Modeling the seed-dependent particle growth via multiphase reactions with the particle-resolved model PartMC-CAMP

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Growth rates of particles depend on the seed

http://pubs.acs.org/journal/aesccq Article

Growth Rate Dependence of Secondary Organic Aerosol on Seed Particle Size, Composition, and Phase

Devon N. Higgins, Michael S. Taylor, Jr., Justin M. Krasnomowitz, and Murray V. Johnston*

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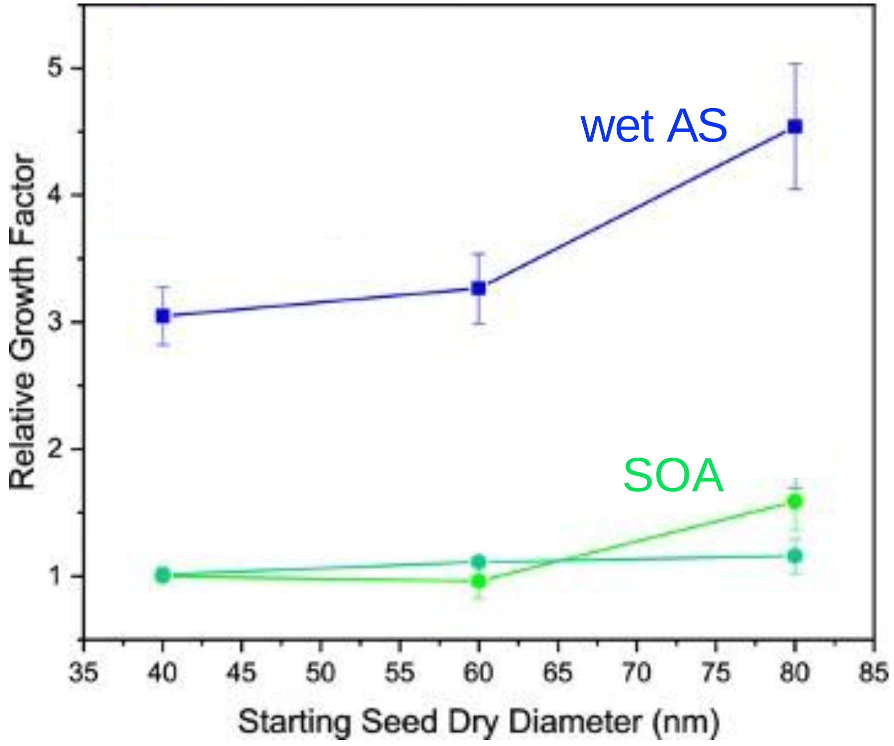
- What is the impact on size distributions and CCN activity?
- **Two steps:**
  Step 1: Develop mechanistic model
  Step 2: Perform population-level simulations



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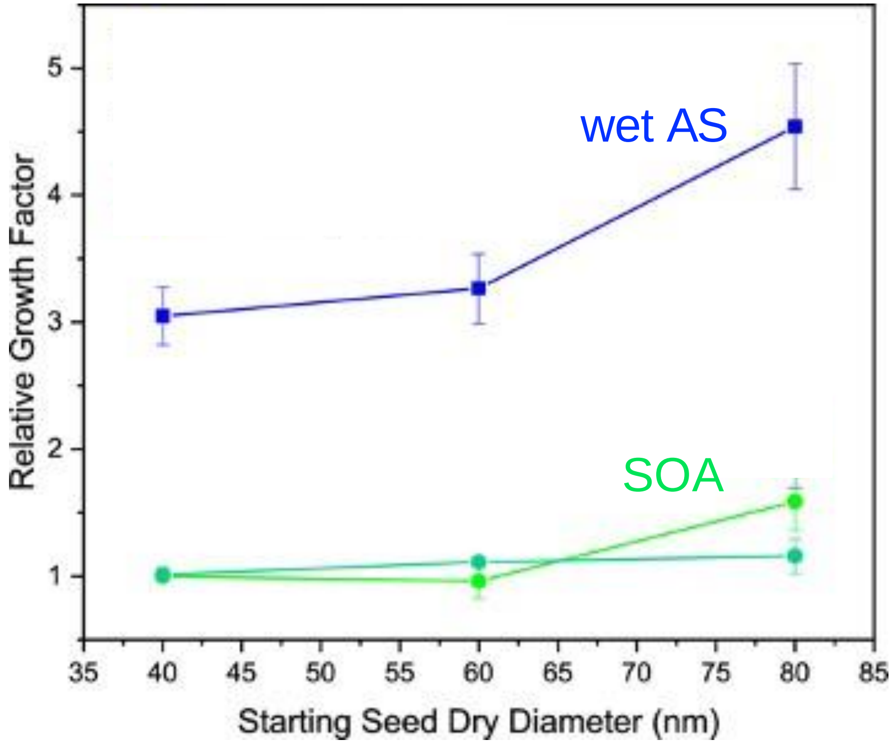

