Evaluating and improving reduced representations of atmospheric aerosol

Laura Fierce
Pacific Northwest National Laboratory

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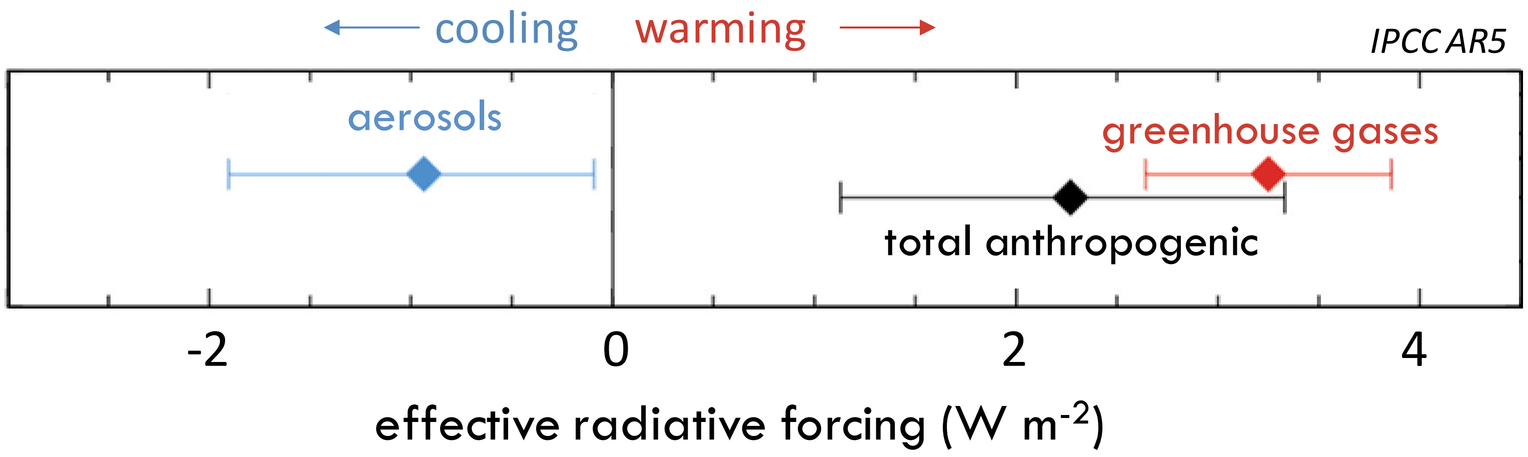
Aerosol effects are large source of uncertainty in total anthropogenic forcing



net warming by greenhouse gases is partially offset by net cooling by aerosols

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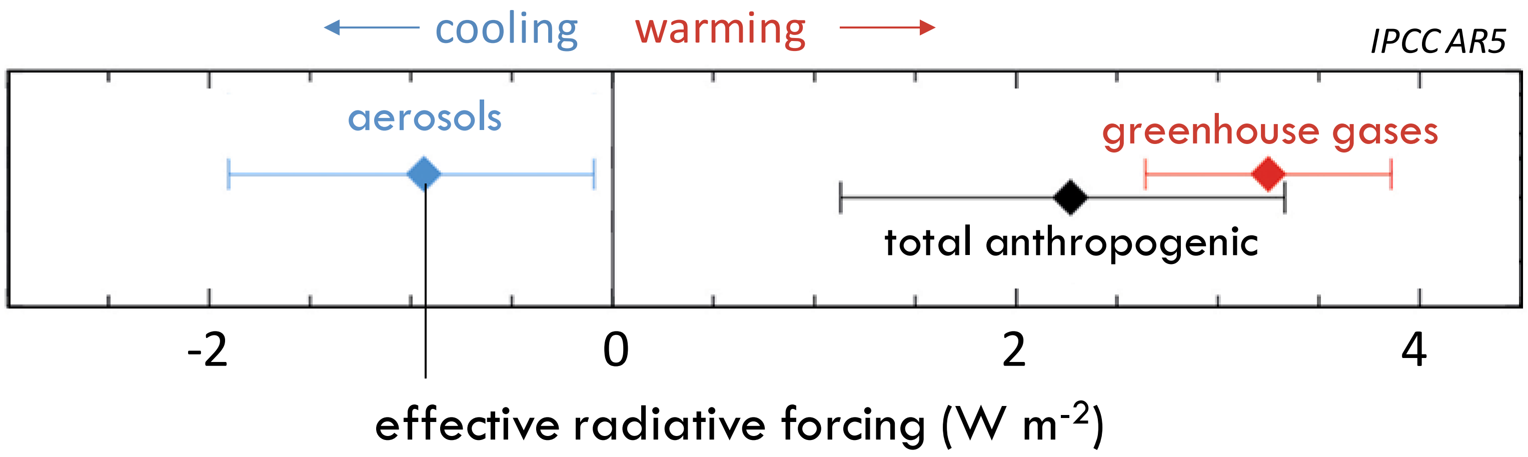
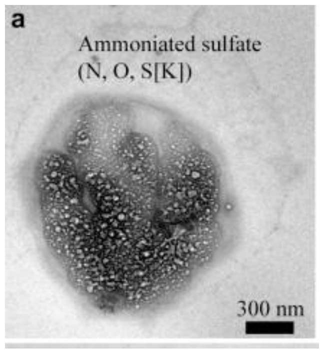
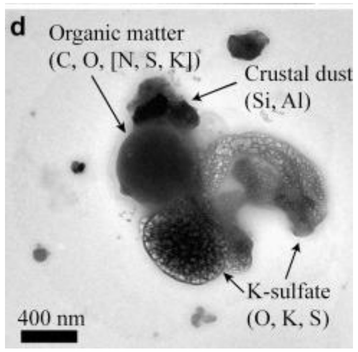


Aerosol effects depend on properties of diverse particles

Li et al., Atmospheric Environment 2011

mixture of inorganics





mixture of inorganics, organics, and dust

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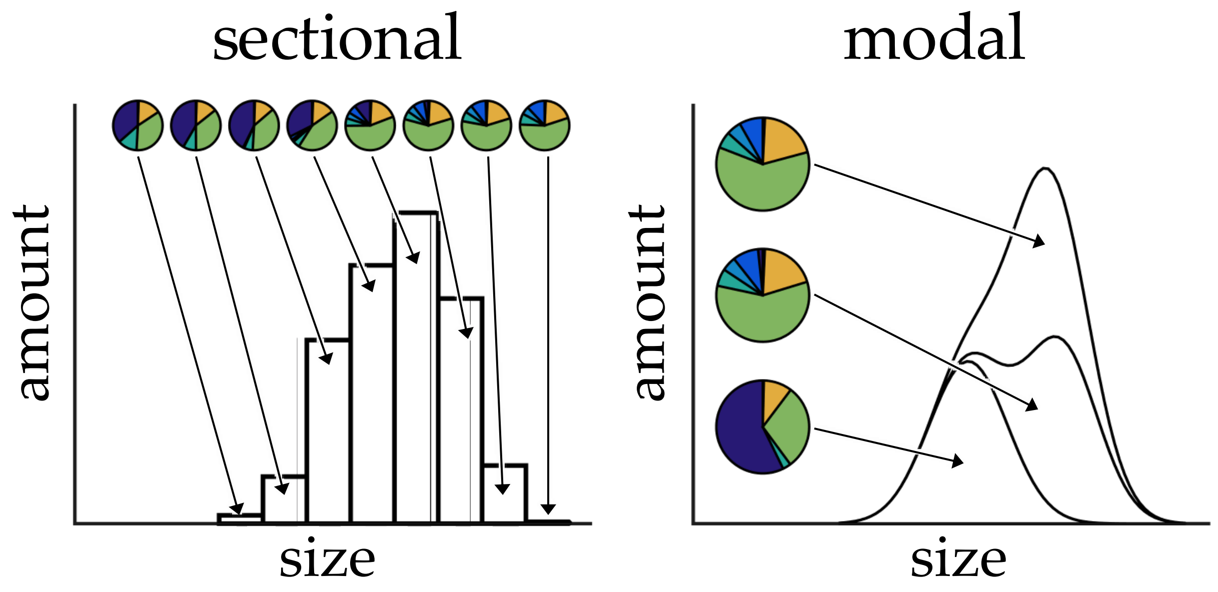
Particle populations are not easily simulated in large-scale models



Common aerosol representations in global-scale aerosol schemes

Large-scale aerosol models simplify the representation of particle size and composition

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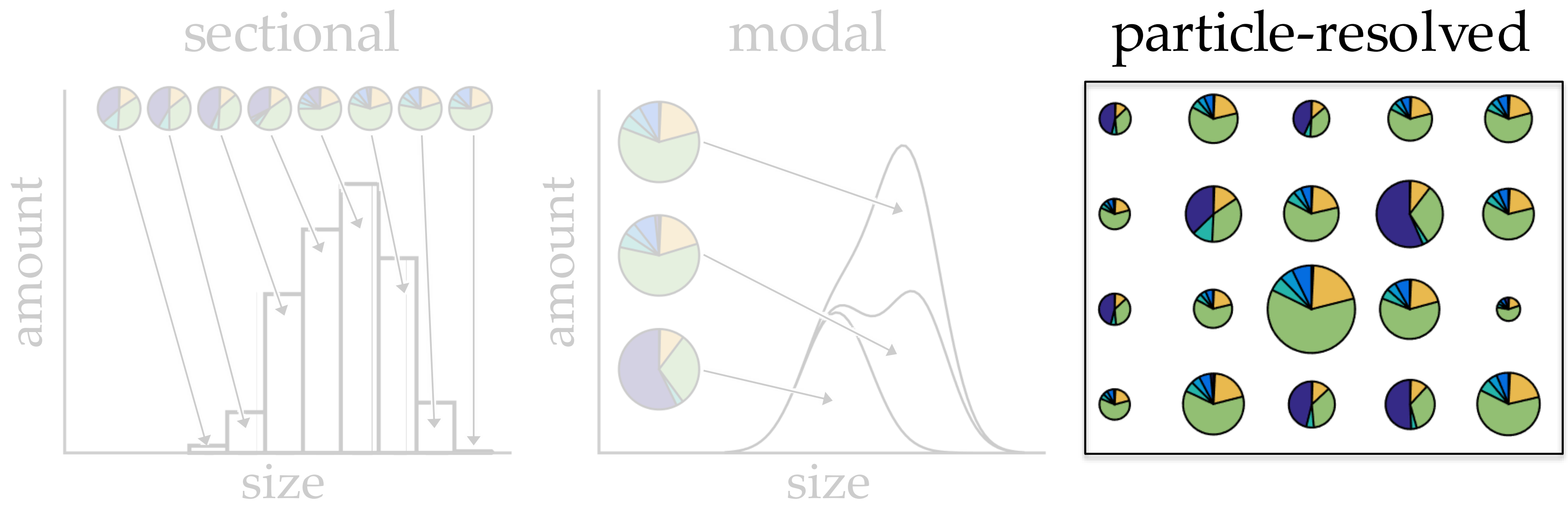
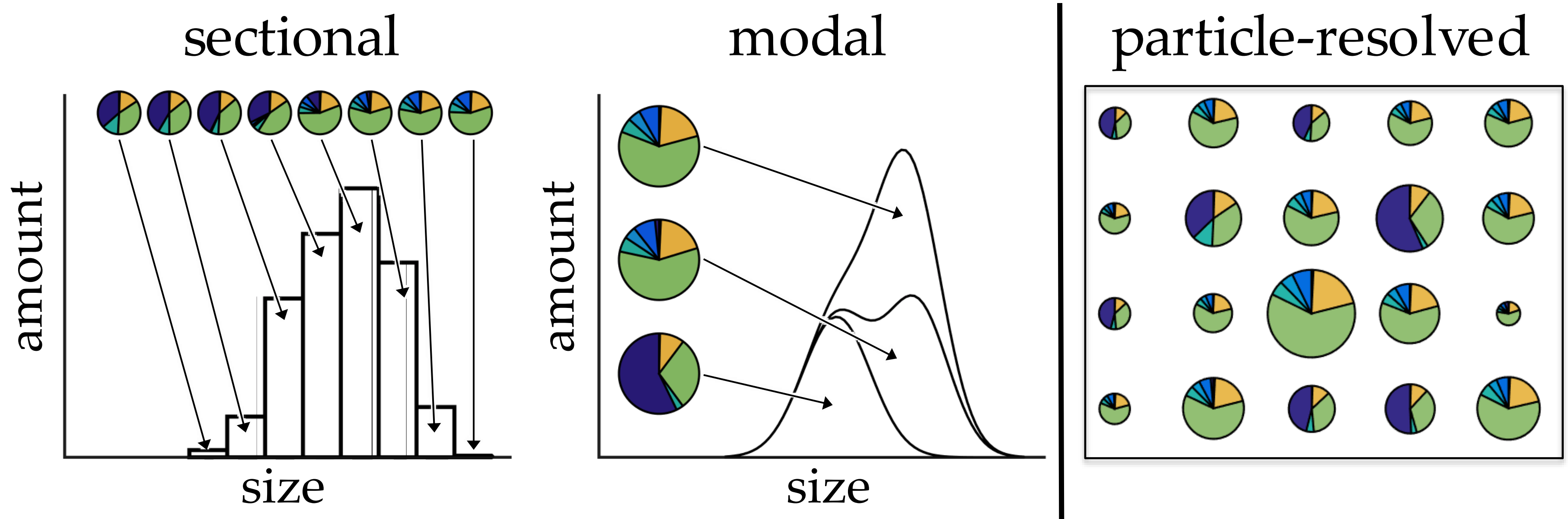


Particle-resolved model tracks per-particle composition for thousands of individual particles

Particle populations are not easily simulated in large-scale models



too simple to accurately resolve particle properties

too complex for global simulations

How well do reduced aerosol schemes represent particle properties?

