Idealized particle-resolved large-eddy simulations to evaluate the impact of emissions spatial heterogeneity on CCN activity

Samuel Frederick, Matin Mohebalhojeh, Jeffrey Curtis, Matthew West, Nicole Riemer

Department of Atmospheric Sciences, University of Illinois Urbana-Champaign

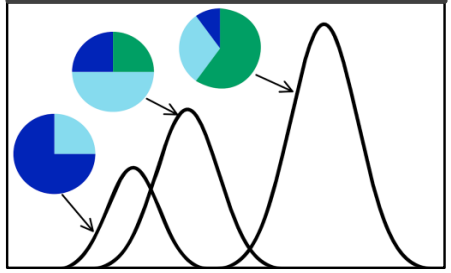
What is structural uncertainty?

Coarse-resolving models

GCM

\+

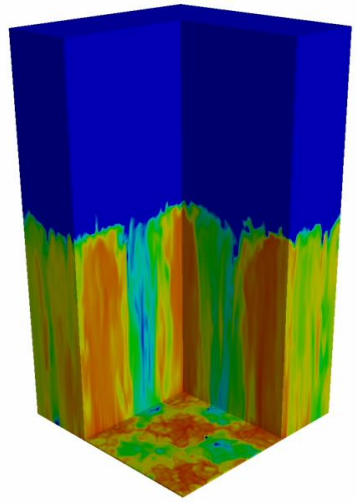
Modal Aerosol Treatment



Structural Uncertainty

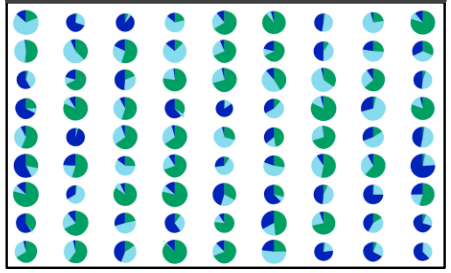
CCN concentrations, aerosol optical properties, etc.

LES



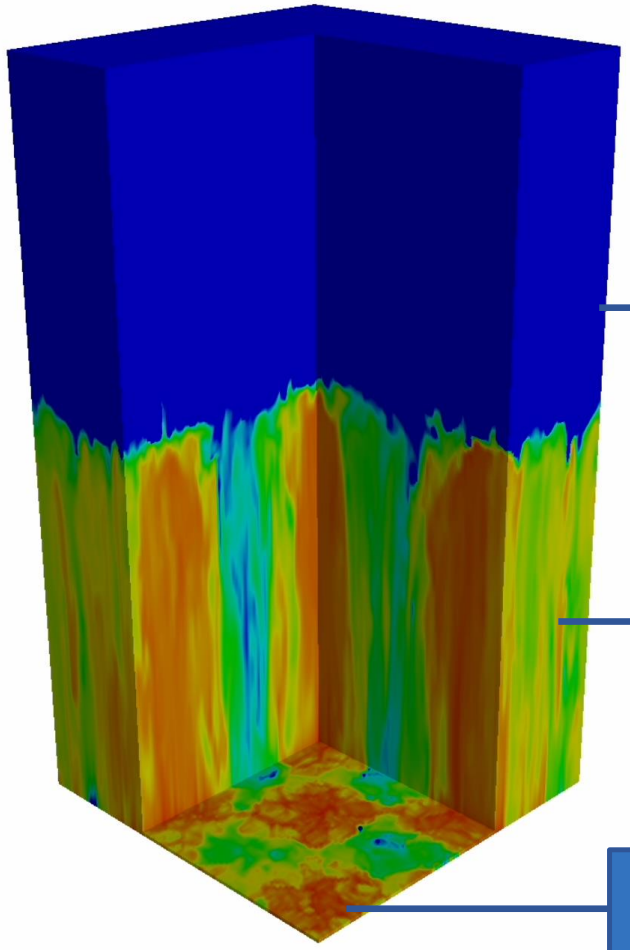
\+

Particle-Resolved Treatment



Fine-resolving models

Emissions contribute to the propagation of structural uncertainty



Cloud properties

Aerosol sub-grid processes

Drivers (Emissions)

Spatial heterogeneity impact?

Spatial heterogeneity impact?

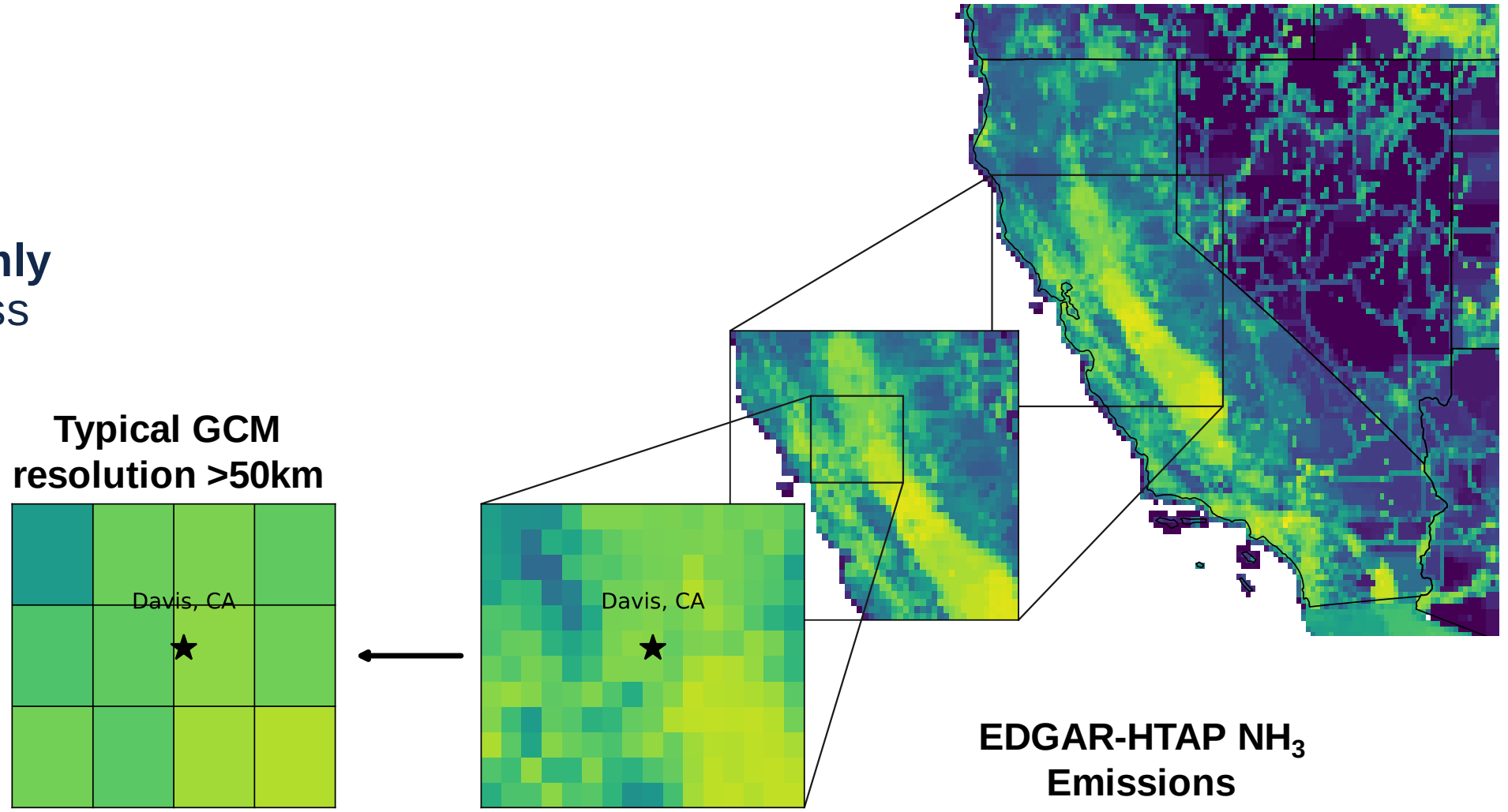
Climate-relevant quantities like CCN activity

Depend on aerosol number concentration, composition, etc.