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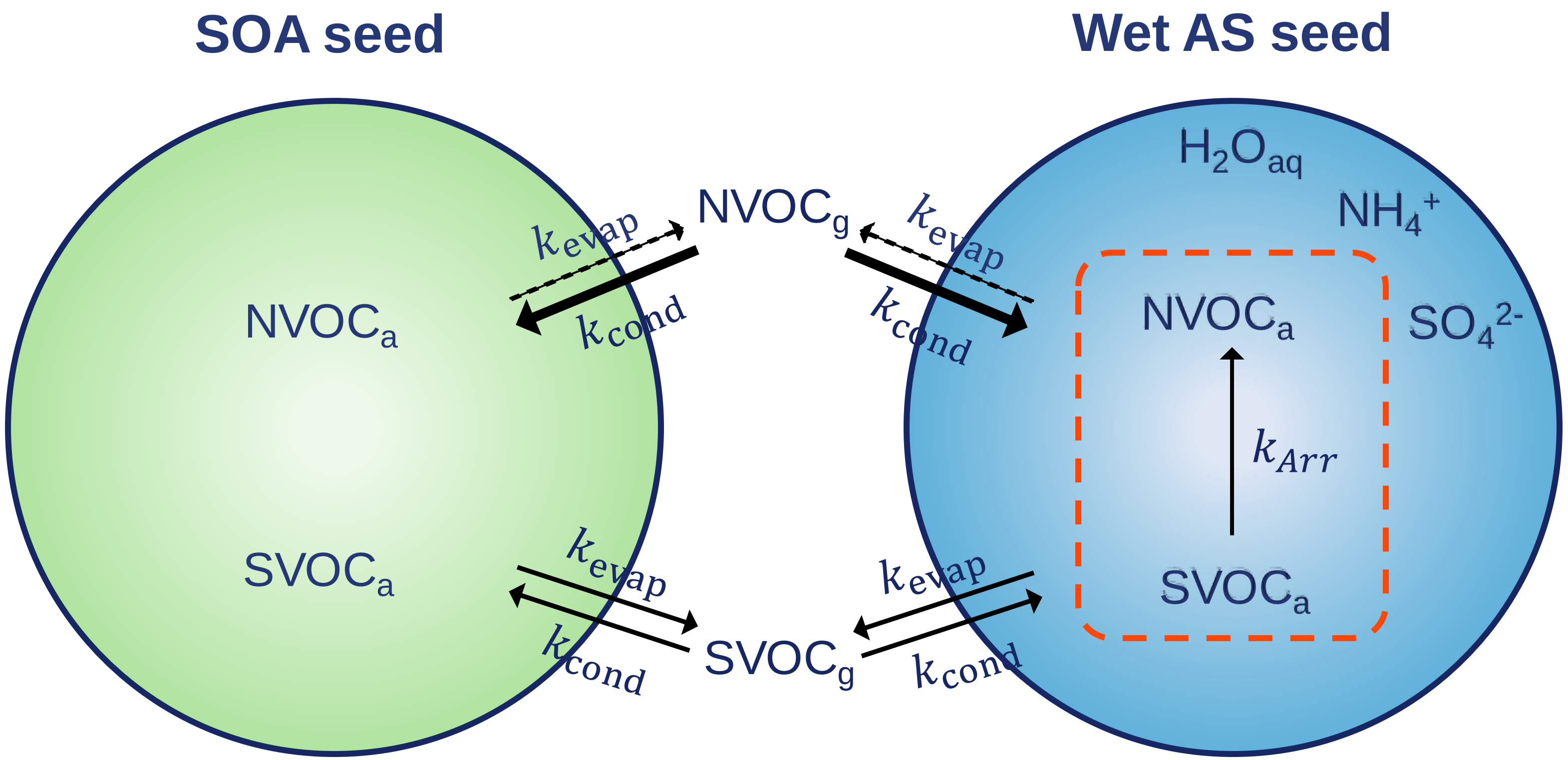
Construct inorganic/organic multiphase system in model



- **SOA seed**

$$NVOC_g \underset{k_{evap}}{\overset{k_{cond}}{\rightleftharpoons}} NVOC_a$$

$$SVOC_g \underset{k_{evap}}{\overset{k_{cond}}{\rightleftharpoons}} SVOC_a$$

- **Wet AS seed**

$$NVOC_g \underset{k_{evap}}{\overset{k_{cond}}{\rightleftharpoons}} NVOC_a$$

$$SVOC_g \underset{k_{evap}}{\overset{k_{cond}}{\rightleftharpoons}} SVOC_a$$

$$SVOC_a \xrightarrow{k_{Arr}} NVOC_a$$

Our tools: integrated multiphase chemistry model (CAMP)

- JSON-based: allows for detailed description of any gas- and particle- phase chemical mechanism.

```
"reactions" : [
    {
      "type" : "SIMPOL_PHASE_TRANSFER",
      "gas-phase species" : "SVOC",
      "aerosol phase" : "organic_seed",
      "aerosol-phase species" : "SVOC_aero",
      "B" : [ 3.50e3, -2.13e1, 0.0, 0.0 ]
    },
              ...
    {
      "type" : "CONDENSED_PHASE_ARRHENIUS",
      "aerosol phase" : "inorganic_seed",
      "units" : "mol m-3",
      "reactants" : {
        "SVOC_aero" : {}
      },
      "products" : {
        "NVOC_aero" : {}
      },
      "A" : 3.65e-6,
      "B" : 1e-10,
      "C" : 1426.6
    },
              ...
```

Configuration for SVOC partitioning to the organic seed

Reminder:

$$SVOC_g \underset{k_{evap}}{\overset{k_{cond}}{\rightleftharpoons}} SVOC_a$$

Configuration for in-particle reaction within inorganic seed

Reminder:

$$SVOC_a \xrightarrow{k_{Arr}} NVOC_a$$

M. L. Dawson, et al., *Geosci. Model Dev.*, 15, 3663–3689, 2022

Our tools: Integrated multiphase chemistry model (CAMP)