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Consider two factors:

- Simplification of particle mixing state
- Simplification of particle size distributions

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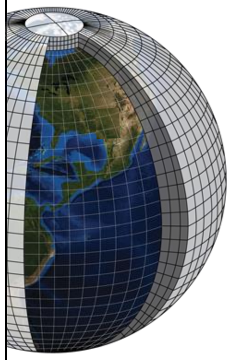
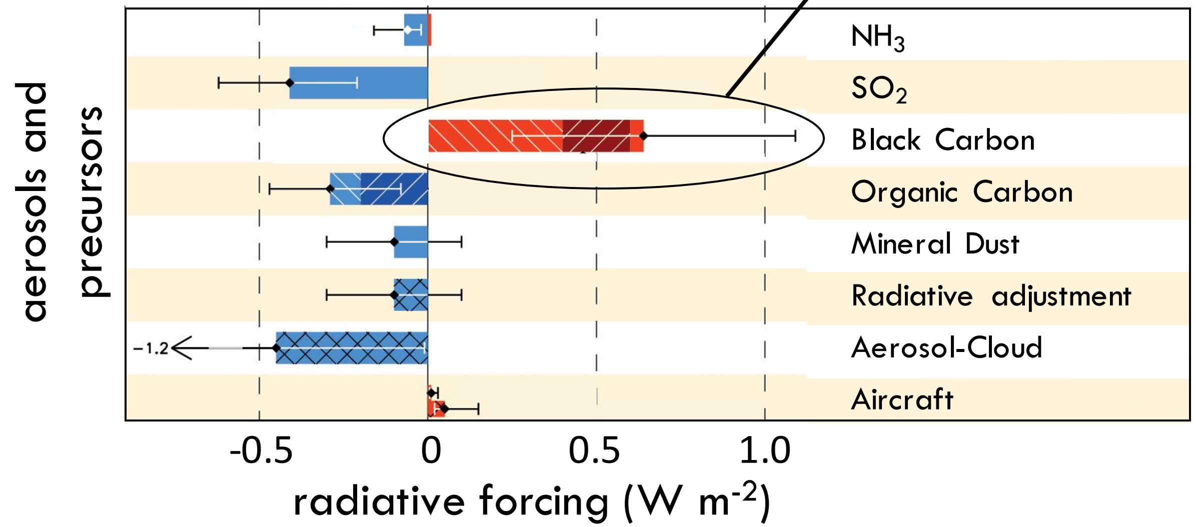
- **Simplification of particle mixing state**
- Simplification of particle size distributions

Example: radiative effects of black carbon are determined by properties of diverse particles

Simulations of the global atmosphere are used to quantify radiative forcing



Need **optical coefficients** in each grid cell to compute **direct forcing**



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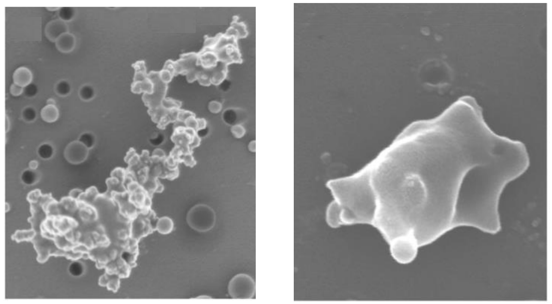
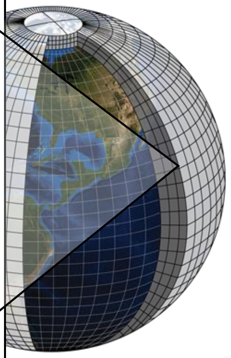


China et al., 2013

Need to integrate over **optical cross sections** of diverse particles to find **optical properties**



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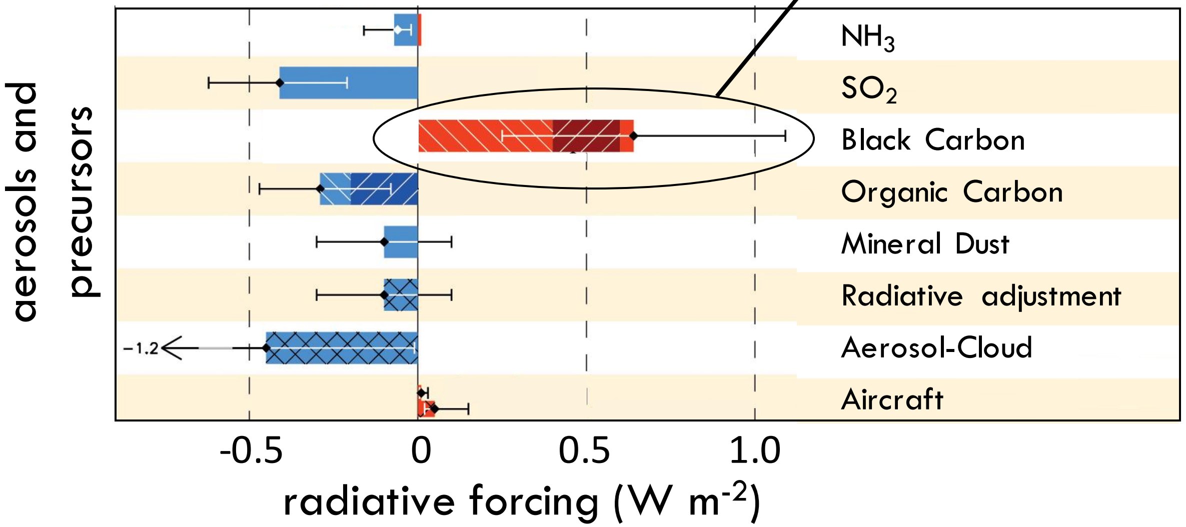
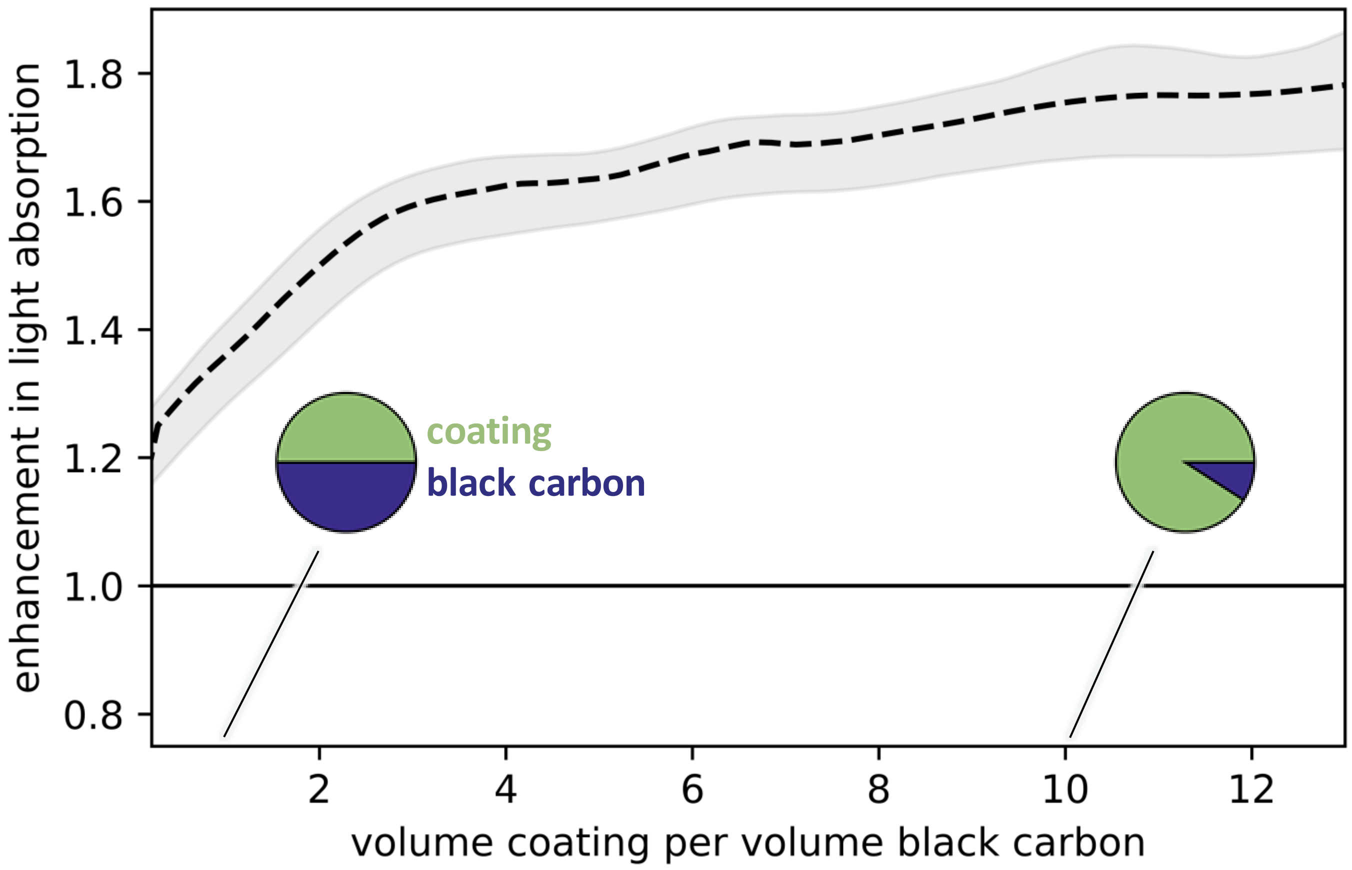


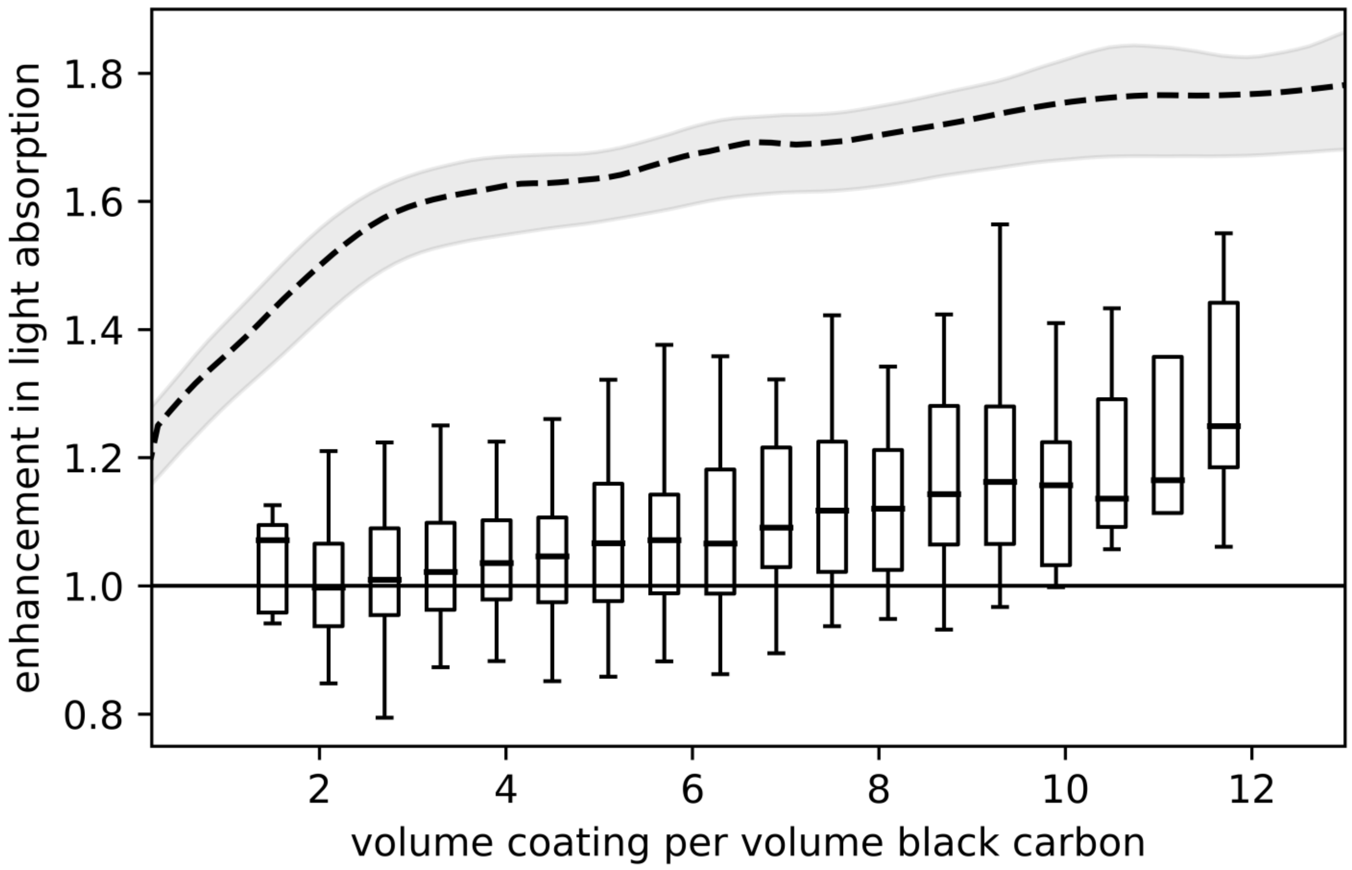
Common models predicts strong increase in black carbon's light absorption when coated



widely-used model for absorption

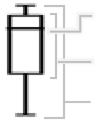
Fierce, Onasch et al., PNAS 2020

Field studies show only weak enhancements in light absorption by ambient black carbon.