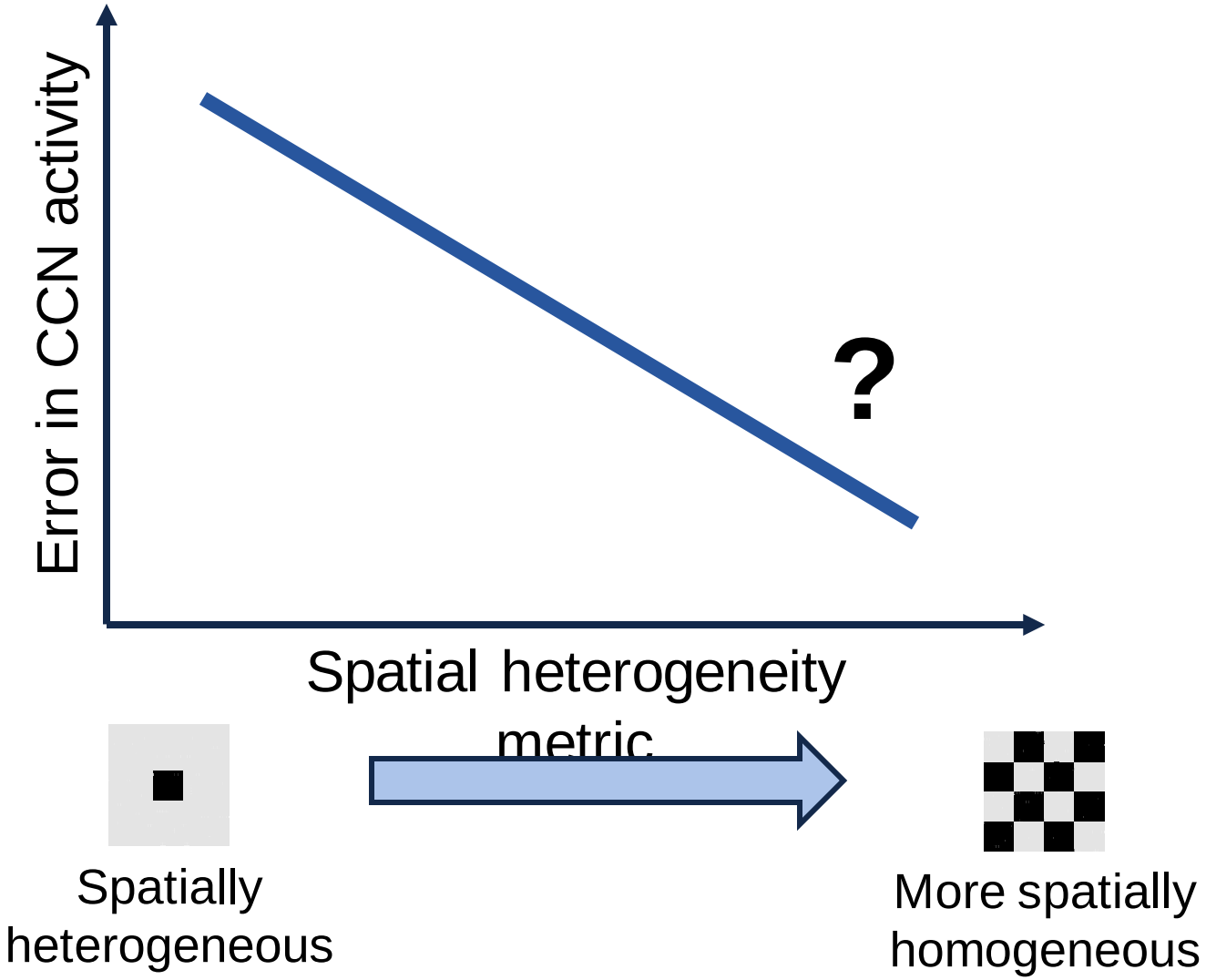
Convective and turbulent transport of aerosols and gases

Emissions are heterogeneous

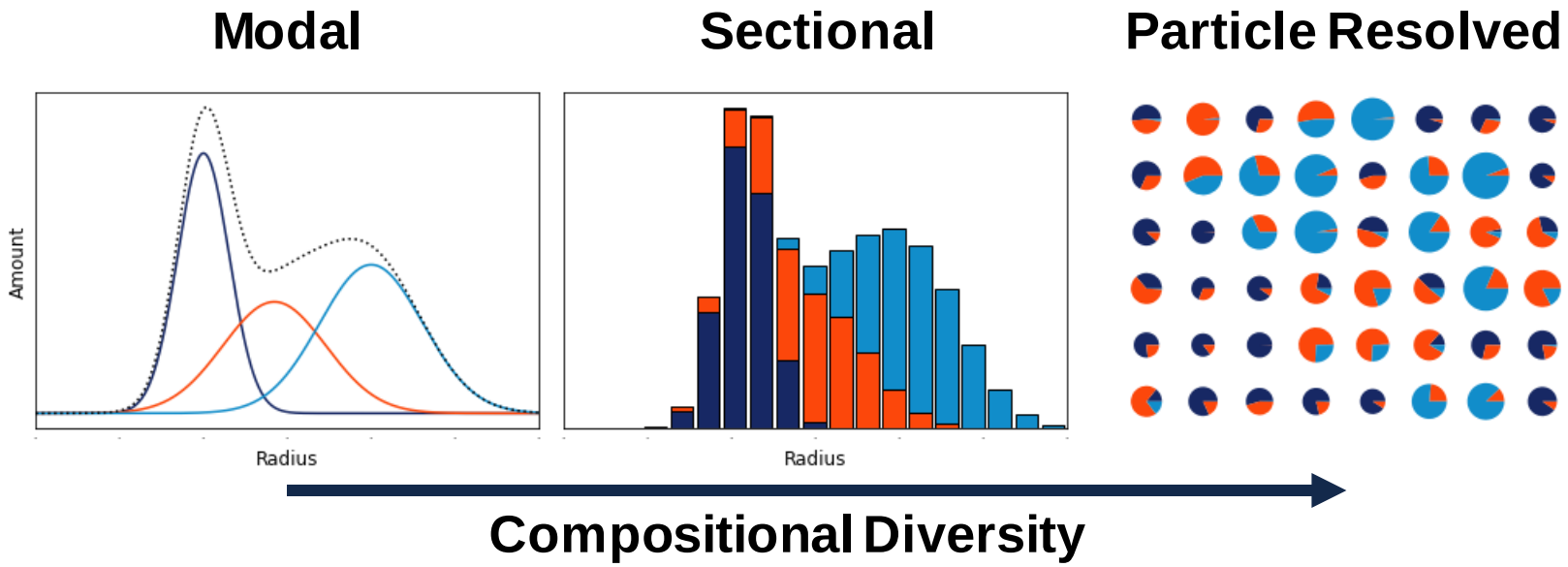
Emissions are assumed **uniformly distributed** across grid cells



How does CCN activity vary with spatial heterogeneity?



Particle-Resolved Aerosol Modeling



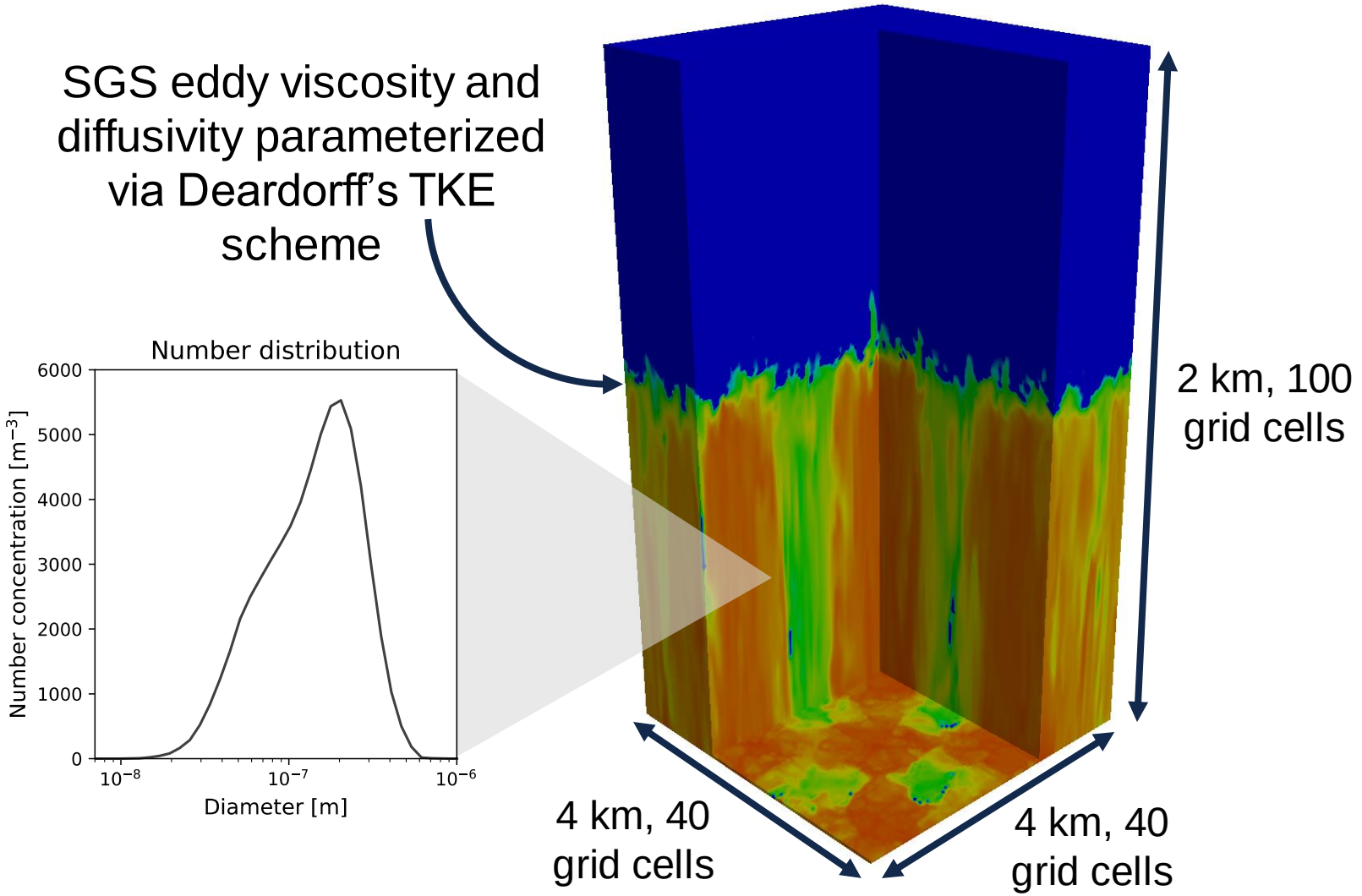
Particle-resolved models allow **simulation of per-particle aging** (highly dependent on the properties of the aerosol!)