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        "NVOC_aero" : {}
      },
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    },
              ...
```

$$p_{eq}(T) = 101325.0 \times 10^{b(T)}$$

$$b(T) = \frac{B_1}{T} + B_2 + B_3 T + B_4 \ln T$$

Reminder:

$$SVOC_g \underset{k_{evap}}{\overset{k_{cond}}{\rightleftharpoons}} SVOC_a$$

$$k_{Arr} = Ae^{\left(-\frac{E_a}{k_b T}\right)}\left(\frac{T}{D}\right)^B = Ae^{\left(\frac{C}{T}\right)}\left(\frac{T}{D}\right)^B$$

Reminder:

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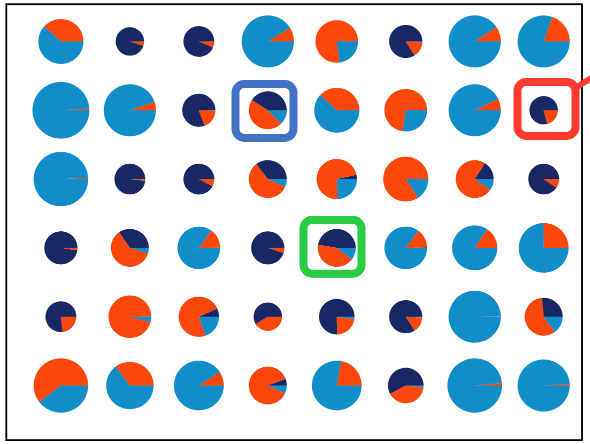
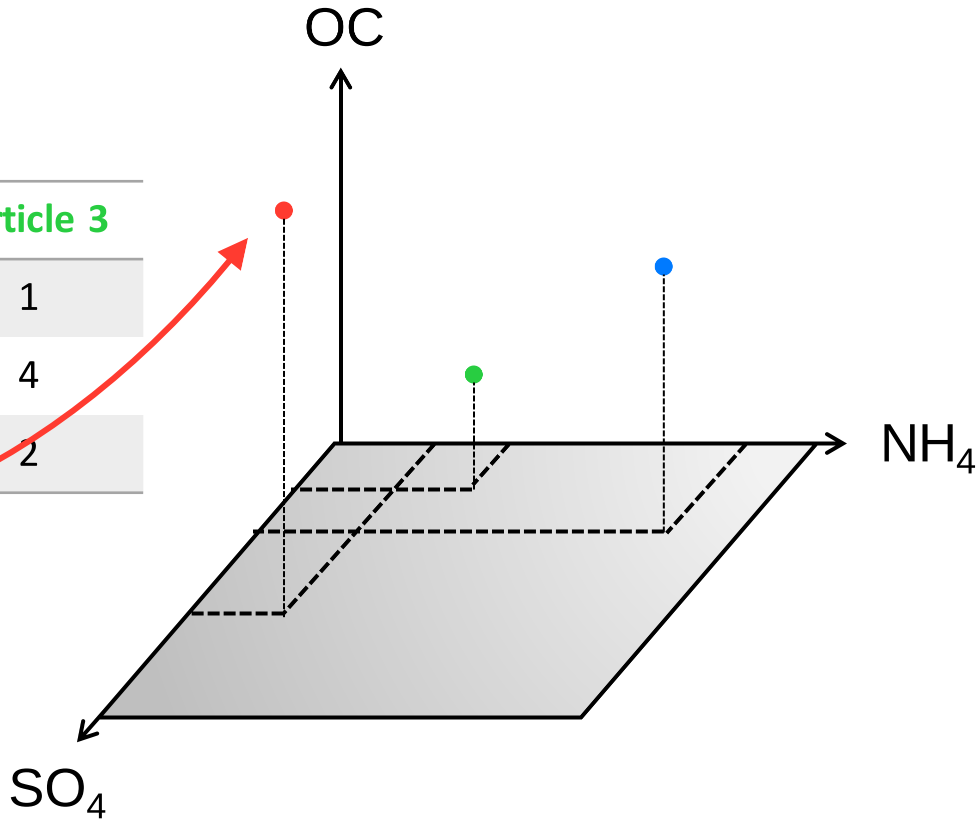
Our tools: Particle-resolved model (PartMC & PyPartMC)

- Each particle is uniquely represented as an *A*-dimensional vector with mass composition components $\{\mu_1^i, \mu_2^i, \ldots, \mu_A^i\}$.
