heat fluxes, etc.)
- Initial conditions (meteorology, gas and aerosols), mention spin up period as well

2.3 Emissions Scenarios

- Scenarios chosen across a range of spatial heterogeneity, note that emission rates are scaled by 1 over the fraction of area covered by emissions relative to the uniform base case in which emissions cover the entire domain surface
- Spatial heterogeneity is quantified using SH metric (Mohebalhojeh et al. 2024)

3 Results

3.1 Aerosol size distributions

- Number distributions: Number concentration of Aitken mode particles reduced as SH increases due to brownian coagulation, correspondingly, number of accumulation mode particles increases
- Mass distribution: Mass concentrations in accumulation mode increase for high SH scenarios due to both coagulation of smaller particles and gas-particle partitioning of ammonia, nitric acid

3.2 Aerosol composition

- Sulfate time-height plot: Sulfate levels decrease as SH increases due to greater competition for OH in high-concentration plumes resulting in less H2SO4

- Ammonium and Nitrate time-height plots: Notable increase in the upper boundary layer for each under high SH scenarios. Present in upper boundary layer because of strong temperature dependence on ammonium nitrate formation. Under high SH scenarios with less sulfate, there will exist more free ammonia available to neutralize nitrate thus resulting in ammonium nitrate formation.

- Mass fraction figures: Comparing particles in the size range 50-100 nm, particles in the base case are primarily composed of BC and OC (low hygroscopicity), whereas sulfate, nitrate, and ammonium dominate the mass fraction in the same size range for high heterogeneity scenarios. This indicates that particles in the size range whose CCN activity hinges on aerosol hygroscopicity are more hygroscopic under high SH scenarios and thus activate at lower supersaturations.

- 2D kappa distributions: Supporting the conclusions of the mass fraction figures, there are considerably more particles with high kappa at 100 nm compared to the base case. We also find that enhancement of coagulation and gas-particle partitioning in high SH scenarios increases the hygroscopicity of emitted primary aerosol.

3.3 CCN activity

- Vertical profiles of CCN concentrations at each supersaturation: Competition between coagulation enhancement under high emissions SH and increased ammonium nitrate formation results in a complex effect on CCN that is dependent on supersaturation. At lower supersaturations characterized by larger, hygroscopic particles, CCN activity increases with with emissions SH. Smaller particles will activate at higher supersaturations, and we find that the enhancement of coagulation for scenarios with high emissions SH wins out, resulting in fewer CCN activating at high supersaturations.

- Time height plots for percent difference in CCN concentrations relative to the base case

3.4 Influence of ammonia on aerosol composition and CCN activity

- Vertical profile for CCN concentrations in absence of ammonia

4 Conclusions

- Restate the motivation for this work

- Summarize key contributions

- Summarize findings and implications

- Limitations and future work

Code availability. TEXT

Data availability. TEXT

Code and data availability. TEXT

Sample availability. TEXT

Video supplement. TEXT

Appendix A

A1

Author contributions. TEXT

Competing interests. TEXT

Disclaimer. TEXT

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References

REFERENCE 1

REFERENCE 2