Simulation Setup

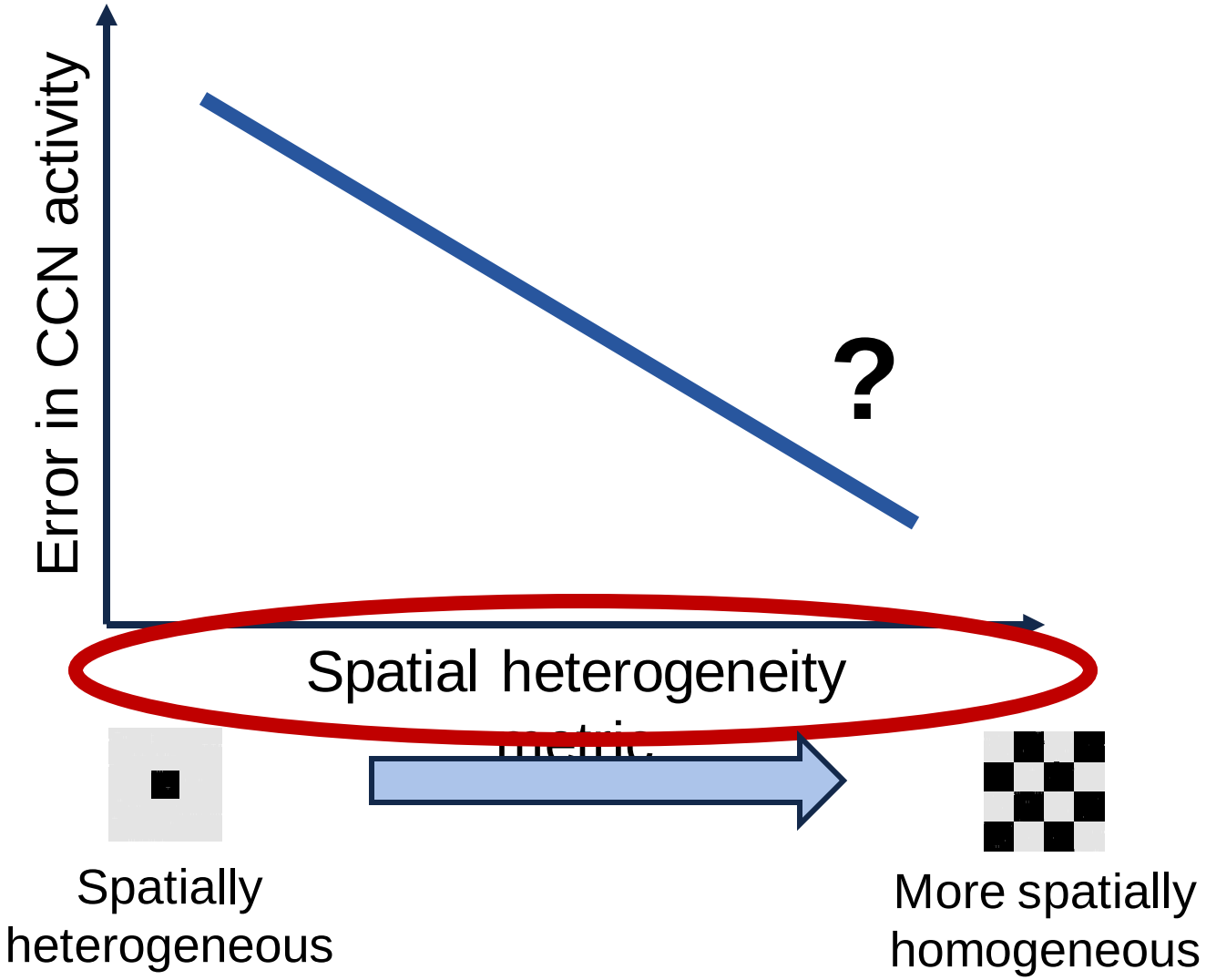
Use coupled **WRF-PartMC** for particle-resolved LES simulations

Multiphase chemistry using **MOSAIC**

Gas, aerosol initial conditions and emissions via Riemer et al. 2009

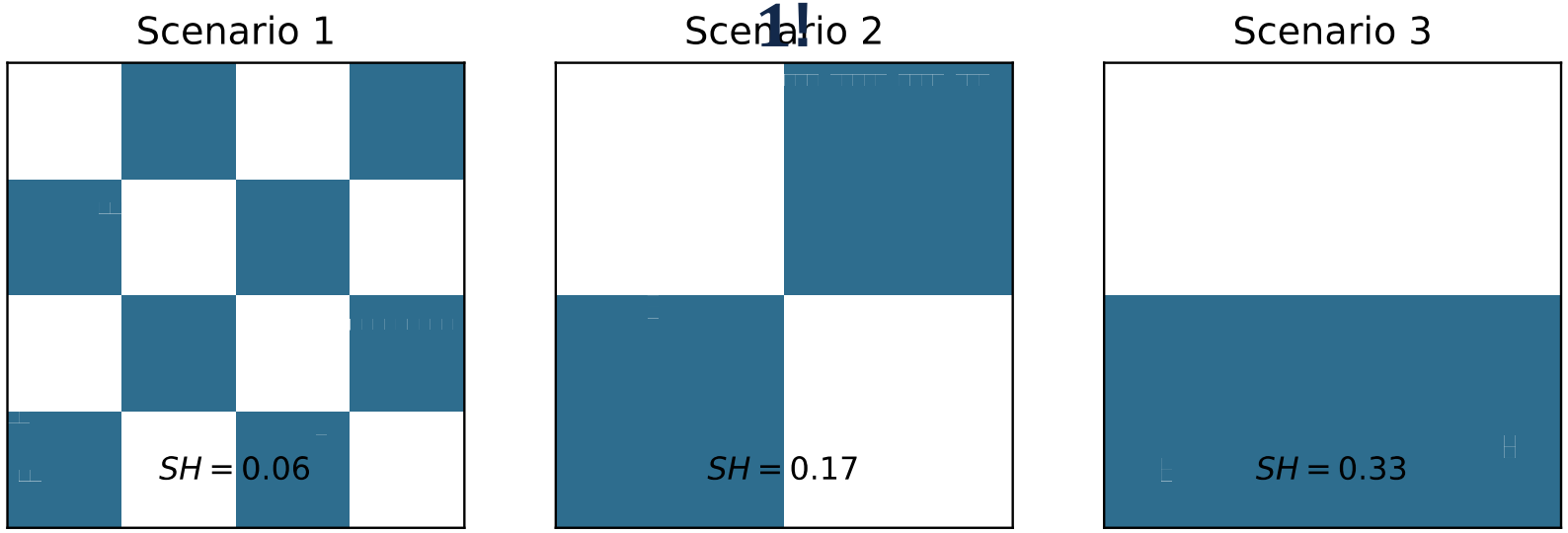


We must quantify spatial heterogeneity to measure its effect



Variance is an imperfect measure of heterogeneity

These scenarios have the same variance, $\sigma^2 = 1$!



...