- Allows for composition-dependent growth rates.



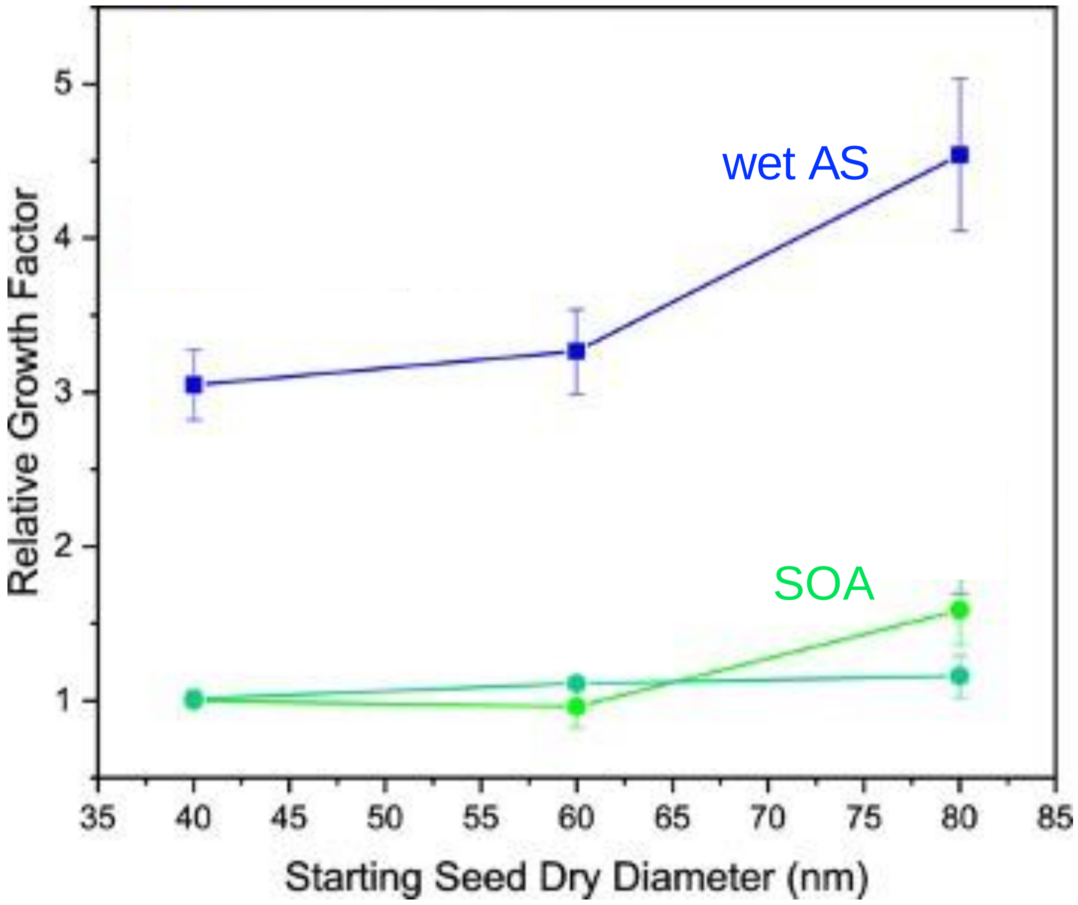
	Particle 1	Particle 2	Particle 3
NH_4	3	10	1
SO_4	12	3	4
OC	5	8	2



PartMC

N. Riemer, M. West, R. A. Zaveri, and R. C. Easter, *Journal of Geophysical Research*, 2009

How to incorporate experimental results into our model?



How can the model be calibrated to translate GF into condensed-phase reaction constant ?

- **Wet AS seeds** – GF ≈ 3
- **SOA seeds** – GF ≈ 1

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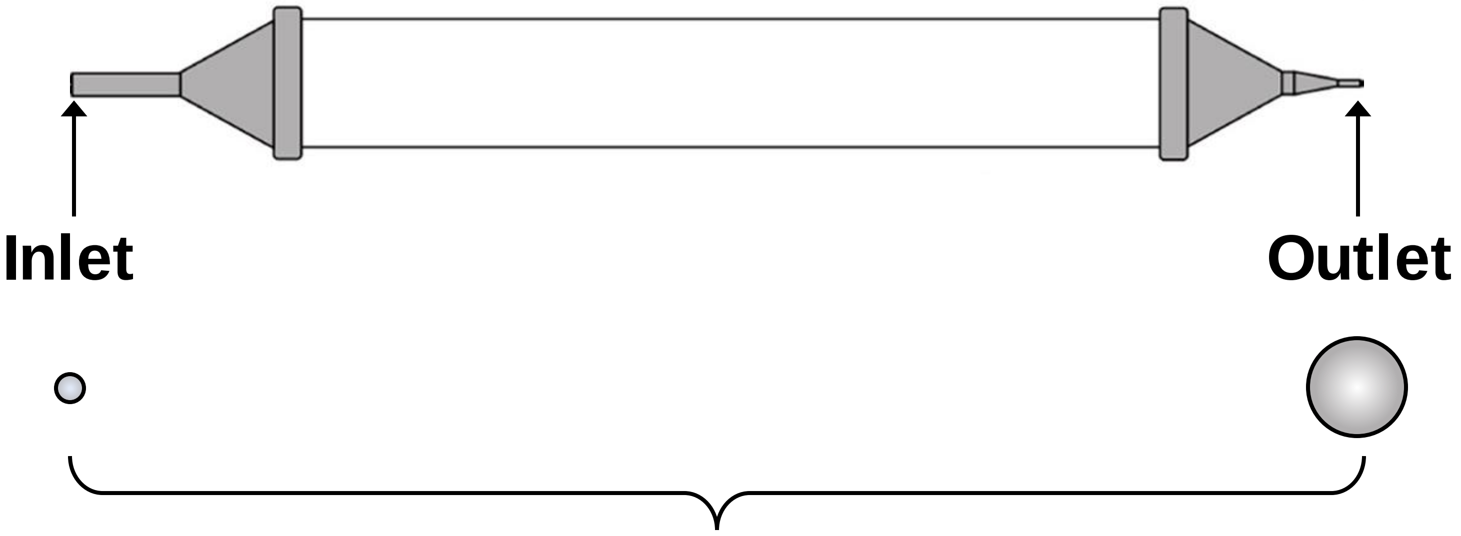
How to incorporate experimental results into our model?

- First, we need to understand the meaning of “growth factor”