widely-used model for absorption

ambient measurements in Fresno, CA*



* Similar findings reported by Cappa *et al.*, 2012, 2019, Healy et al., 2015

Fierce, Onasch et al., PNAS 2020

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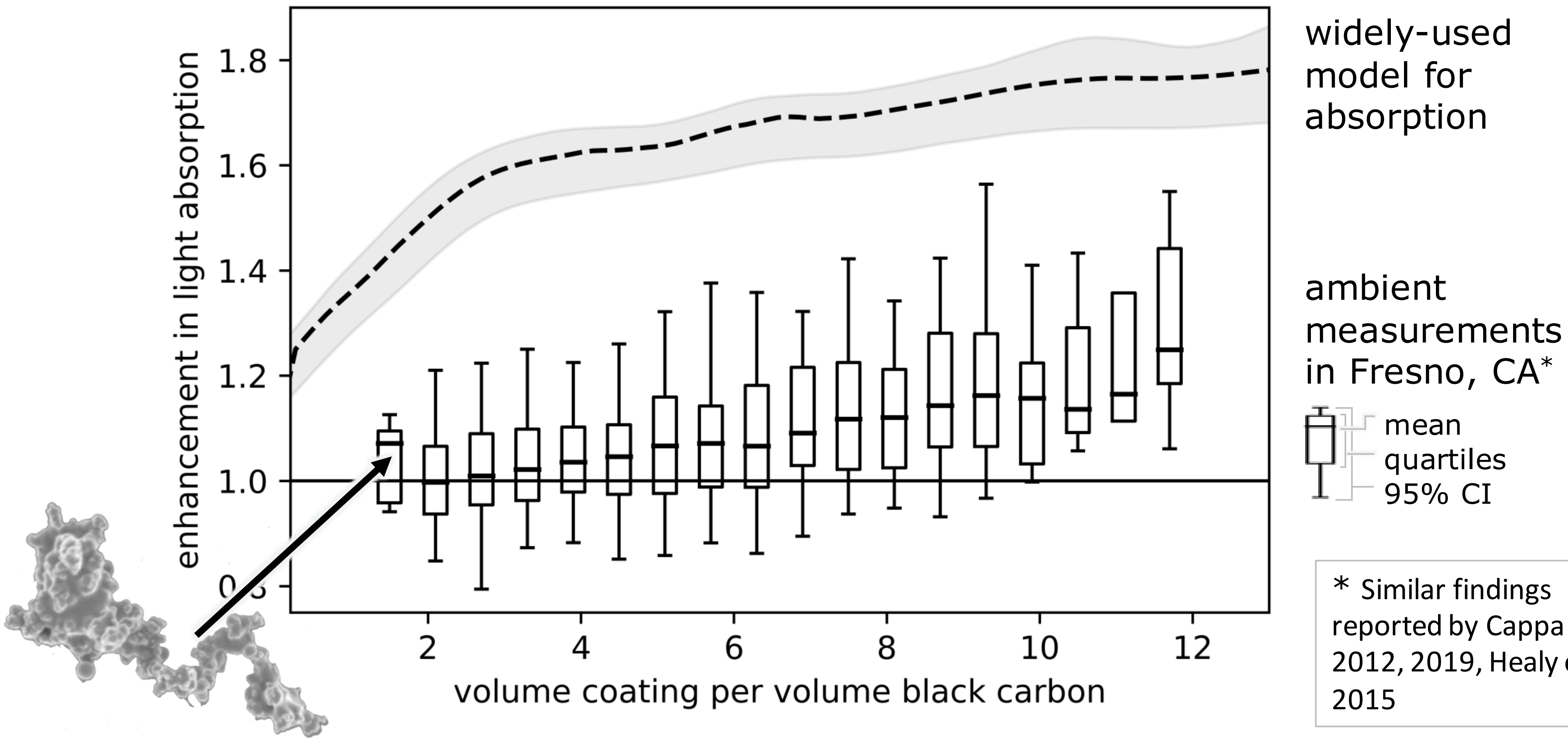
How do models misrepresent particles?
Unrealistic treatment of particle morphology



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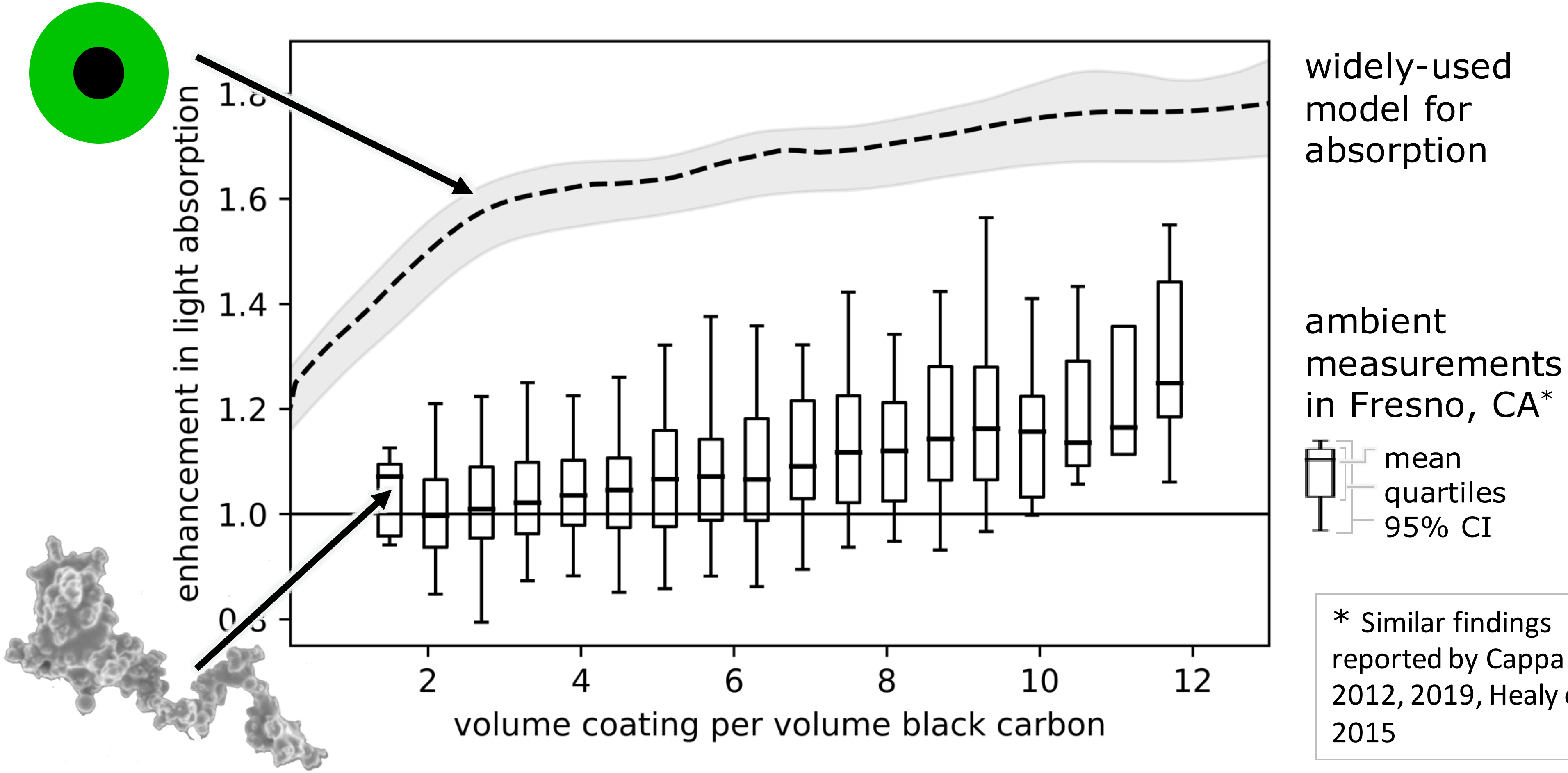

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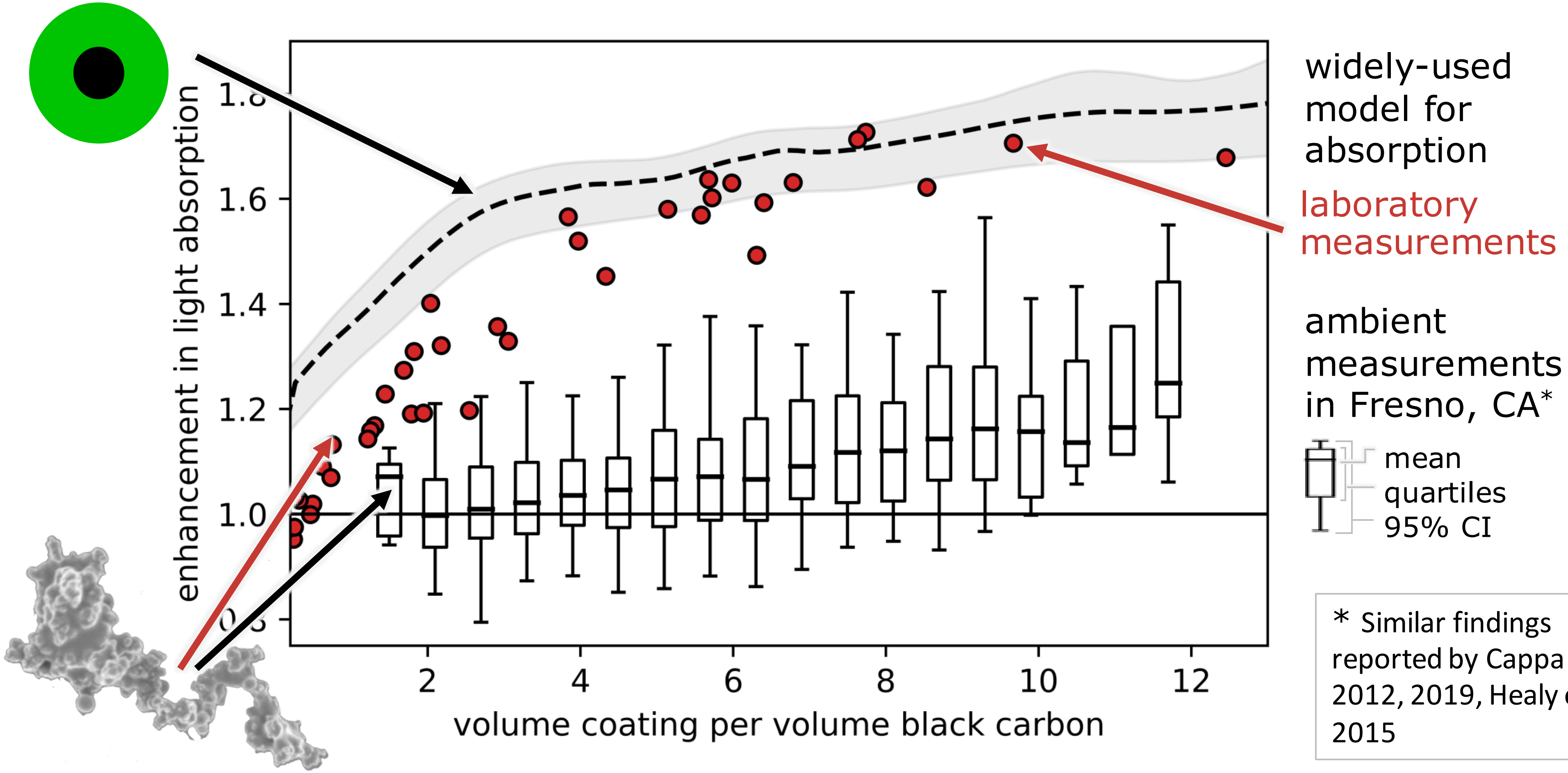