Spatial Heterogeneity Metric

Normalized measure of **how variance of a quantity is arranged** over a domain

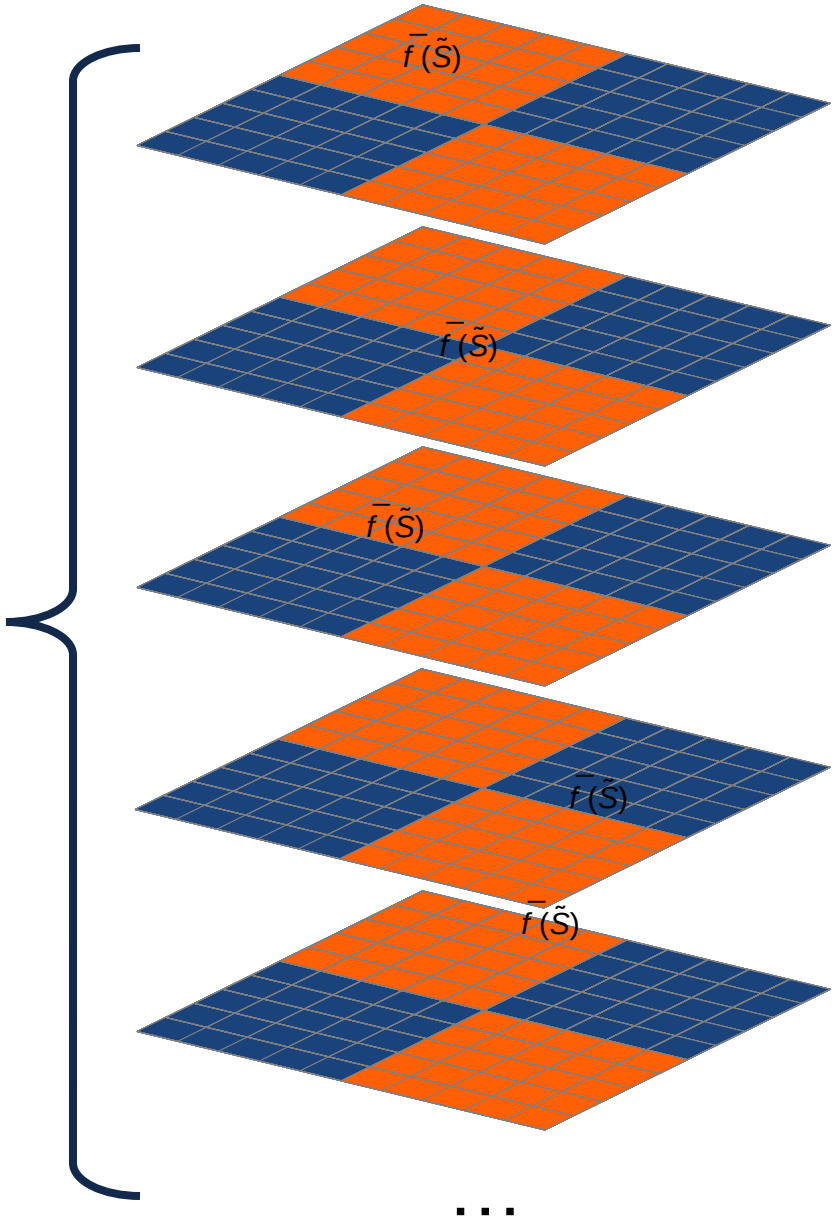
$$SH(f,S) = \frac{1}{N} \sum_{\tilde{S} \in \mathbf{R}} |\bar{f}(S) - \bar{f}(\tilde{S})|$$

N subsets

Mean over entire domain

Mean of subarray

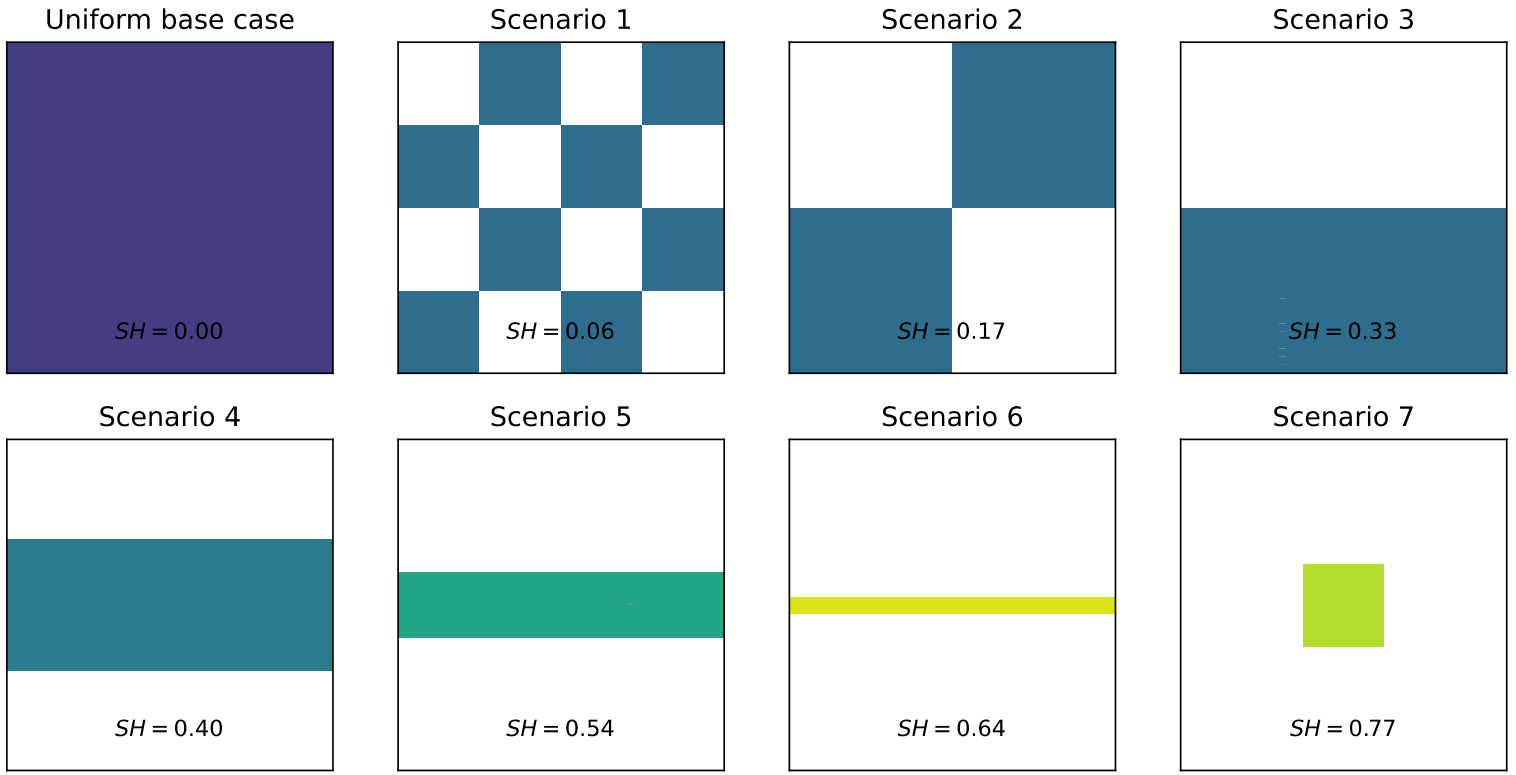
For efficiency, a Monte Carlo implementation is used to estimate SH.