$$\Delta D_p = D_p^{outlet} - D_p^{inlet}$$

Growth factor GF:

Fit parameter that tells us the factor by which we would need to increase the gas phase concentration to obtain the same growth **without having the condensed-phase reaction occurring**.

How to incorporate experimental results into our model?

- Second, we need to design cases to mimic the experiment.

Cases	Seed	$NVOC_g(t=0)$ (ppb)	$SVOC_g(t=0)$ (ppb)	Gas/particle Partitioning	Condensed-phase Reaction
base	AS	0.04	0.132	✓	✗
base3x	AS	0.12	0.132	✓	✗
enhc	AS	0.04	0.132	✓	✓

- "base" – GF = 1
- "base3x" – GF = 3
- "enhc" – what an actual wet AS seed growth should look like

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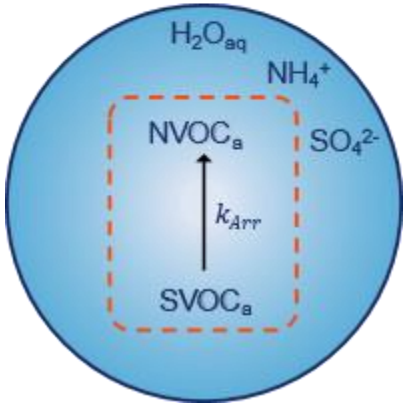
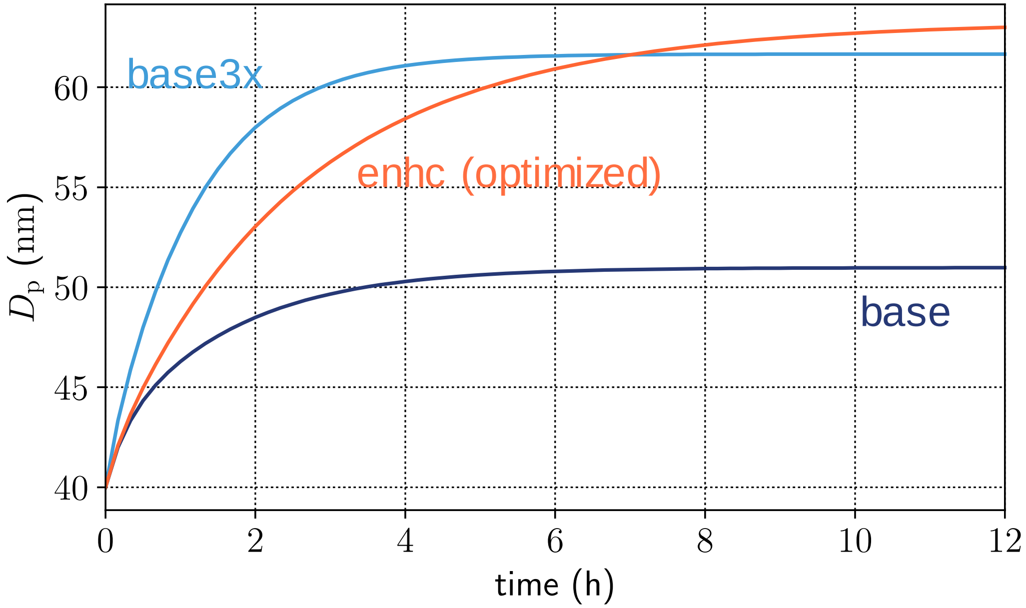