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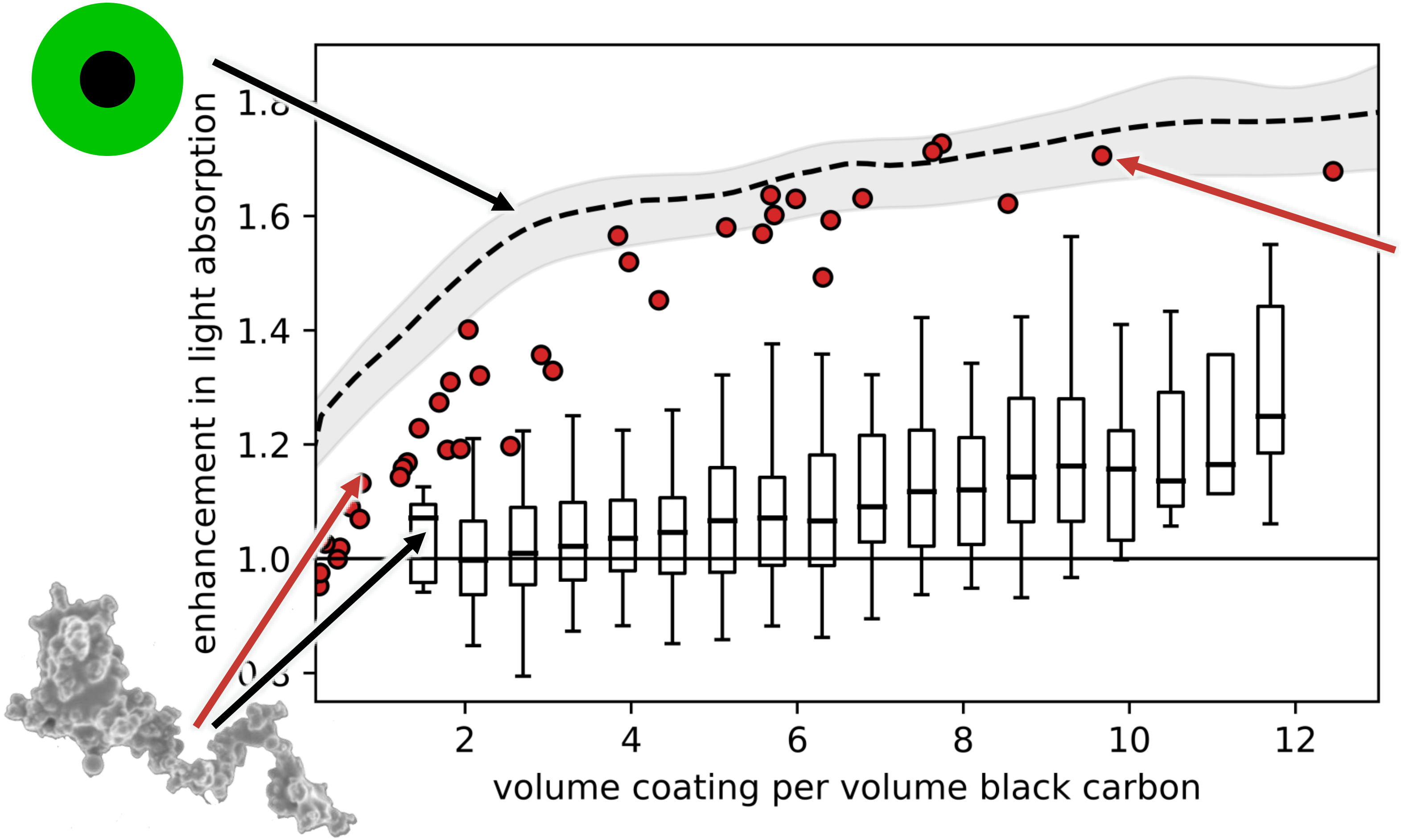

widely-used model for absorption

laboratory measurements

ambient measurements in Fresno, CA*

mean
quartiles
95% CI

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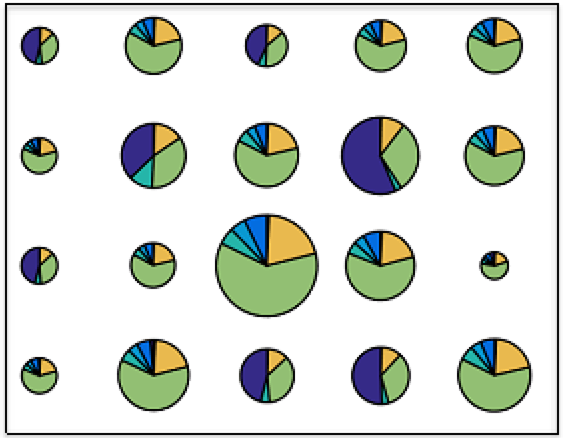
Fierce, Onasch et al., PNAS 2020

How do models misrepresent particles?

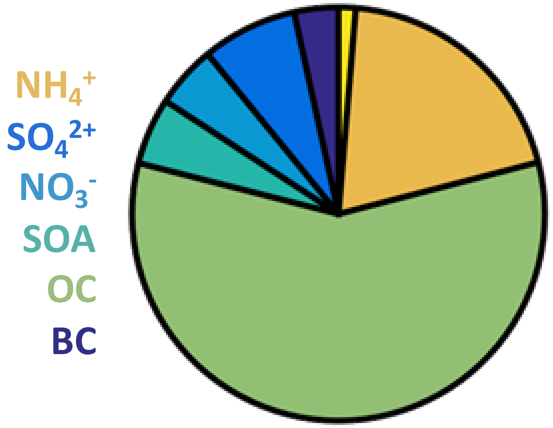
Simplify particle size-composition distribution

Particle-resolved composition



Ambient aerosol particles are diverse in size and composition

Population-averaged composition



Particle-to-particle diversity is not resolved in conventional models

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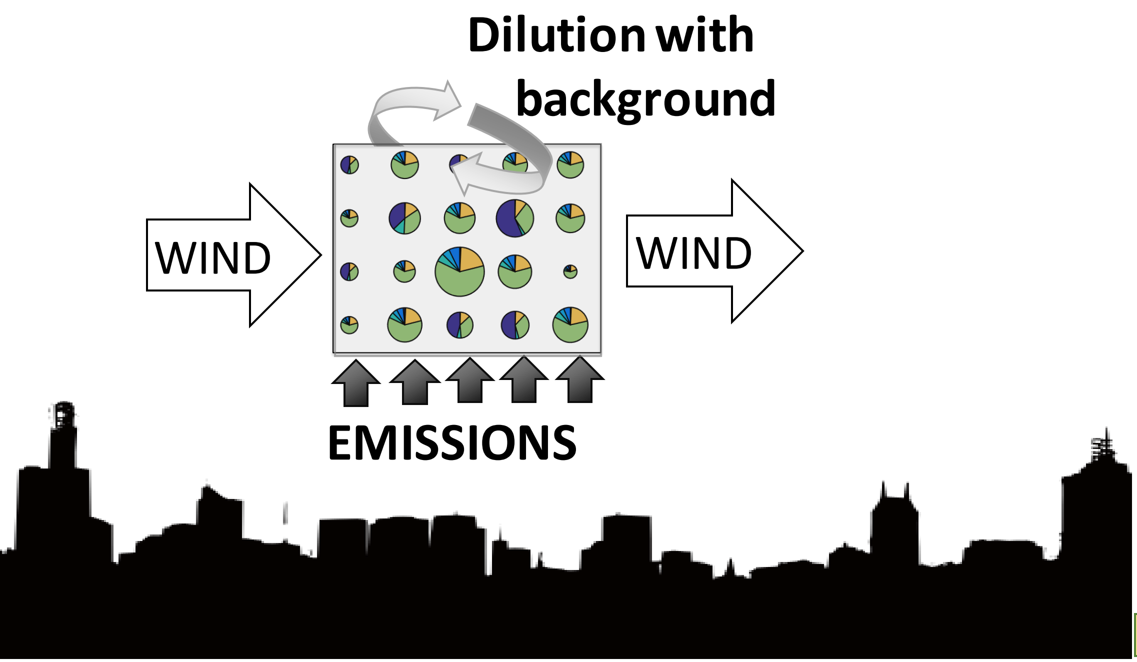
Simulate aerosol evolution in many scenarios with PartMC-MOSAIC

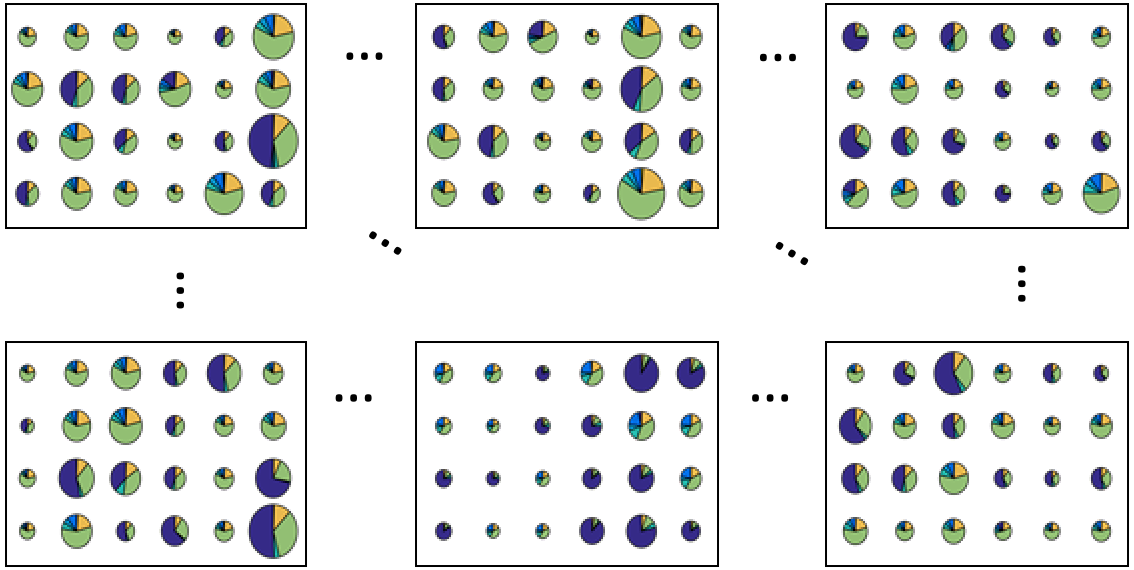


Model tracks per-particle size and composition for ~10^6 aerosol particles

Aerosol aging simulated under variety of conditions

PartMC: Riemer et al., JGR 2009; **MOSAIC:** Zaver et al., 2008

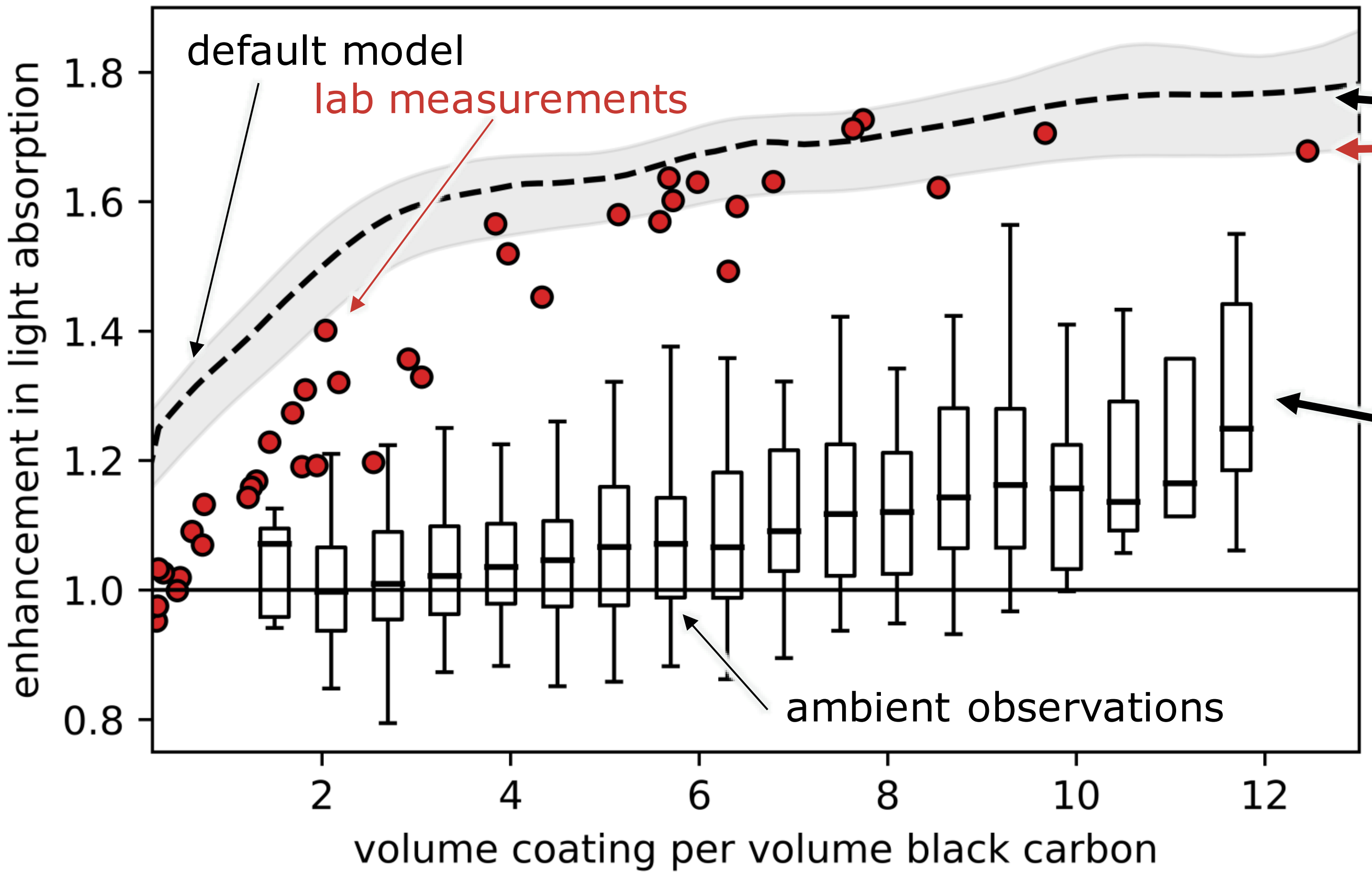
Simulate aerosol evolution in many scenarios with PartMC-MOSAIC

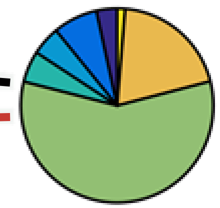


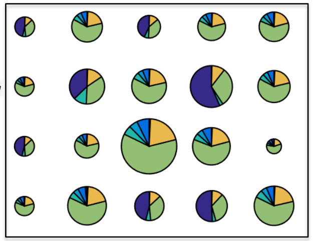
Thousands of aerosol populations

Modeled absorption by black carbon across ensemble of particle-resolved simulations

How do models misrepresent particles?
Simplify particle size-composition distribution