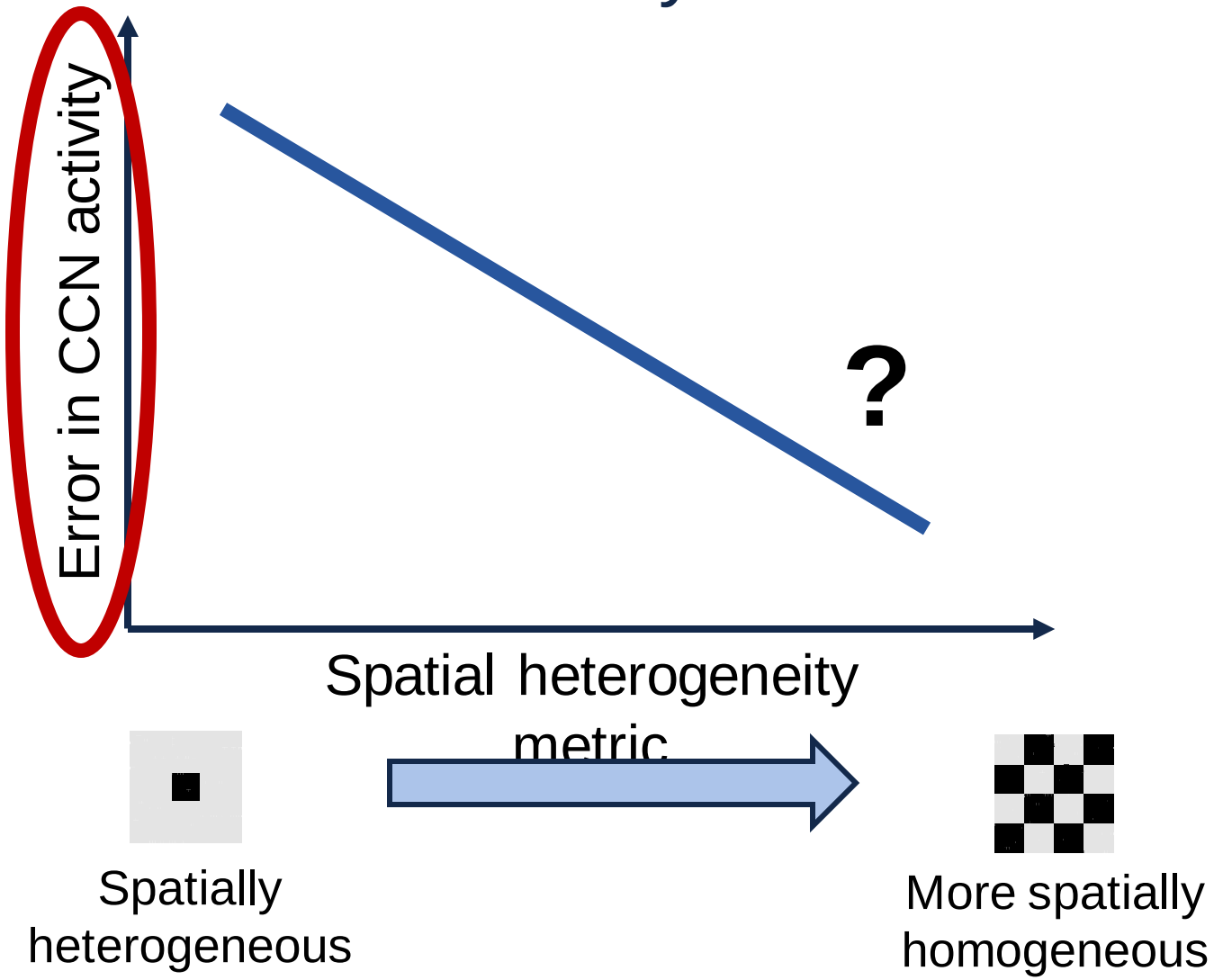
How are the emissions distributed?

Emissions spatial distribution varied using idealized patterns with increasing heterogeneity

Spatial heterogeneity scenarios **compared against the uniform base case**

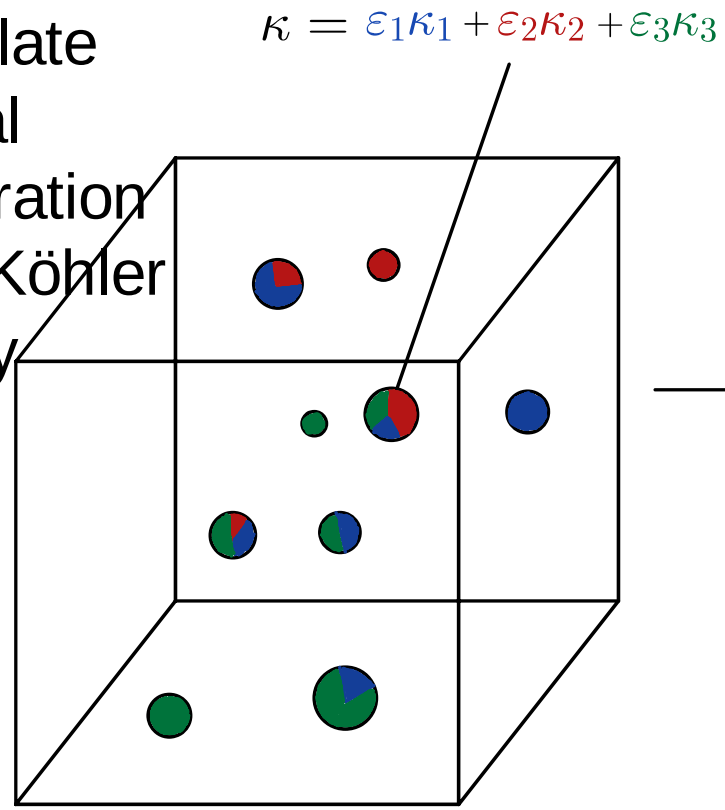


Measuring Error in CCN Activity

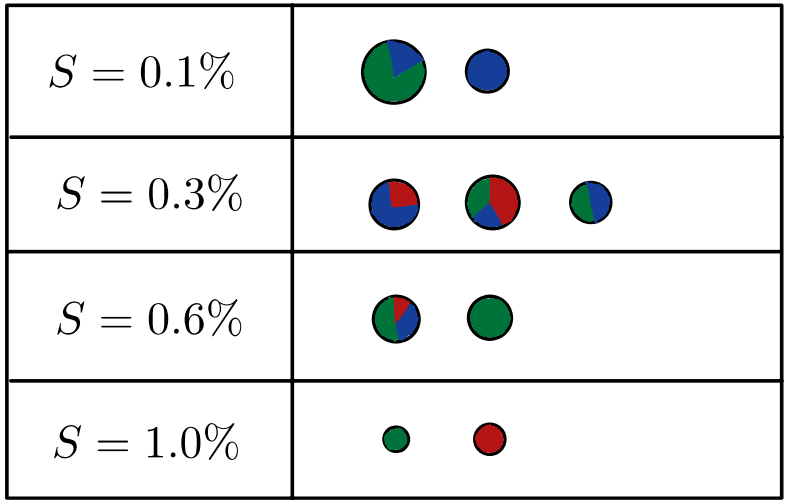


Measuring Error in CCN Activity (for each emissions scenario)

1. Calculate critical supersaturation (S) with κ-Köhler theory



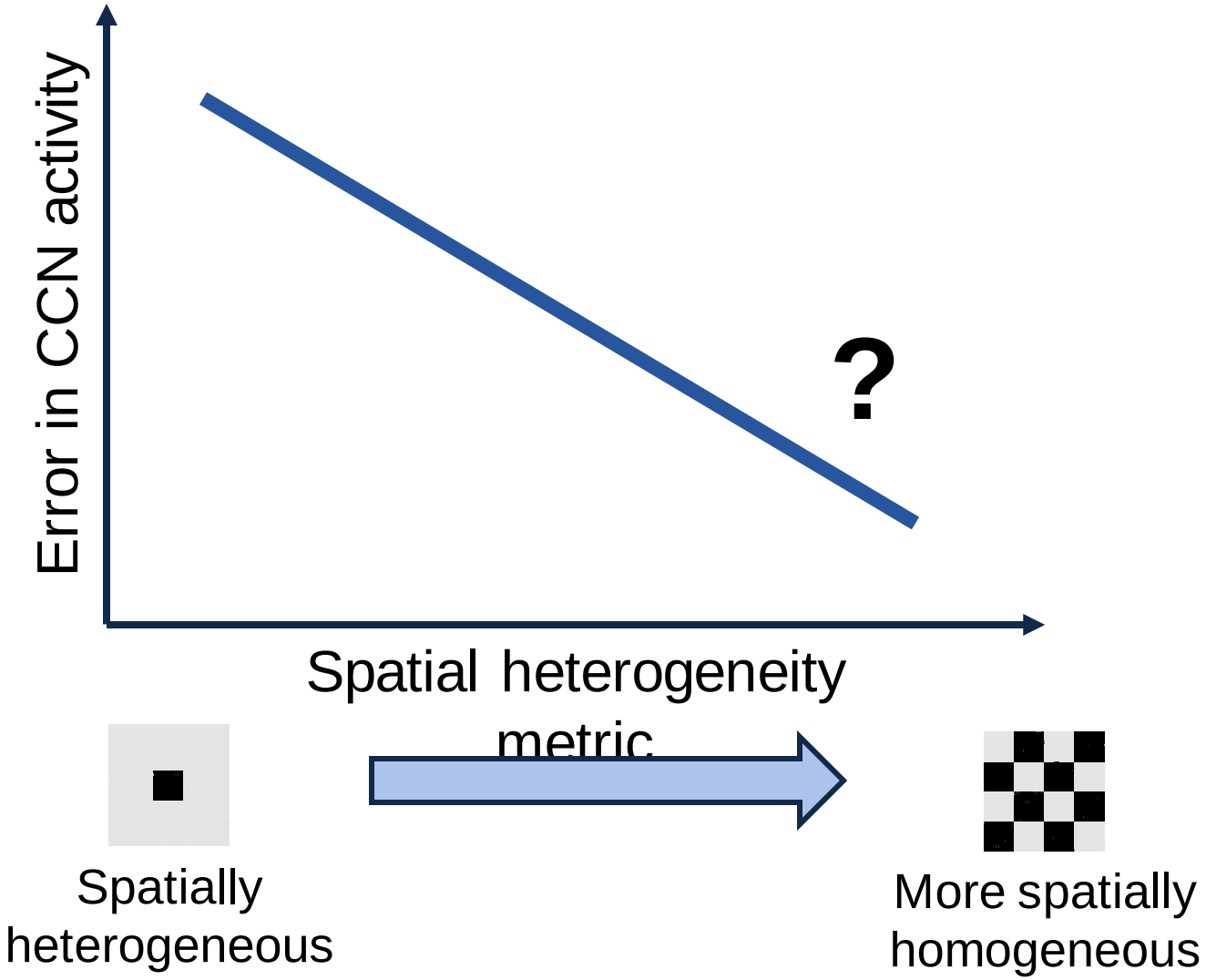
2. Group particles by S and calculate CCN concentration