Use Python SciPy to obtain optimized parameters (A, B, C)



Optimized set:

A = 3.65×10^{-6} s^{-1}

B = 10^{-10}

C = 1426.6 K

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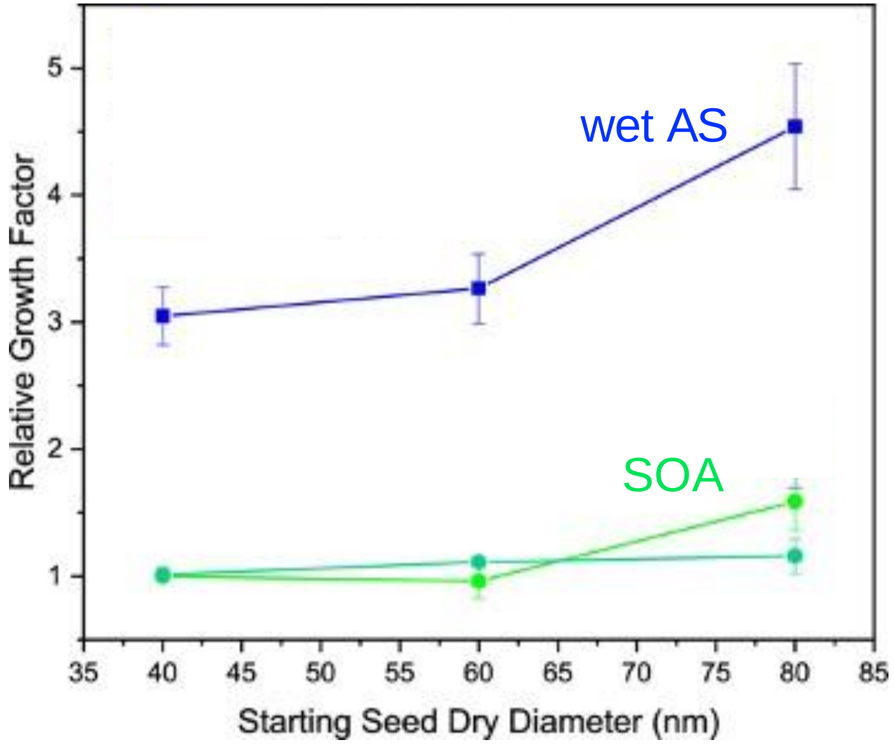

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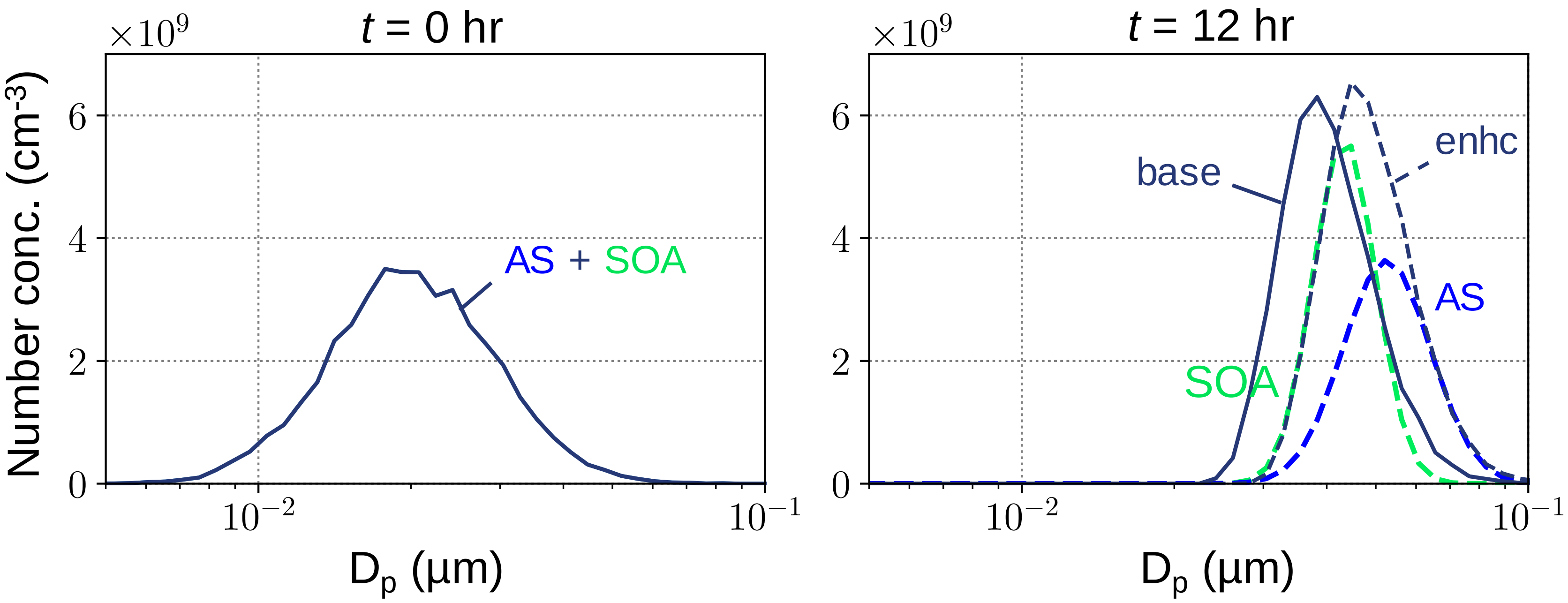
Case A: Unimodal distribution + competing for gases

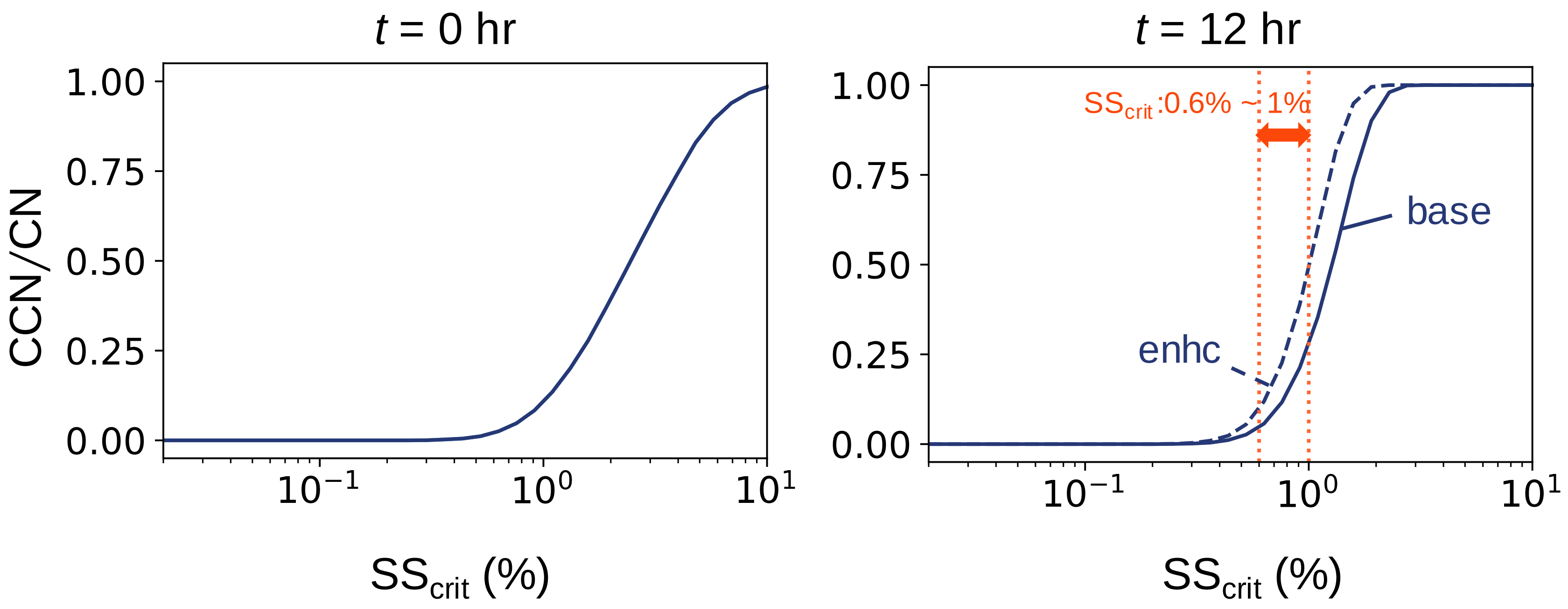
- **Two** independent simulations: base vs. enhc.
- SOA and AS seeds **compete** for NVOC and SVOC.
- Enhanced SOA formation shifts the **size distribution of AS** towards larger sizes.



Increases median diameter by approximately 21%.

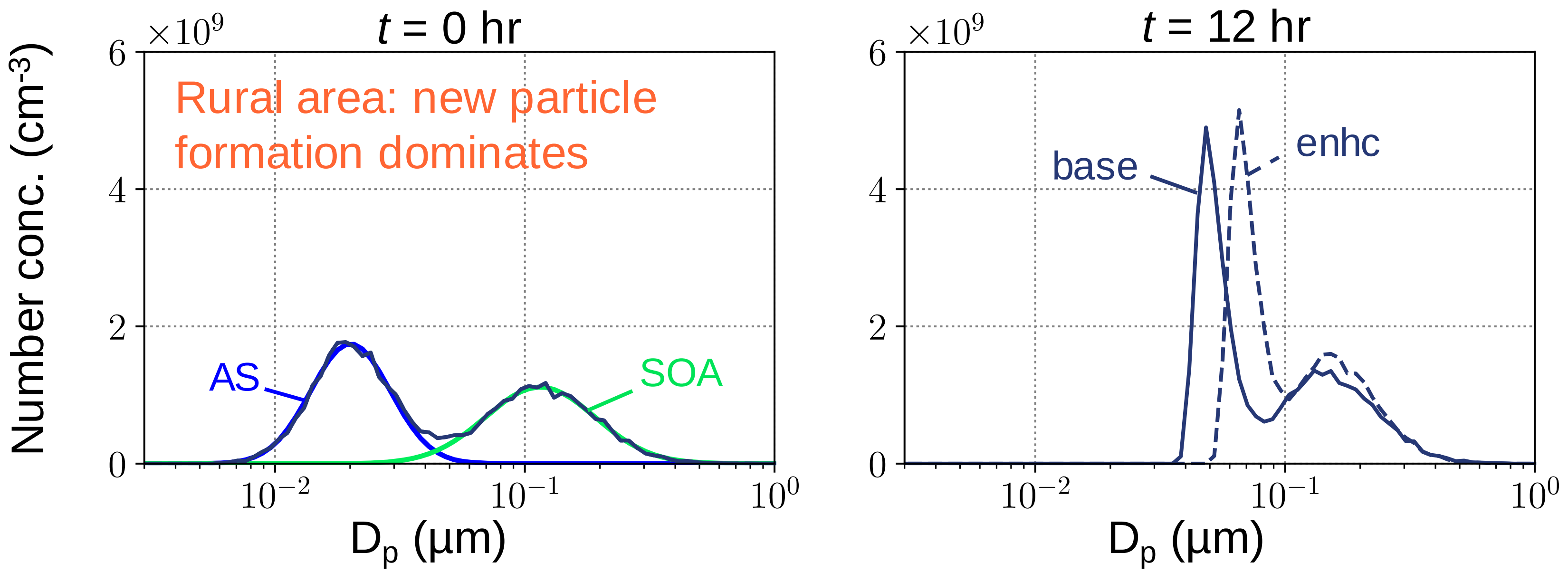
Case A: Unimodal distribution + competing for gases

- **Two** independent simulations : base vs. enhc.