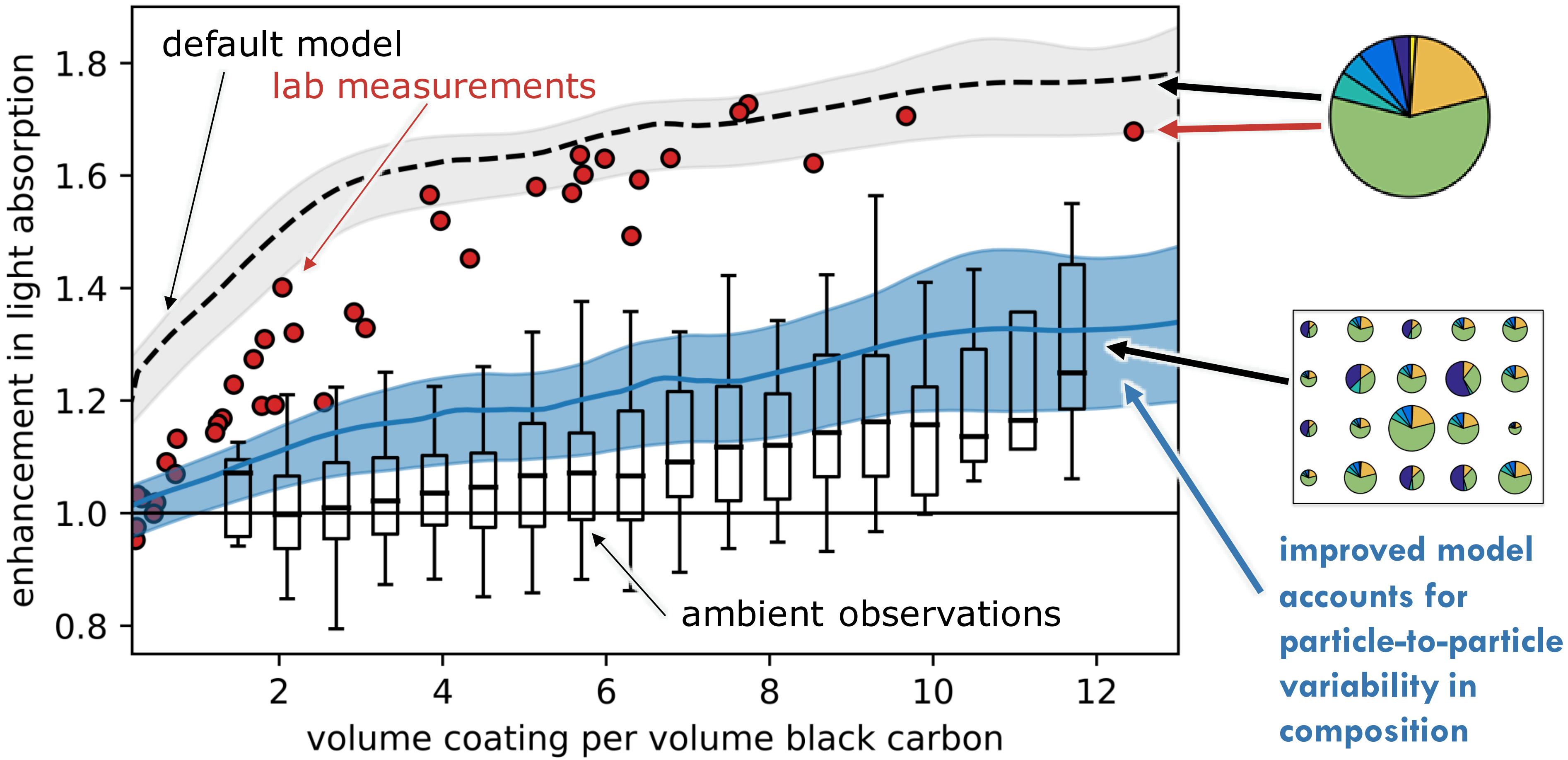






Fierce, Onasch et al., PNAS 2020

How do models misrepresent particles?
Simplify particle size-composition distribution



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How well do reduced aerosol schemes represent particle properties?

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Resolution of the mixing state is inadequate.

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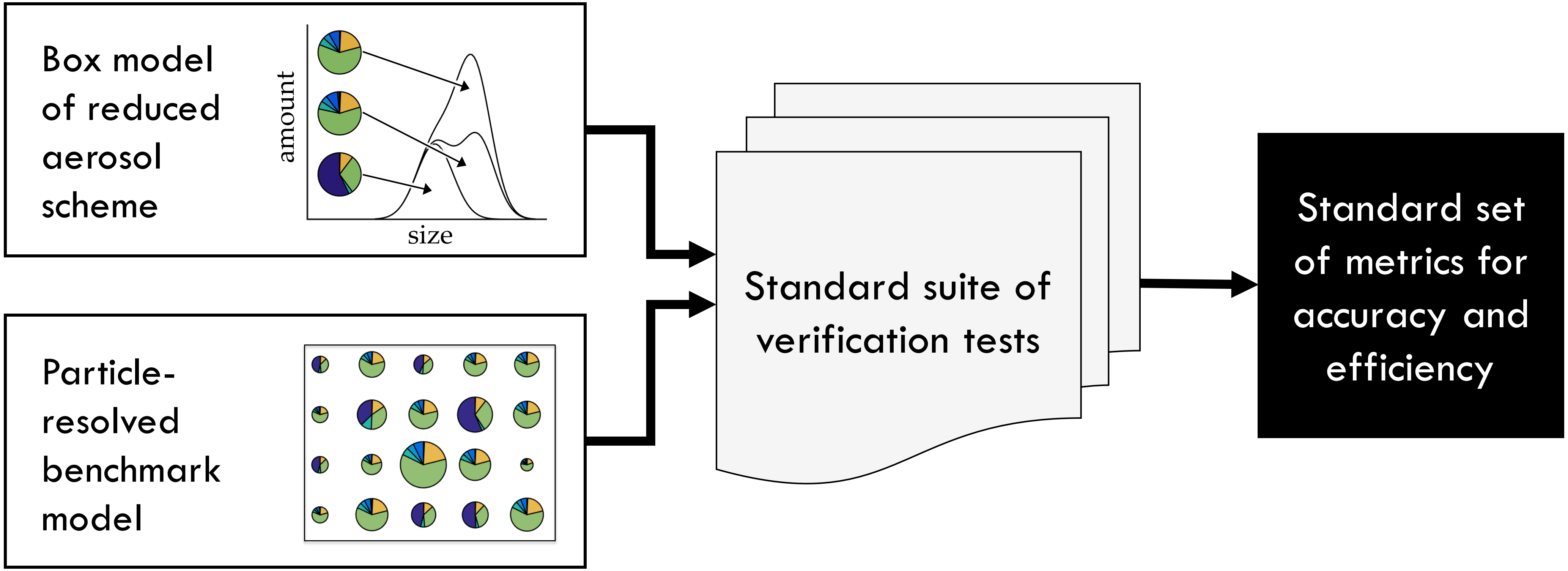
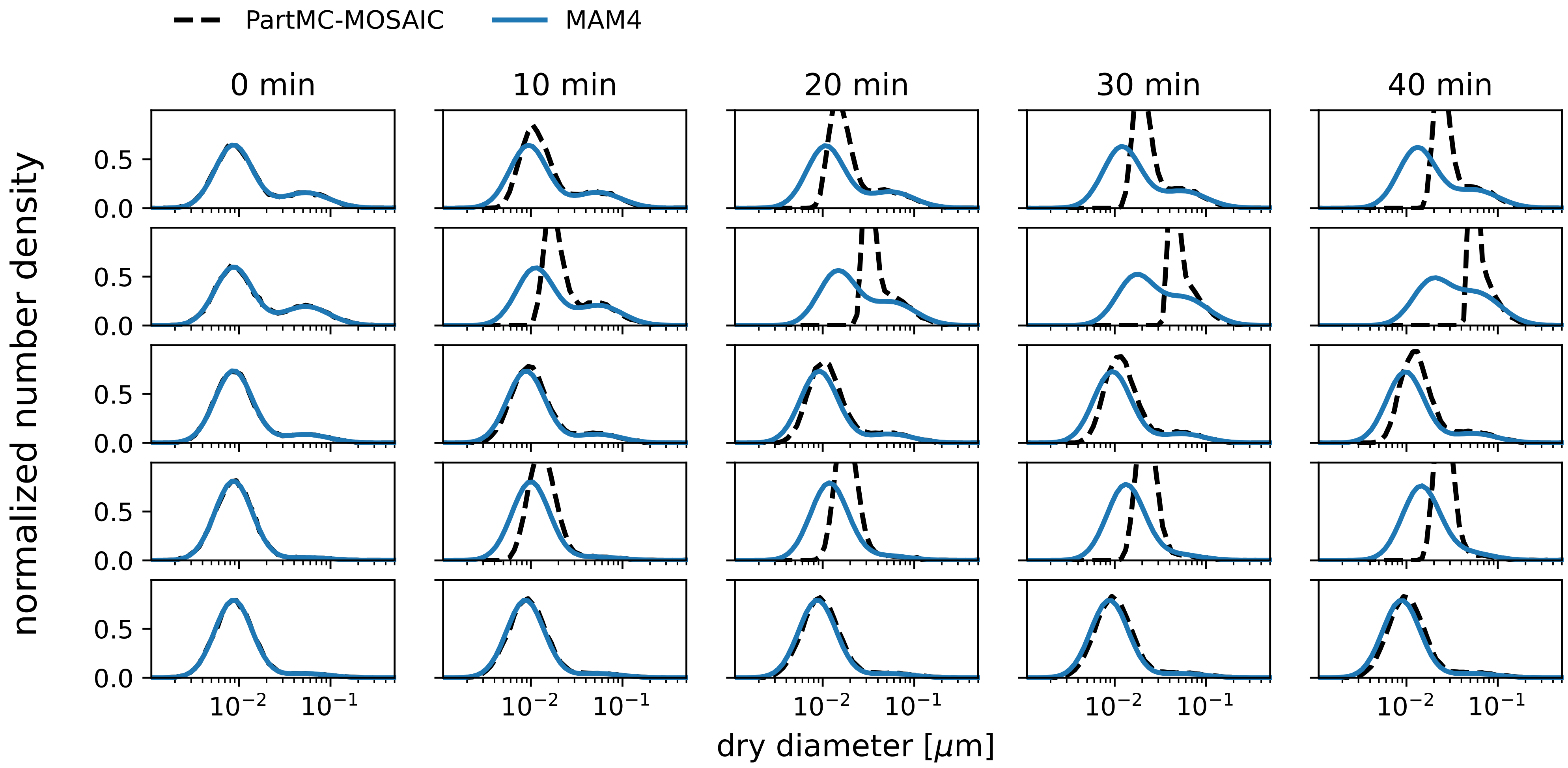
What about the size distribution?

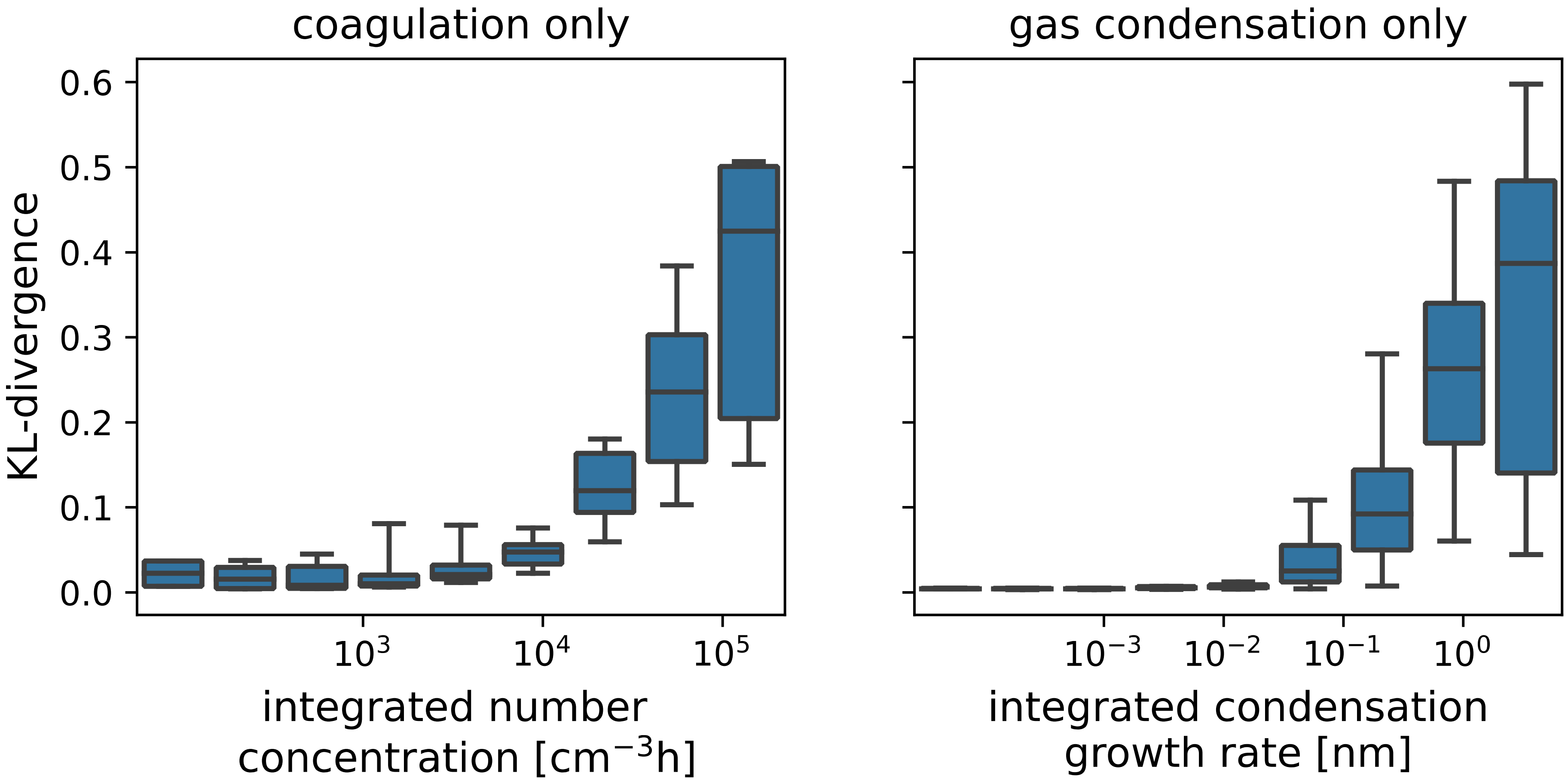
Approach: evaluate aerosol representations through box model comparisons