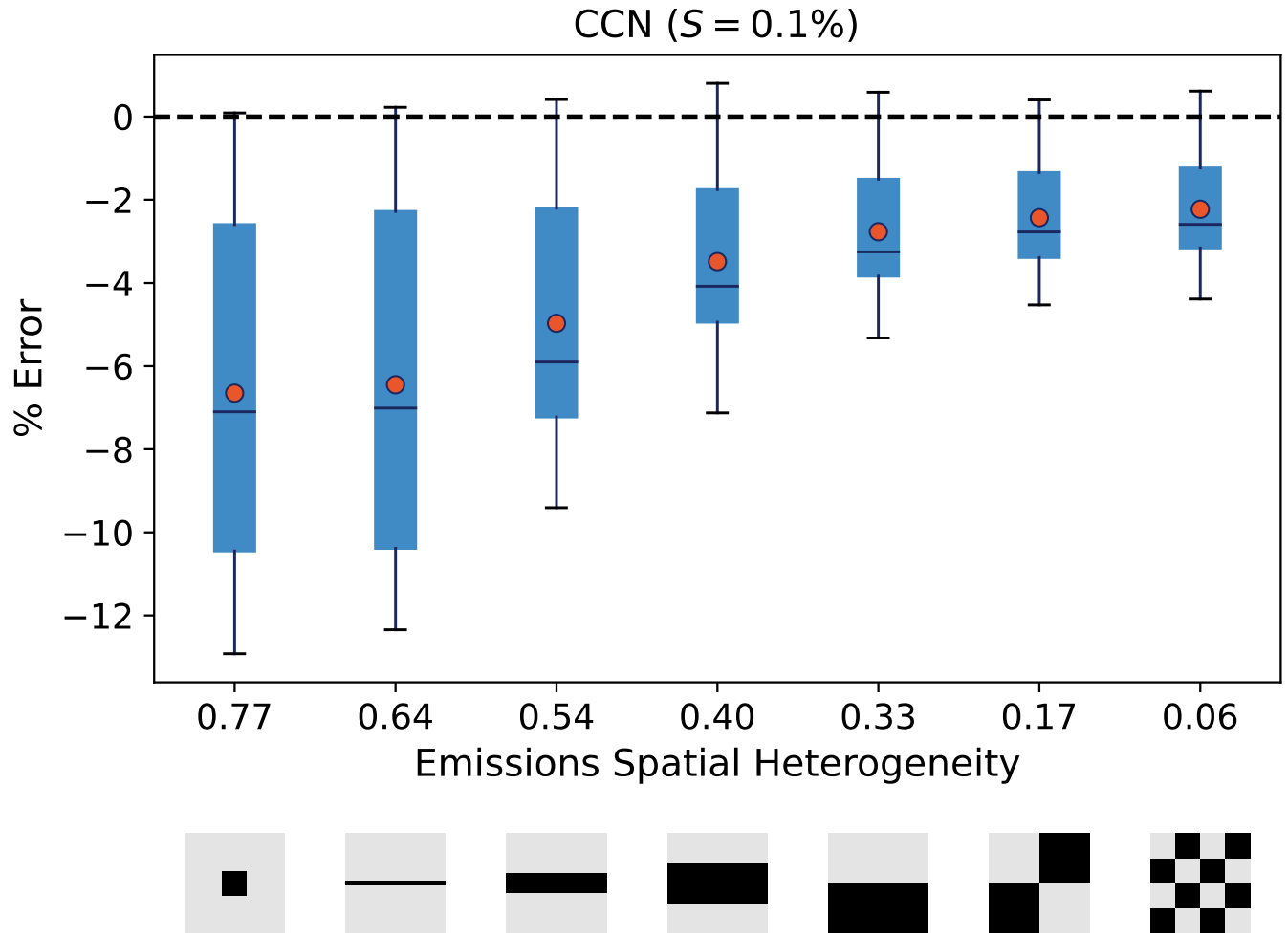
3. Compute average CCN conc. for each vertical level and time step

4. Calculate % error relative to base case

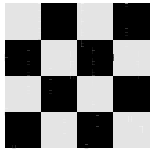
How does CCN activity vary with spatial heterogeneity?



CCN activating at low S are most sensitive to changes in emissions heterogeneity

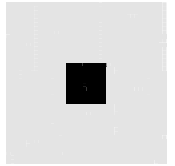


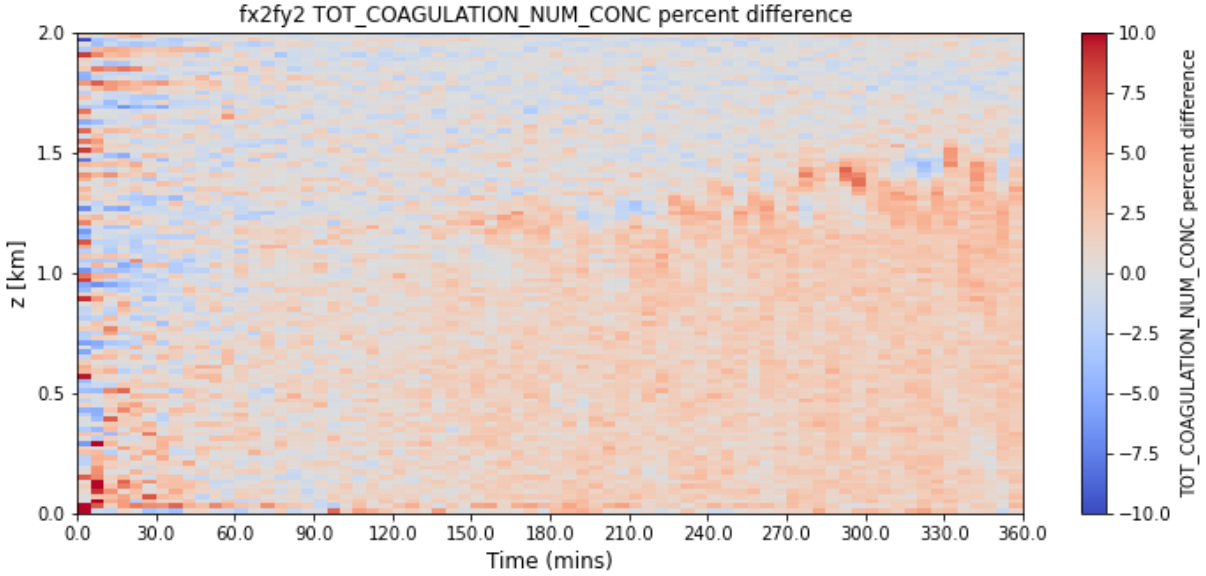
Coagulation increases as spatial heterogeneity increases