- SOA and AS seeds **compete** for NVOC and SVOC.
- Increase in particle sizes affects **CCN activity**.



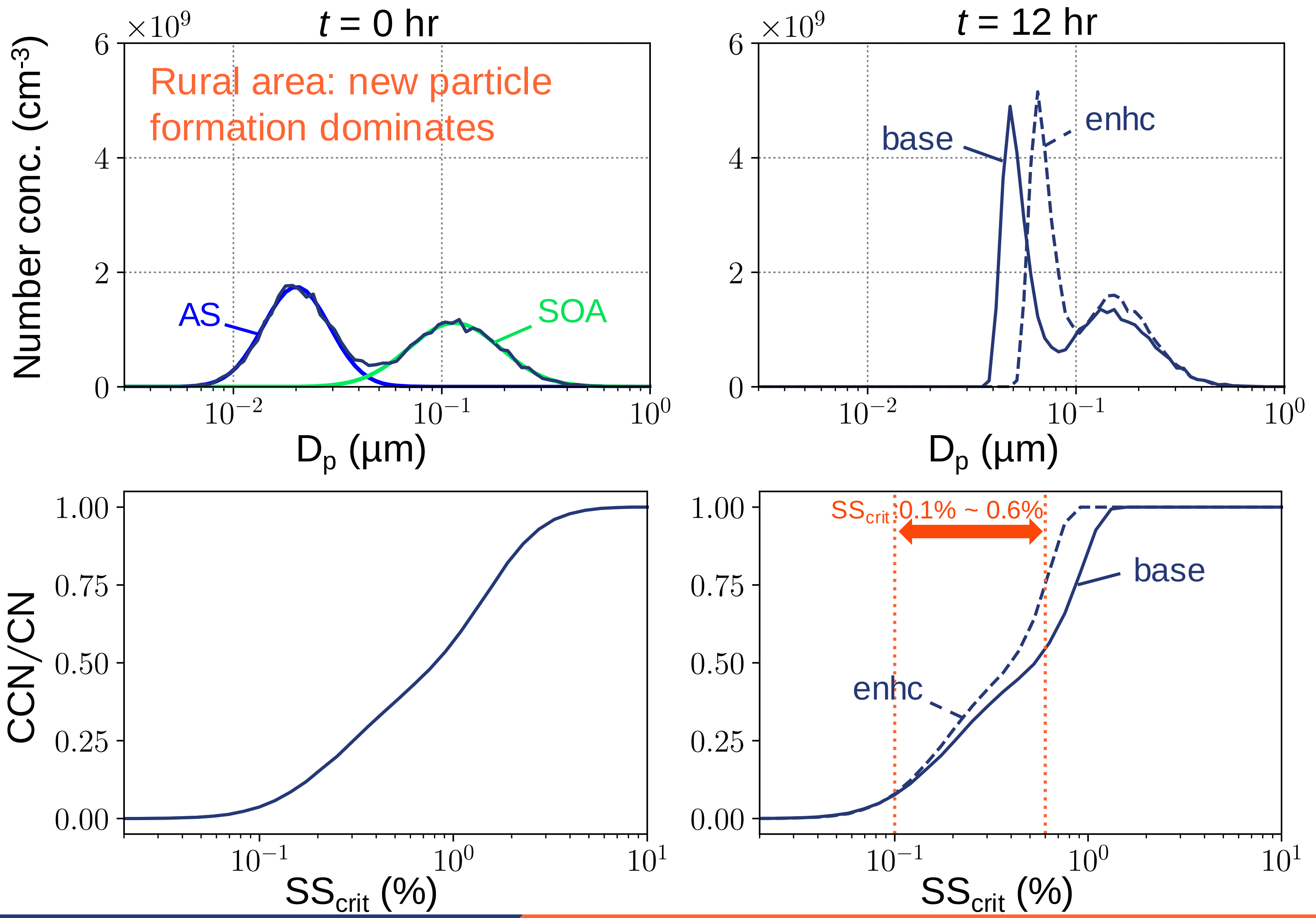
Underestimation in CCN/CN is approximately 52% (SS_{crit} = 0.6%), 43% (SS_{crit} = 1%).

Case B: Bimodal distribution + AS in the small mode



Reduces the distance between the peaks by approximately 36%.

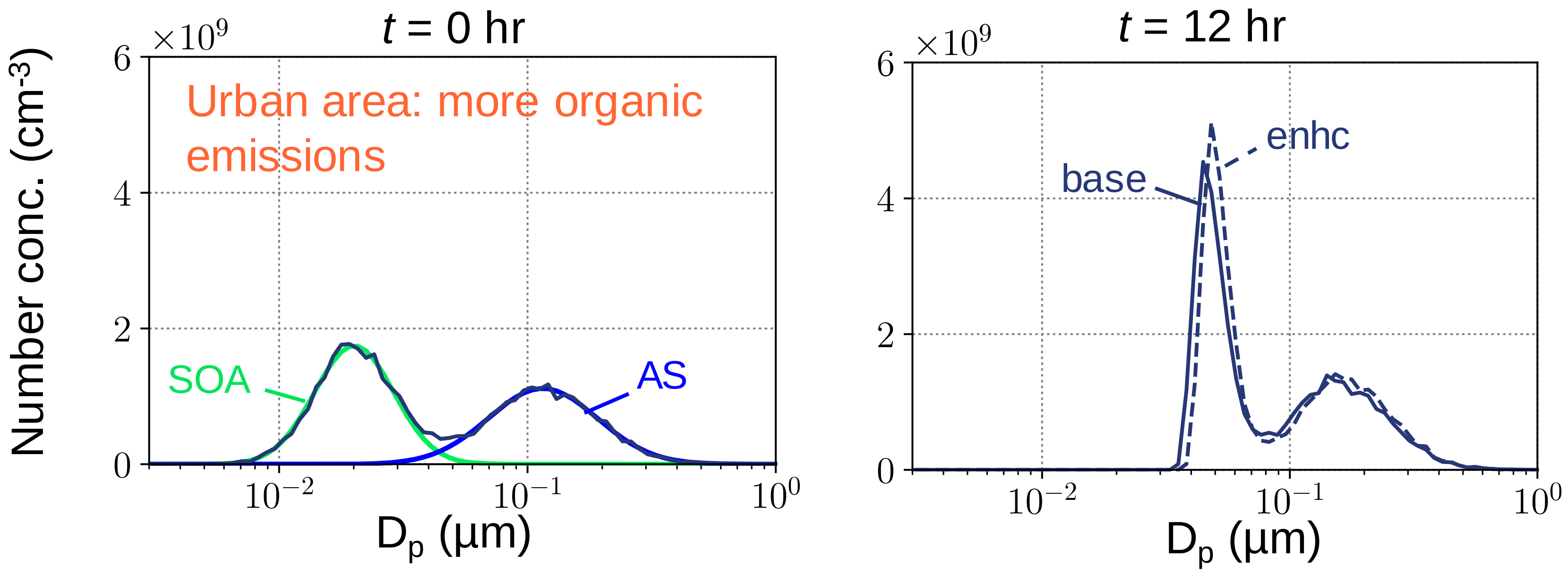
Case B: Bimodal distribution + AS in the small mode



Reduces the distance between the peaks by approximately 36%.

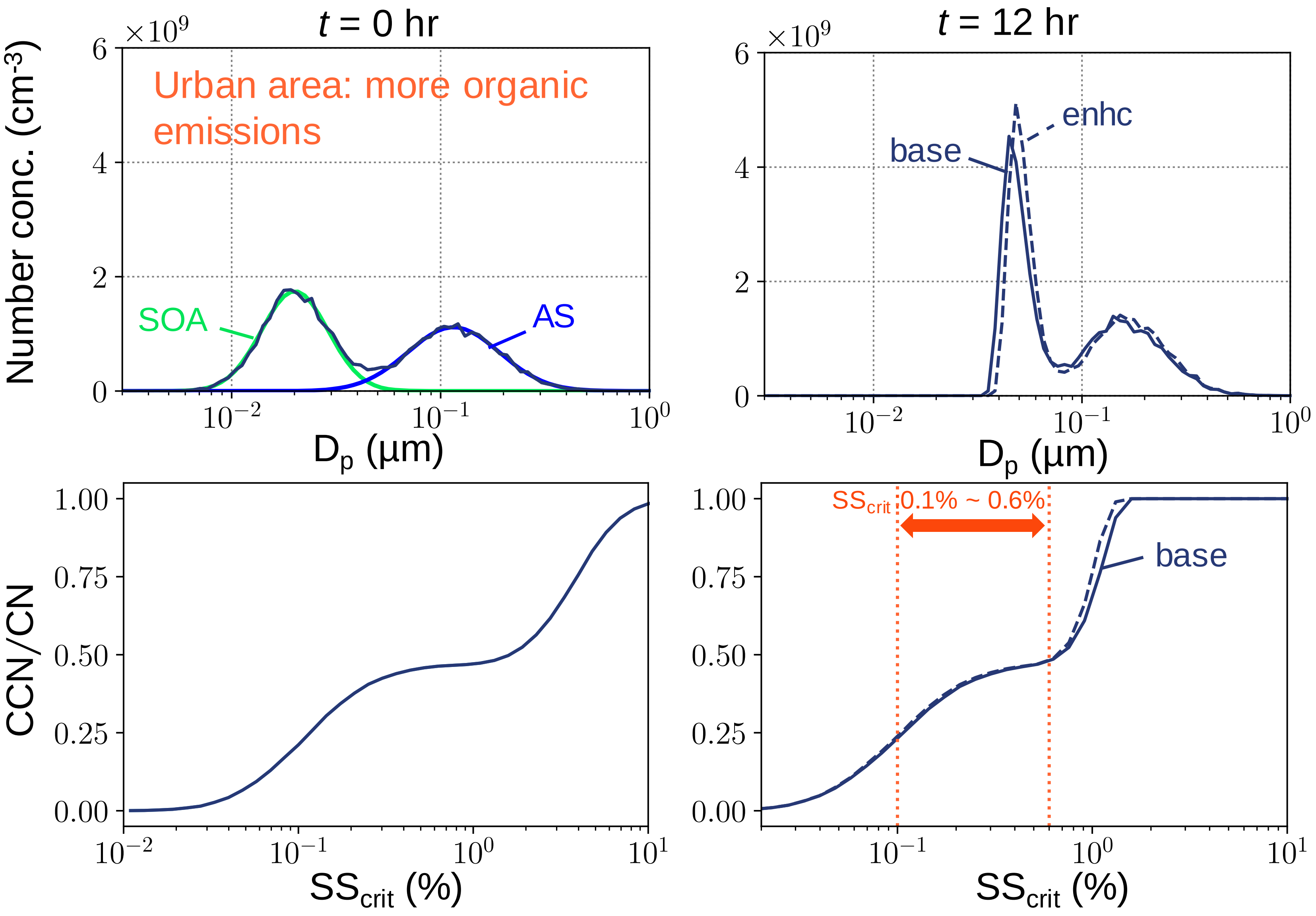
Underestimation in CCN/CN is approximately 0.3% (SS_{crit} = 0.1%), 21% (SS_{crit} = 0.6%).

Case C: Bimodal distribution + SOA in the small mode



Slight increase in both small mode and large mode by ~0.4%.