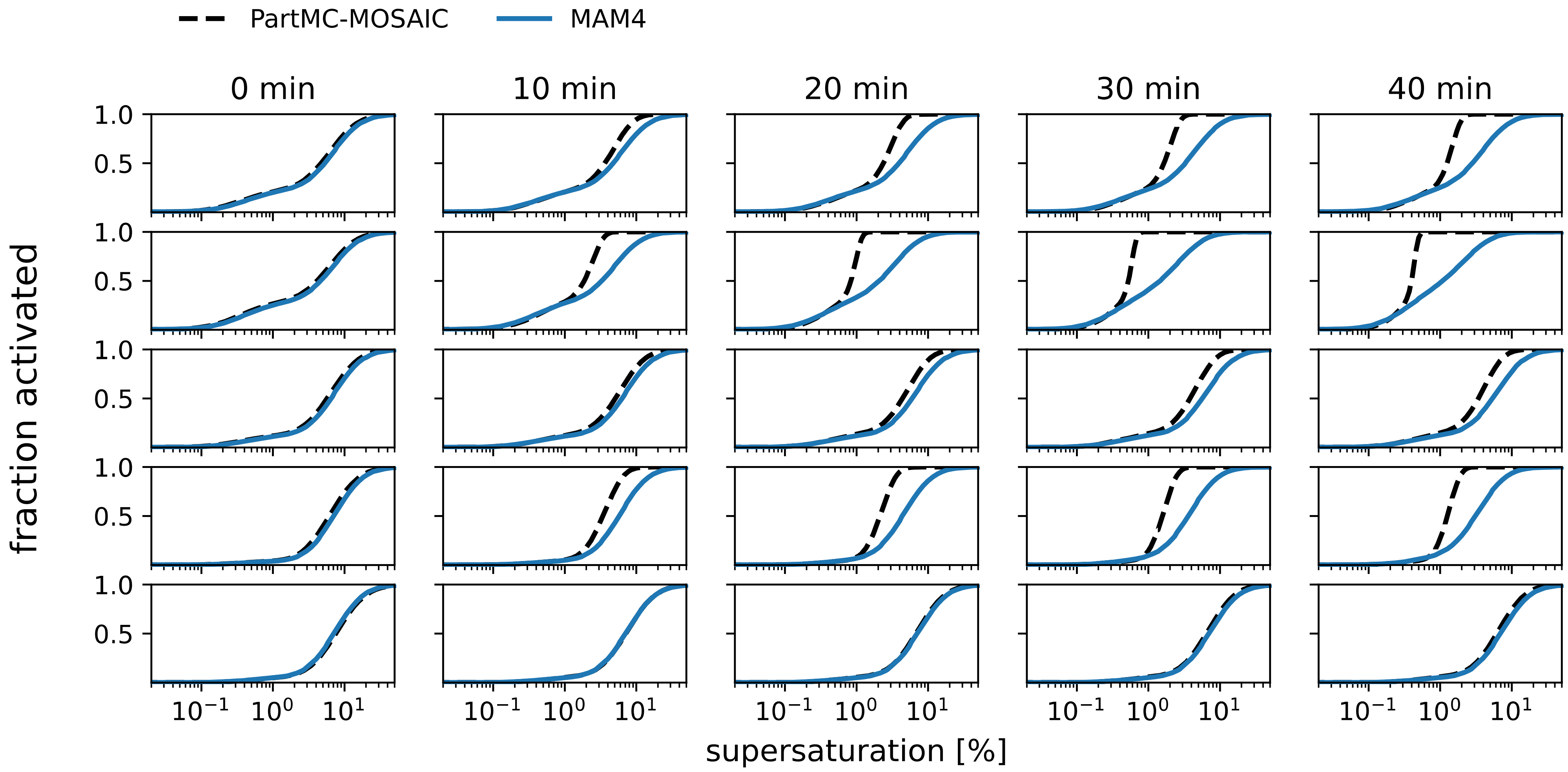
Size distributions simulated by MAM4 diverge from benchmark with aging



Differences in size distribution quantified by KL-divergence



Differences between size distributions lead to differences in CCN activity



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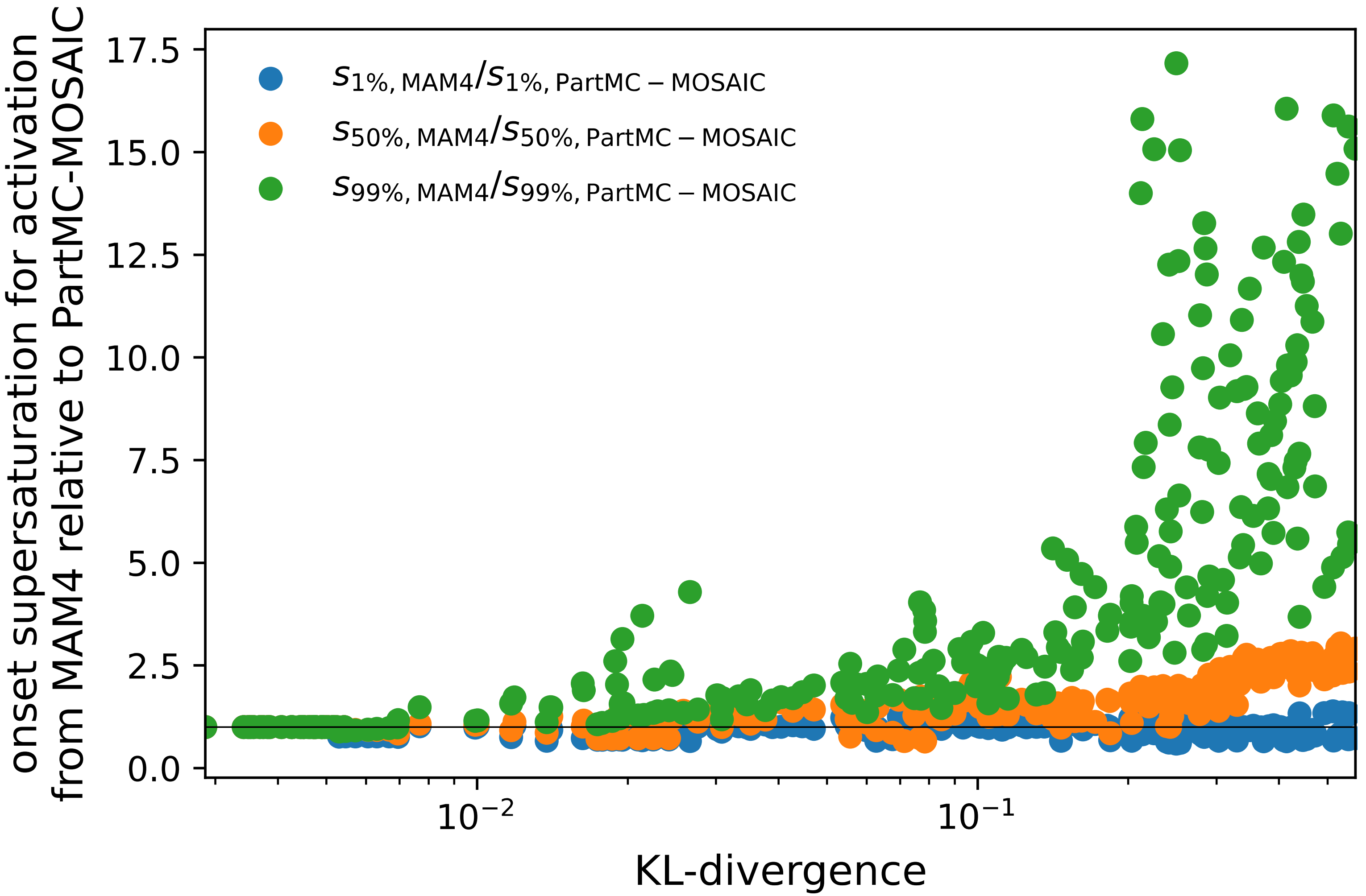
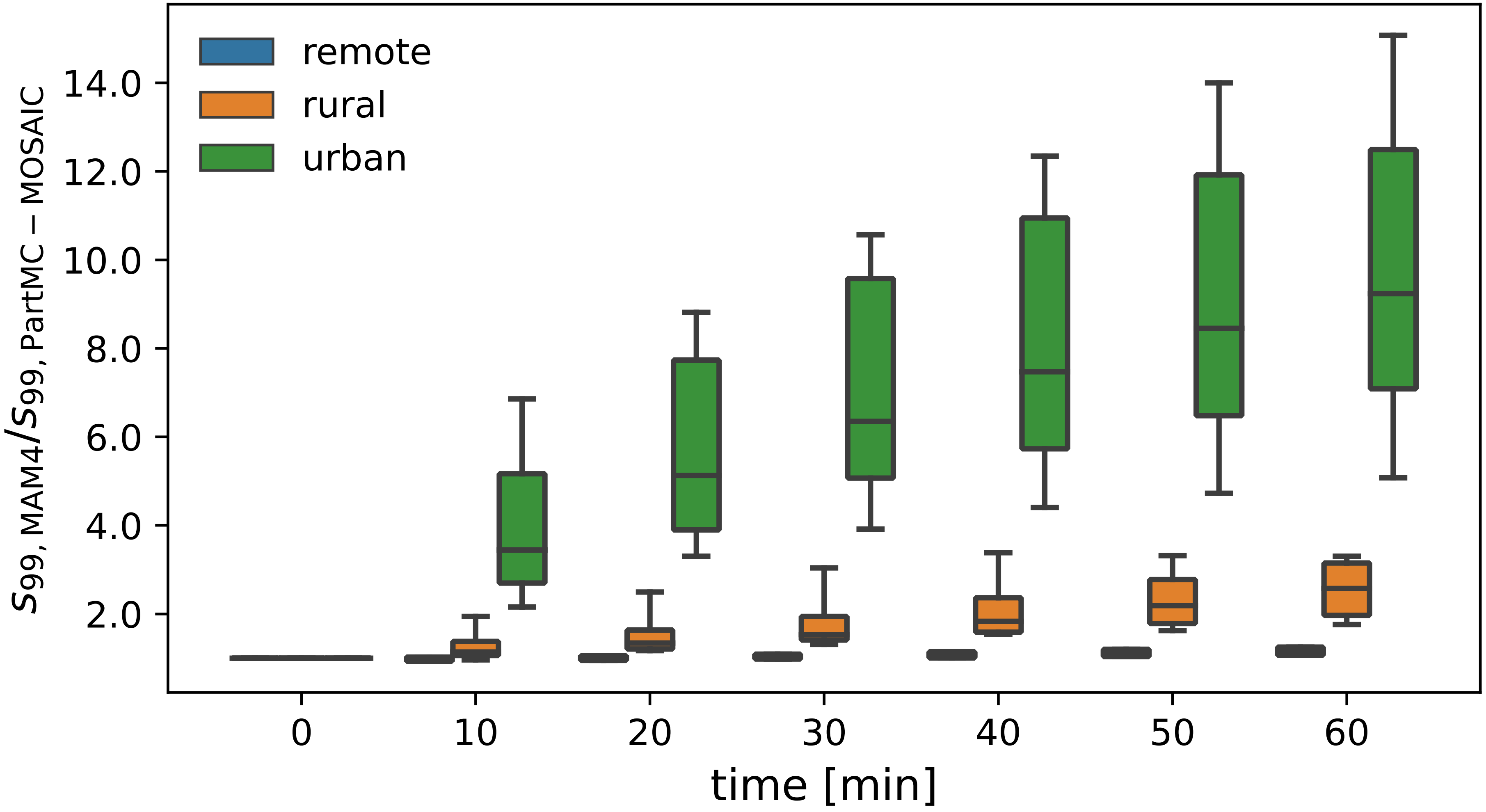


Largest differences in CCN activity in urban areas with rapid aging



How well do reduced aerosol schemes represent particle properties? **Not very well.**

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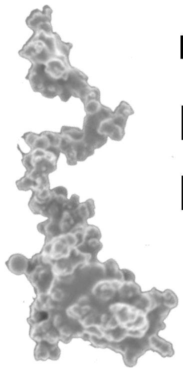
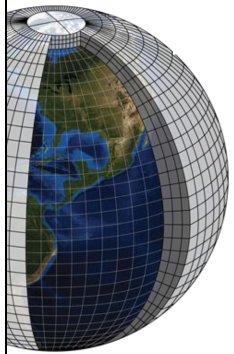
Can we do better?