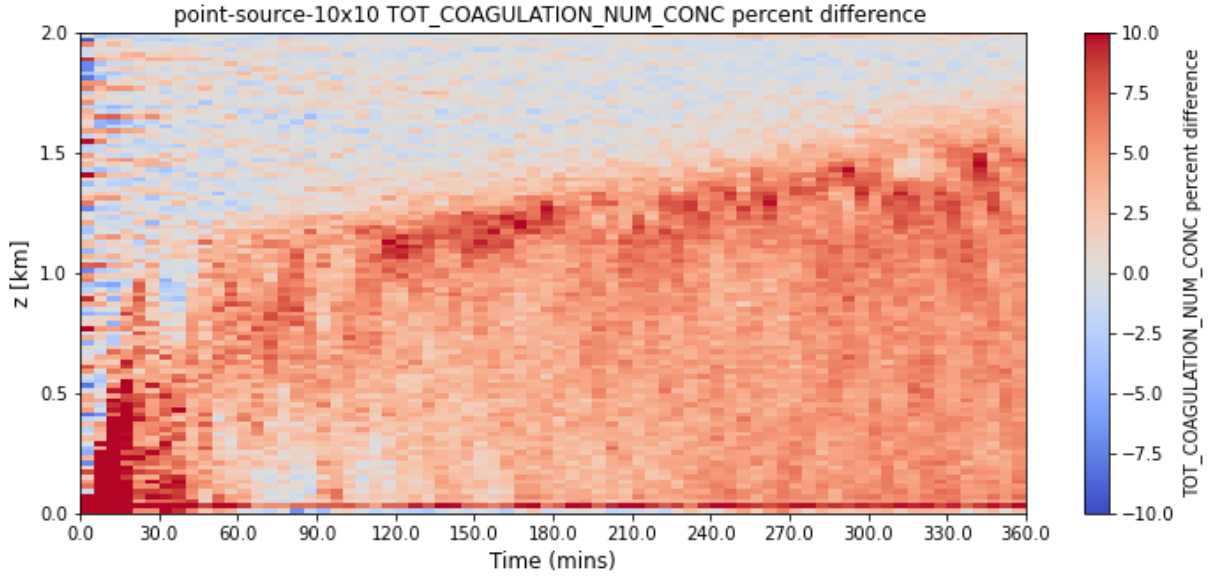


Scenario 1
SH = 0.06

Scenario 7
SH = 0.77



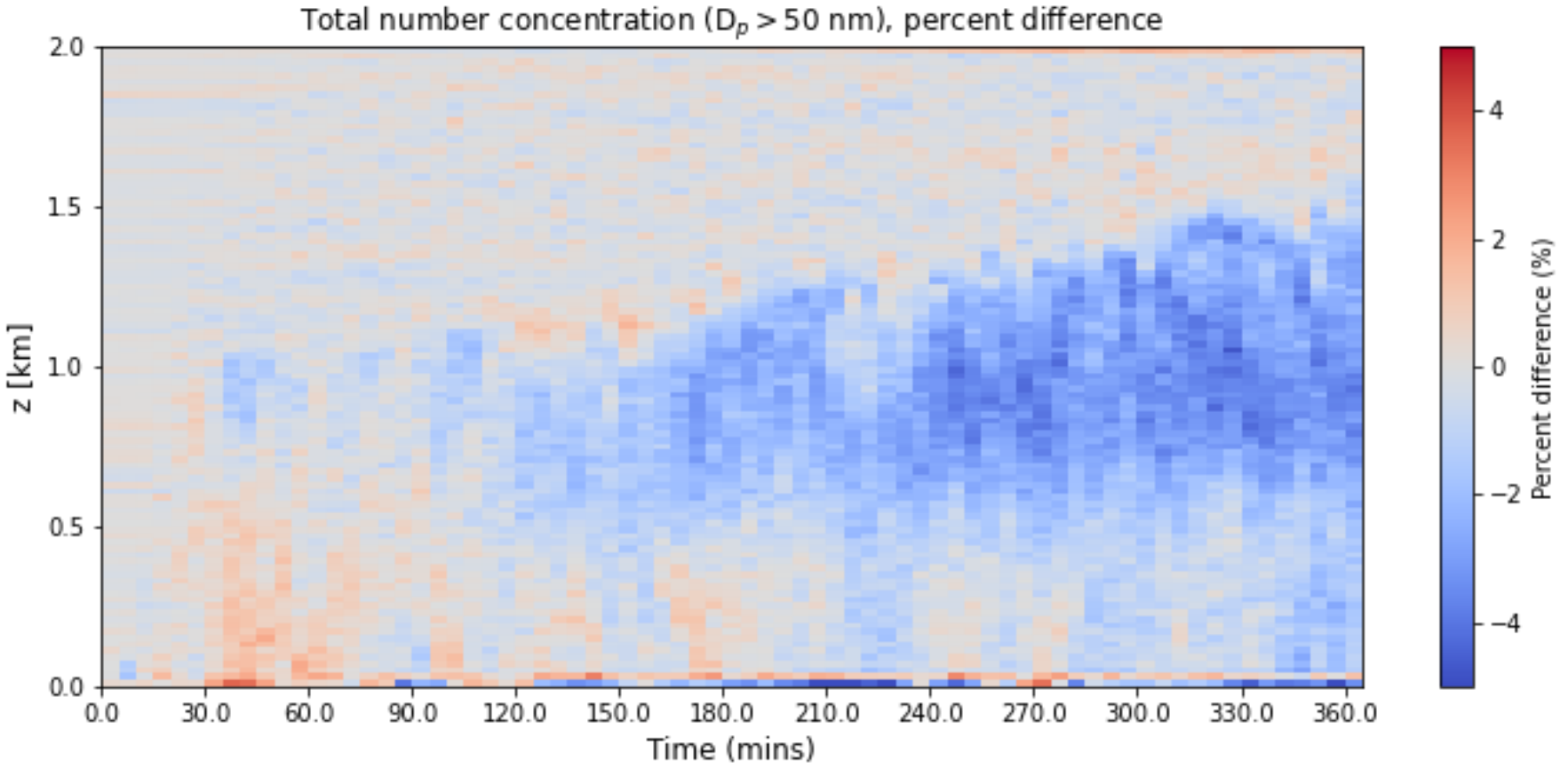




Coagulation reduces the number of particles that activate as CCN

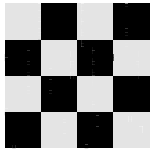
Scenario 7

SH = 0.77

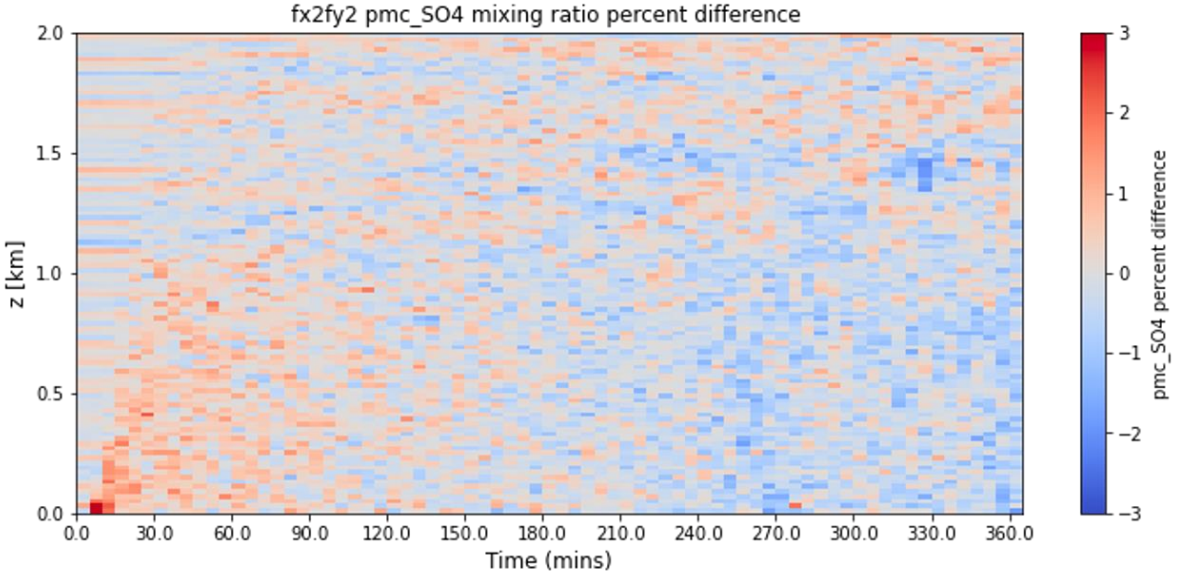


What are the impacts of spatial heterogeneity on chemistry?