Case C: Bimodal distribution + SOA in the small mode

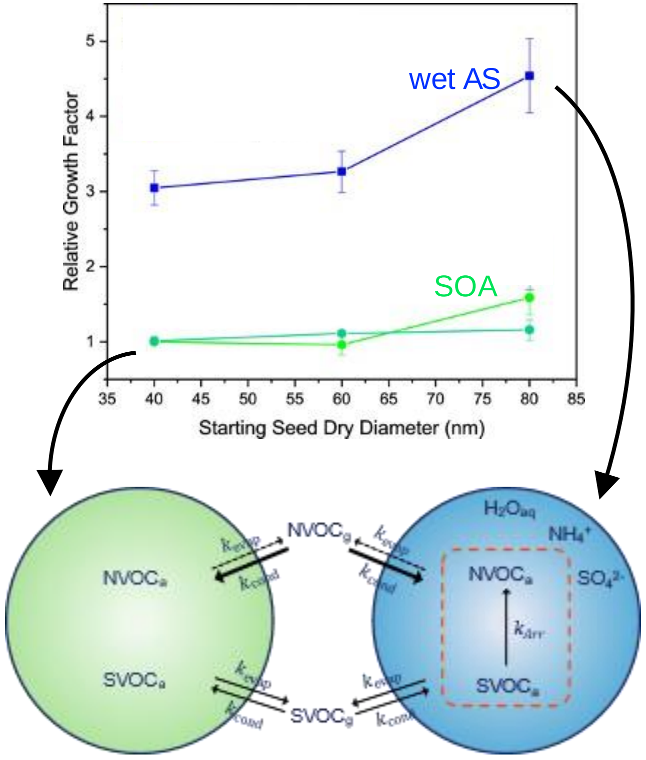


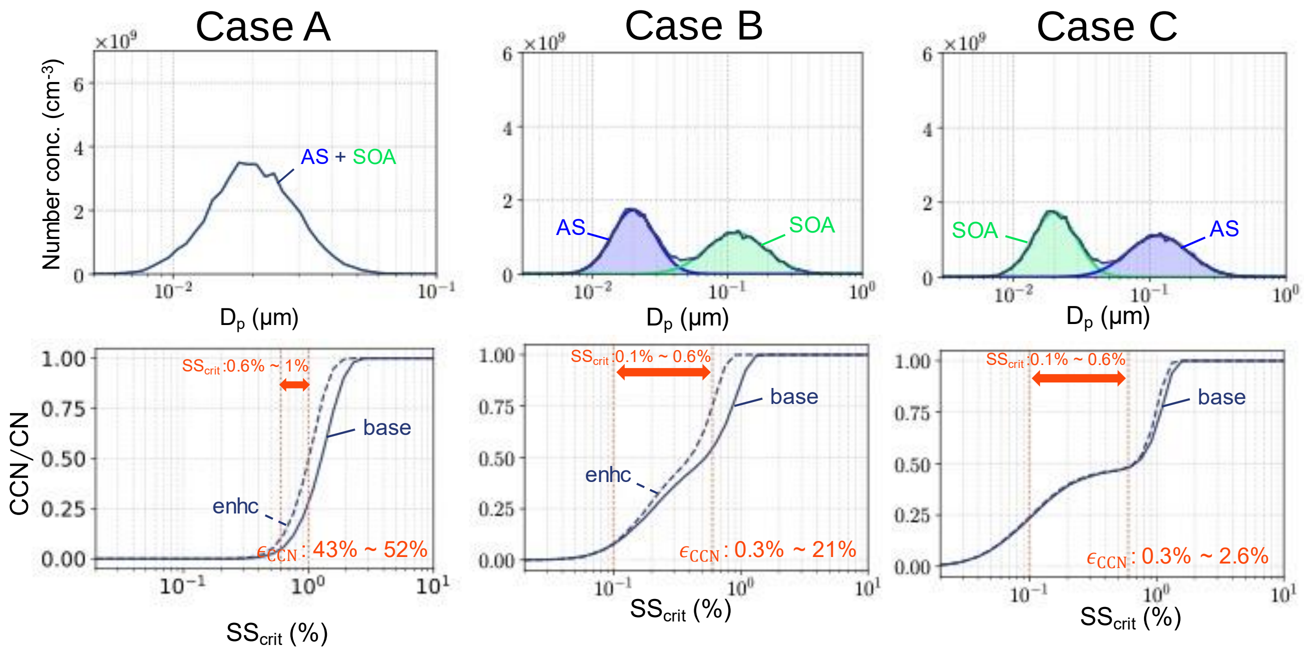
Slight increase in both small mode and large mode by ~0.4%.

Underestimation in CCN/CN is approximately 0.3% (SS_{crit} = 0.1%), 2.6% (SS_{crit} = 0.6%).

Concluding thoughts

- Created framework for seed-dependent particle growth using PartMC and CAMP.
- Impact on CCN concentration when ultrafine AS particles undergo enhanced growth.





Future work: refining mechanisms + scenario analysis (simple mixing states → more complex cases)

Email

yicenl2@illinois.edu

Code availability

PartMC: https://github.com/compdyn/partmc
CAMP: https://github.com/open-atmos/camp

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