How can global models account for the impact of particle-scale heterogeneity?



models for particle-scale processes

laboratory studies to inform particle-scale models



model prediction of global aerosol effects

field observations to evaluate and constrain global-scale models

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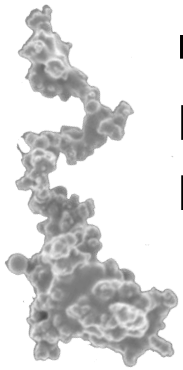
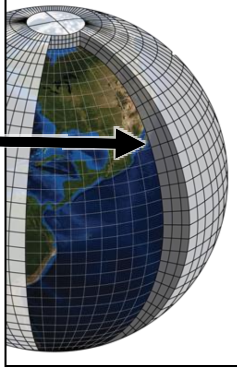


models for particle-scale processes

need to bridge scales by better representing particle heterogeneity



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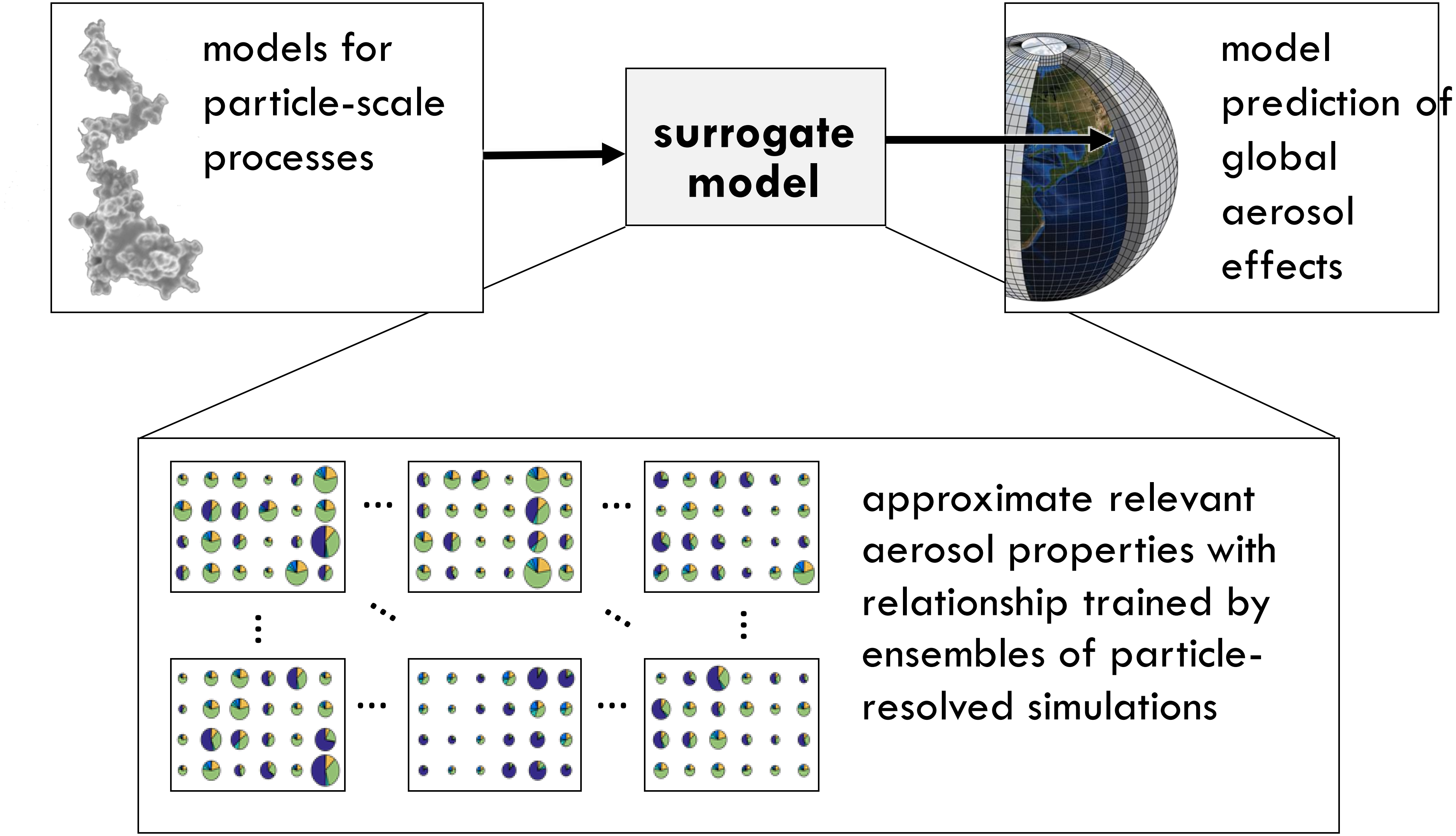
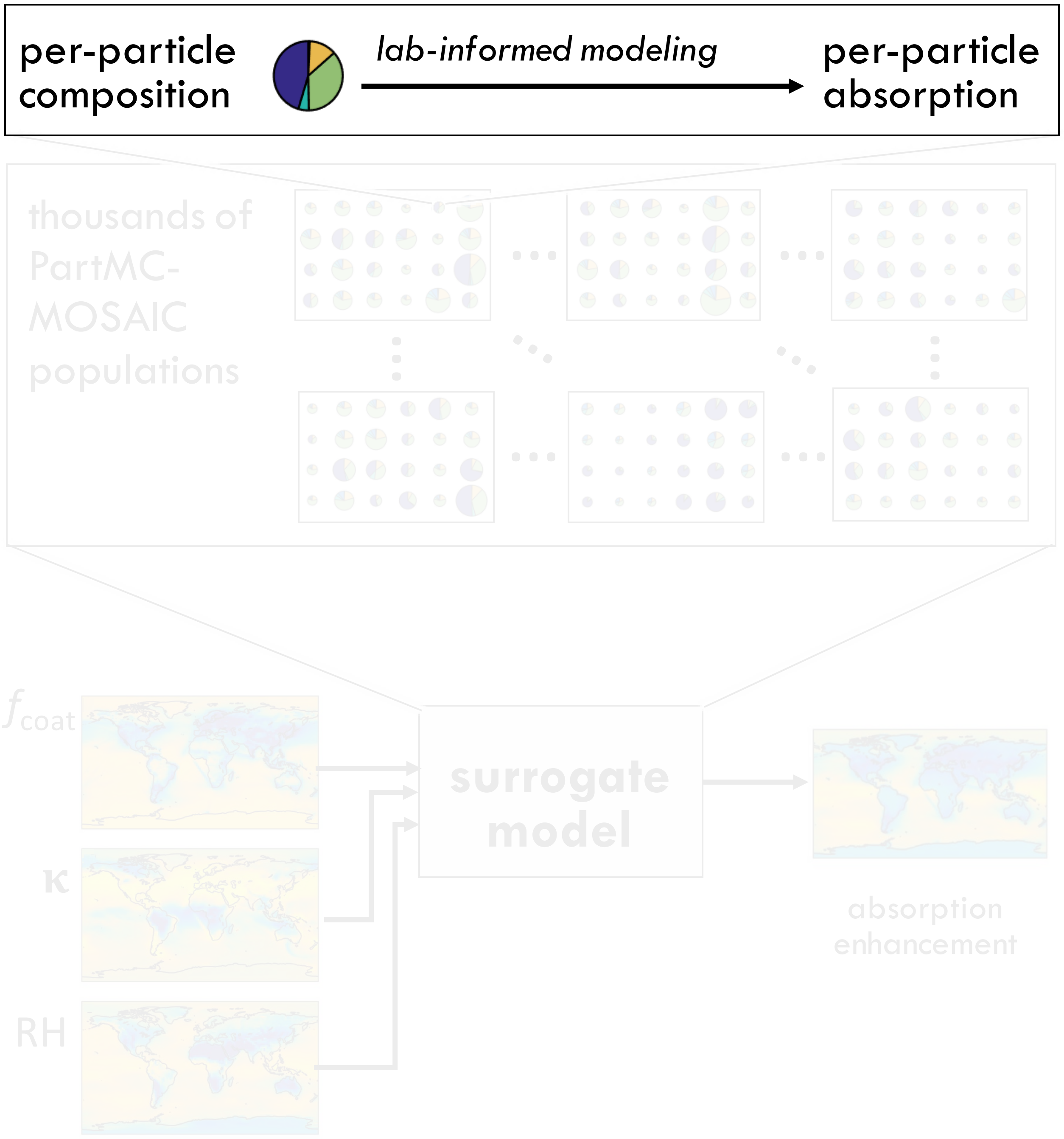
Path forward: surrogate models to bridge particle-scale and global-scale



Challenge:
Particle-resolved model is expensive.

Solution:
Surrogate model that approximately reproduces particle-resolved model predictions.



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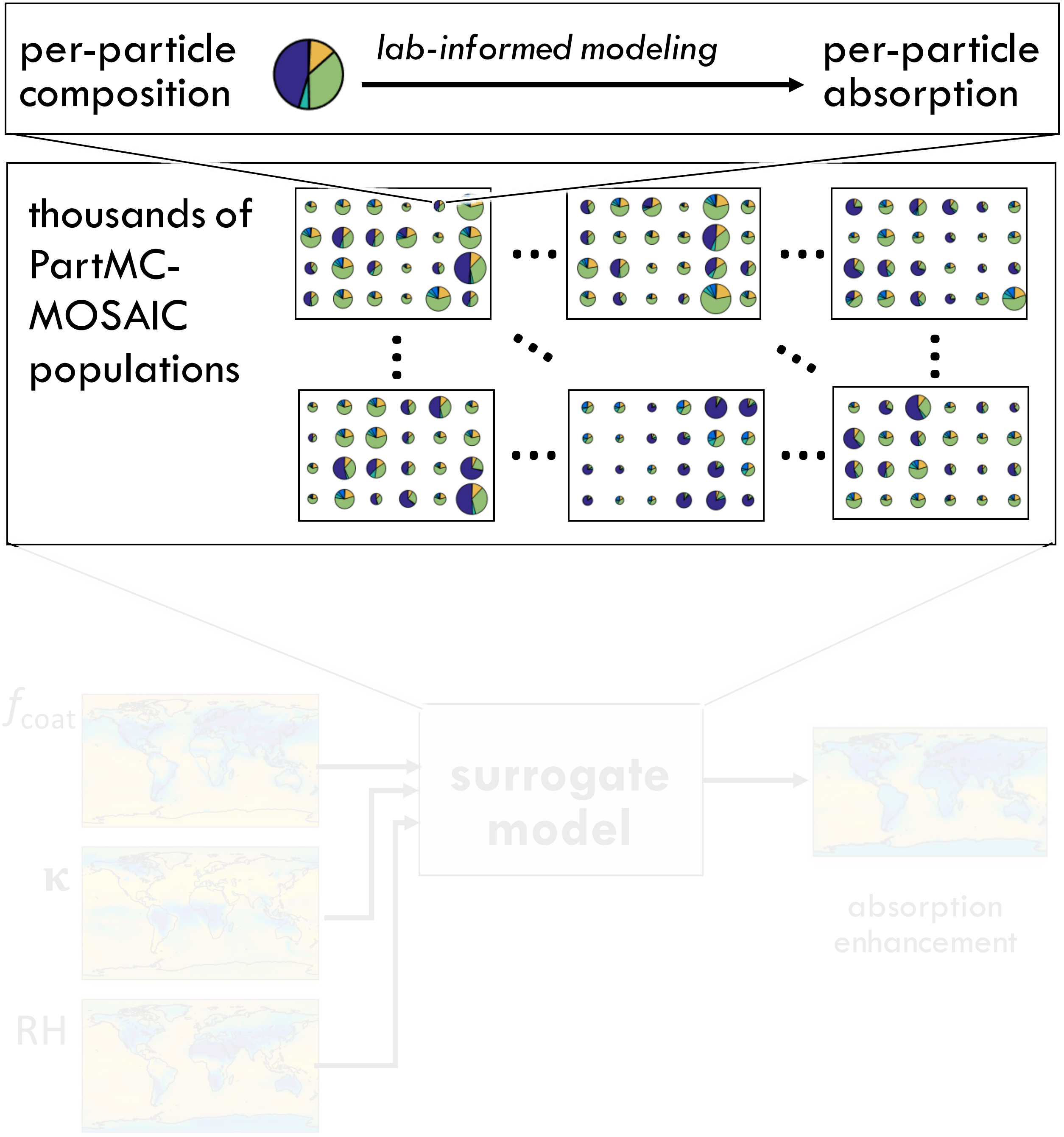