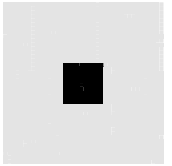
Sulfate

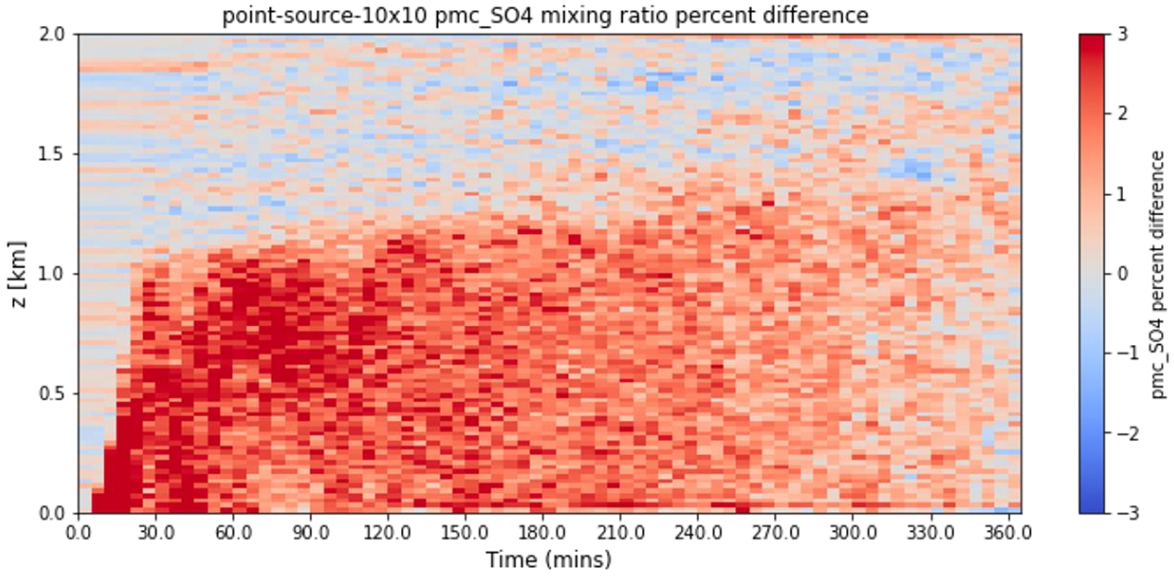


Scenario 1
SH = 0.06



Scenario 7
SH = 0.77



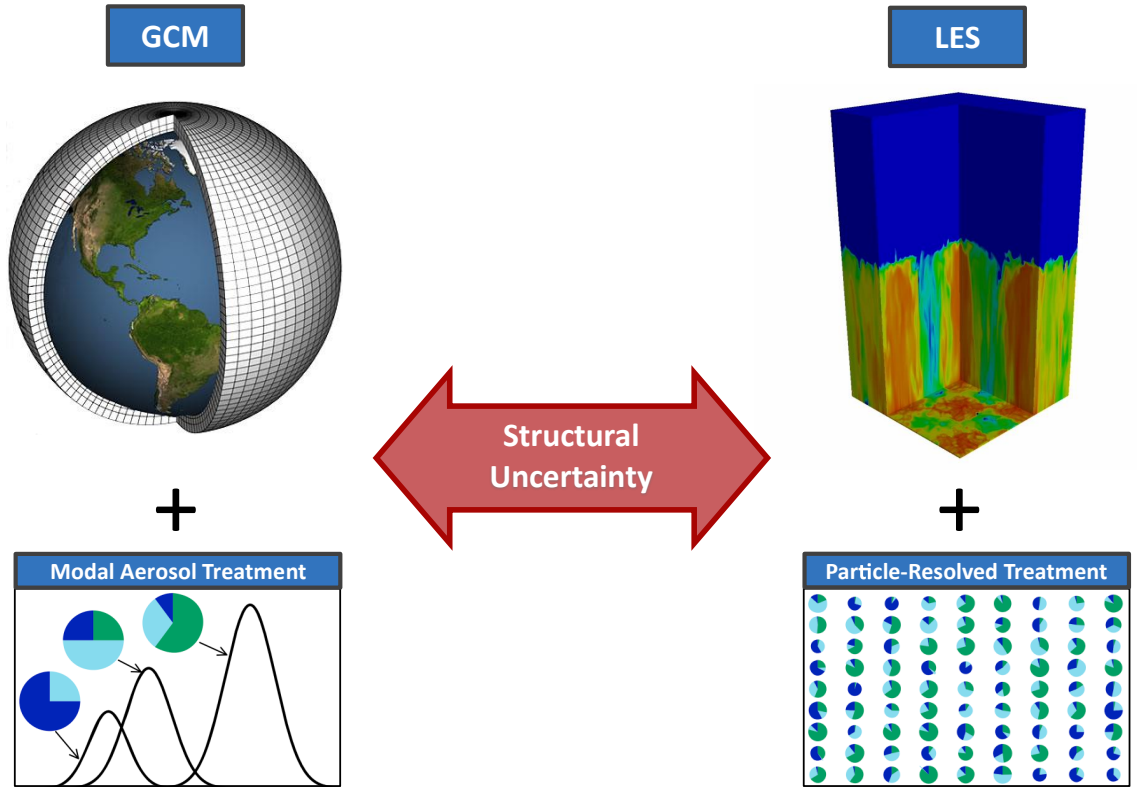


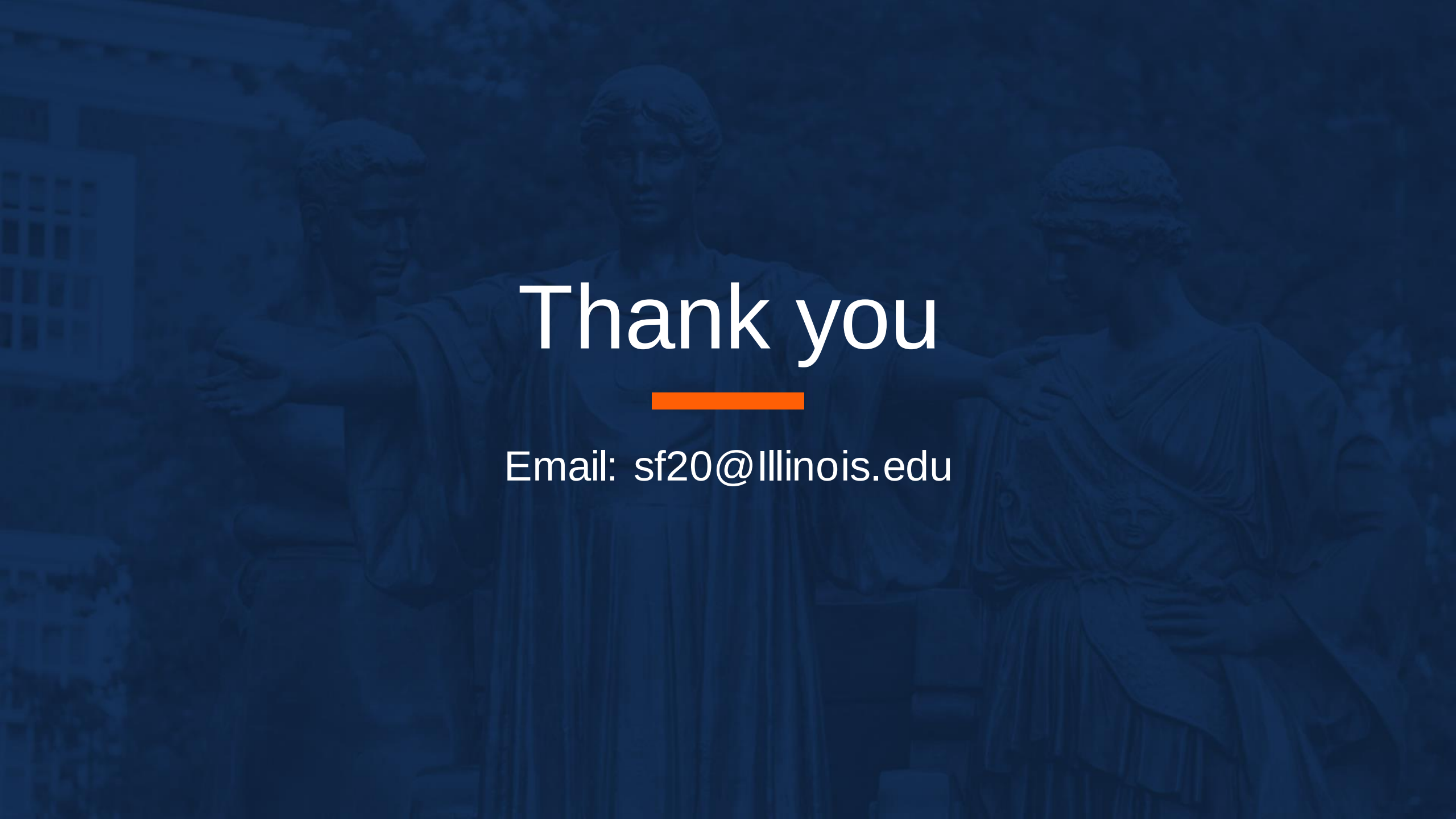
Key takeaways

CCN activity tends to **decrease** with increasing spatial heterogeneity

Coagulation increases with SH, likely reducing the concentration of CCN

Regional and global models that assume uniform emissions **may overestimate CCN activity** in geographic regions with high emissions heterogeneity





Thank you

Email: sf20@Illinois.edu