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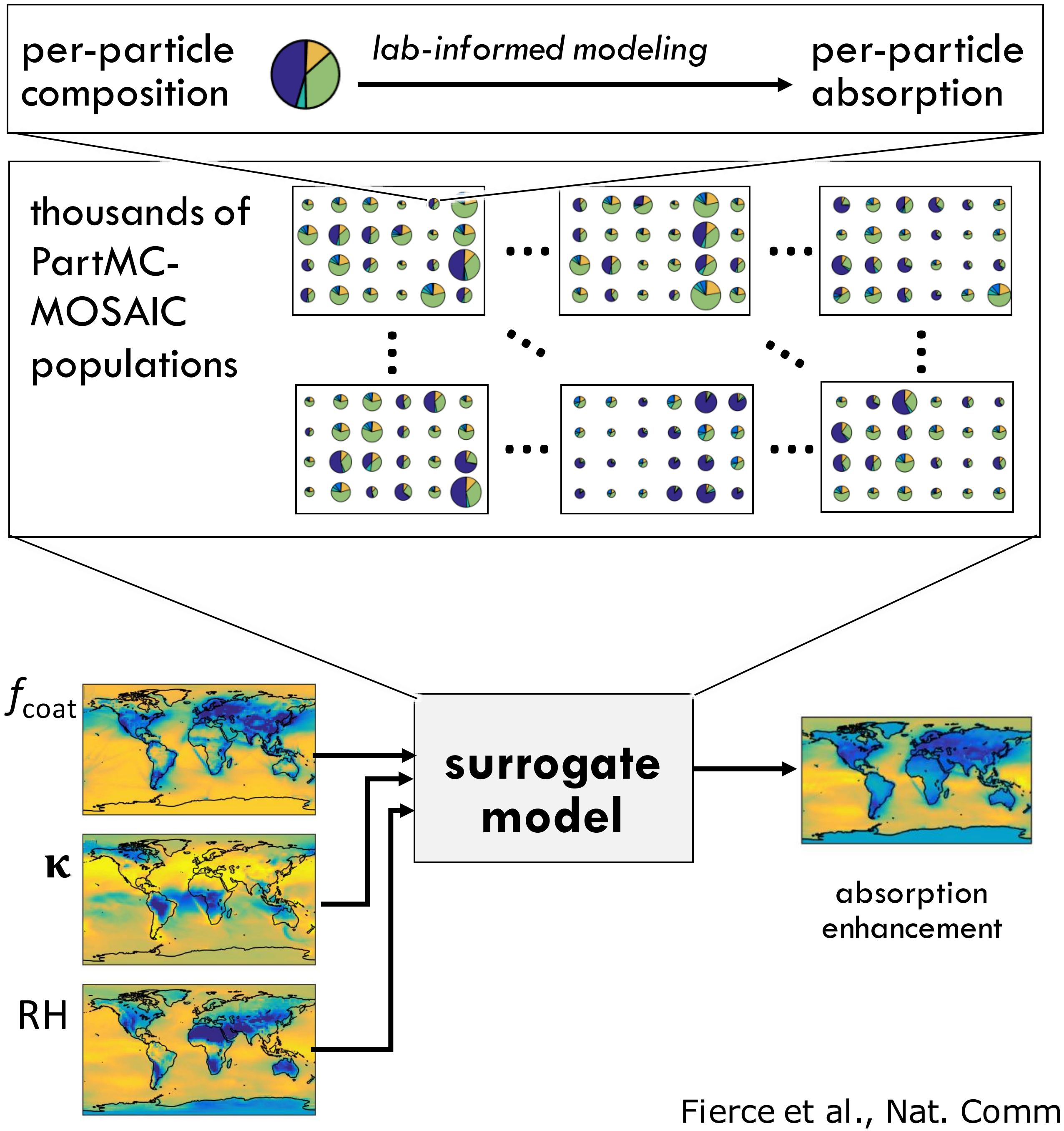
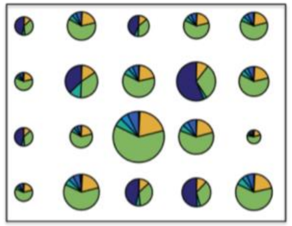


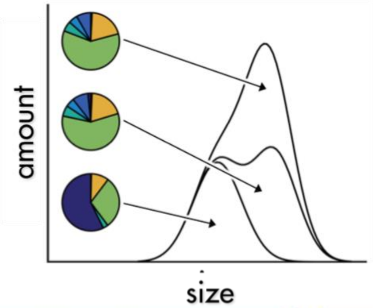
Fierce et al., Nat. Comm. 2016

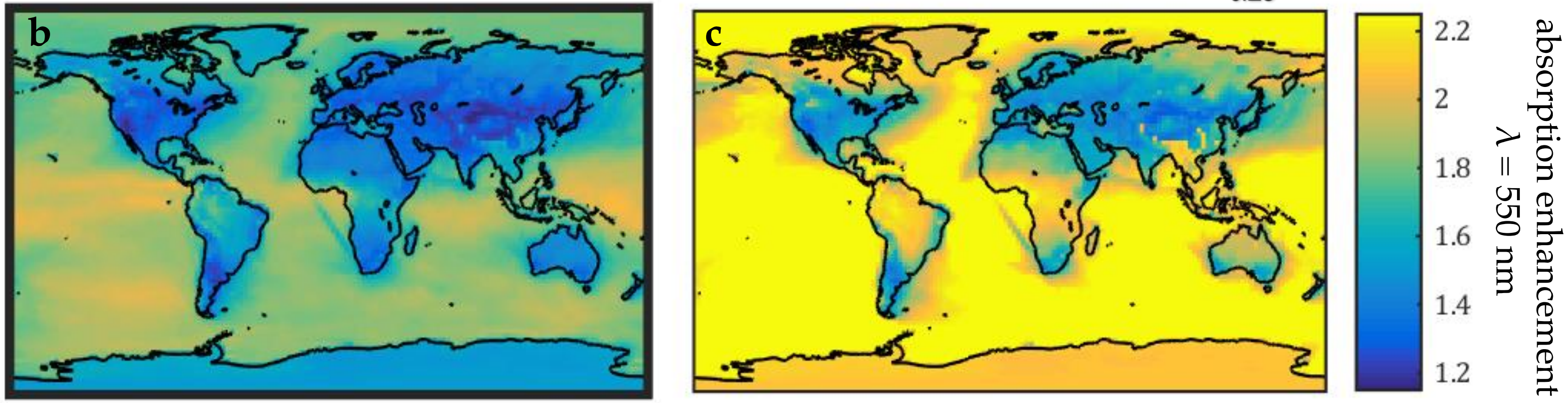
Surrogate model can improve predictions of climate-relevant aerosol properties …

Account for compositional diversity



Default: assume uniform composition across each mode





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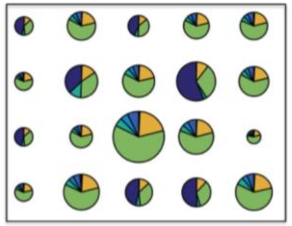

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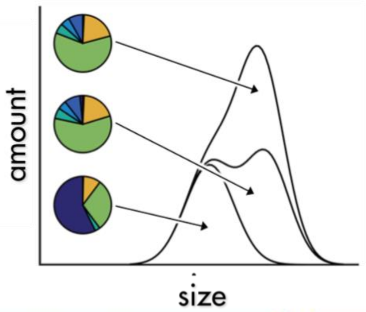
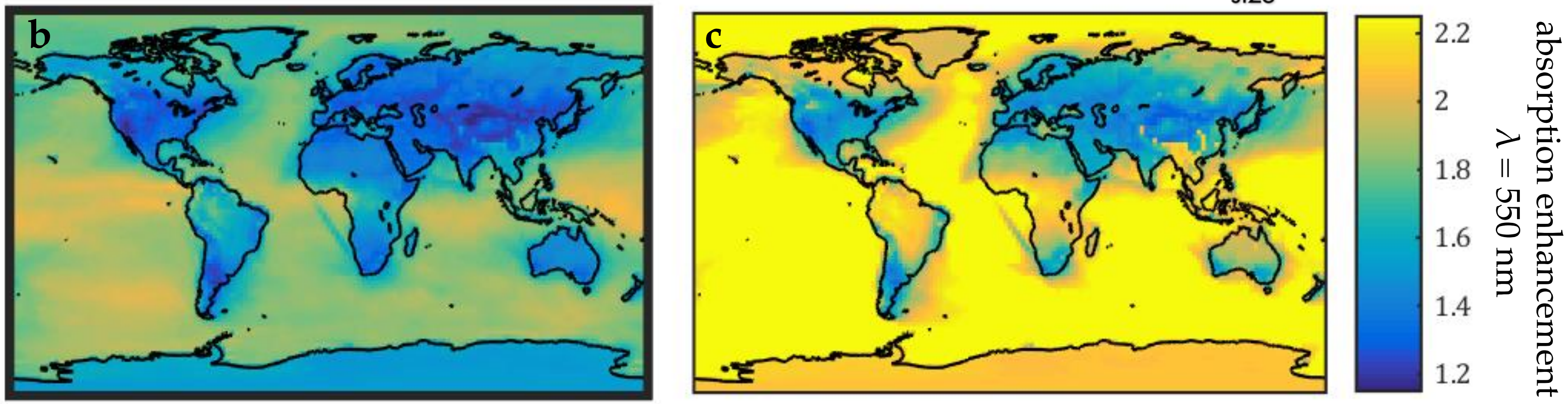
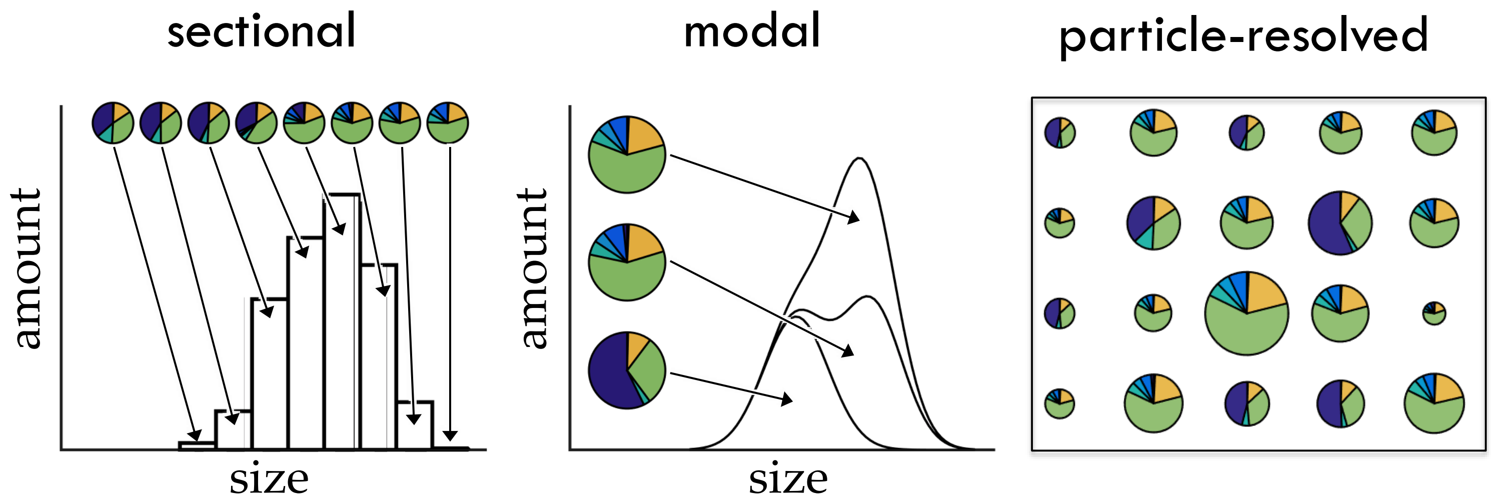




… if those models track the right information.

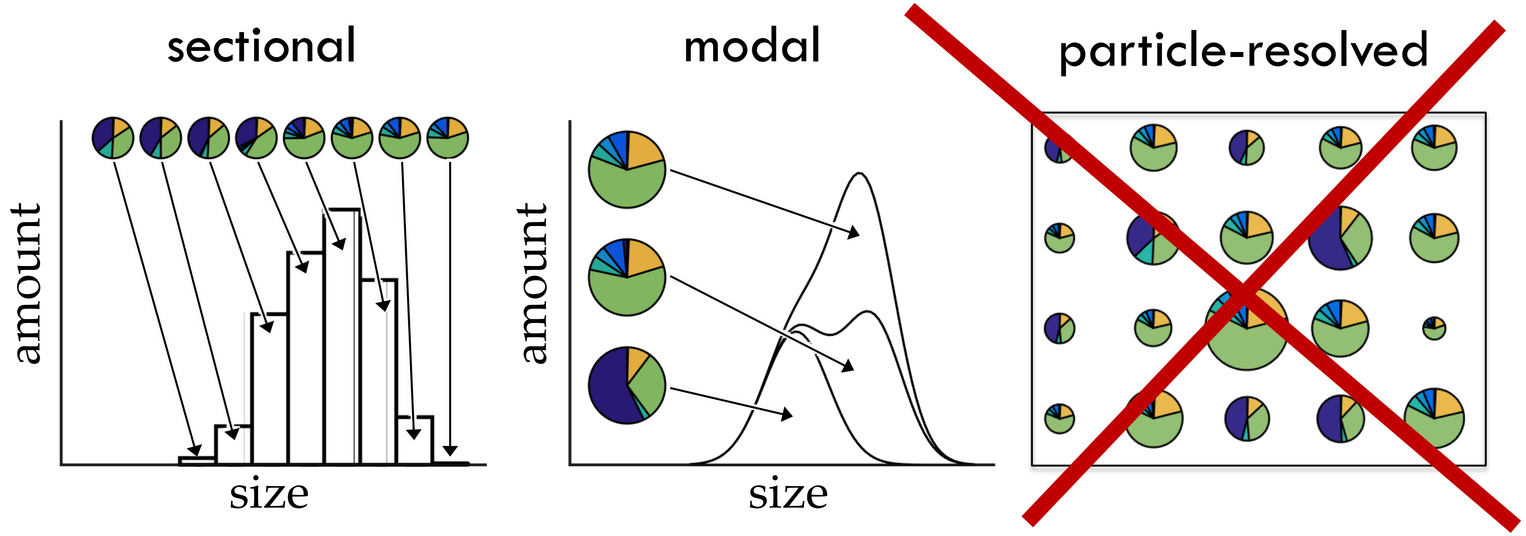
Fierce et al., Nat. Comm. 2016

Need a different approach for simulating aerosol distributions



Current approaches for modeling atmospheric aerosol

Need a different approach for simulating aerosol distributions



Particle-resolved scheme is impractical for global simulations

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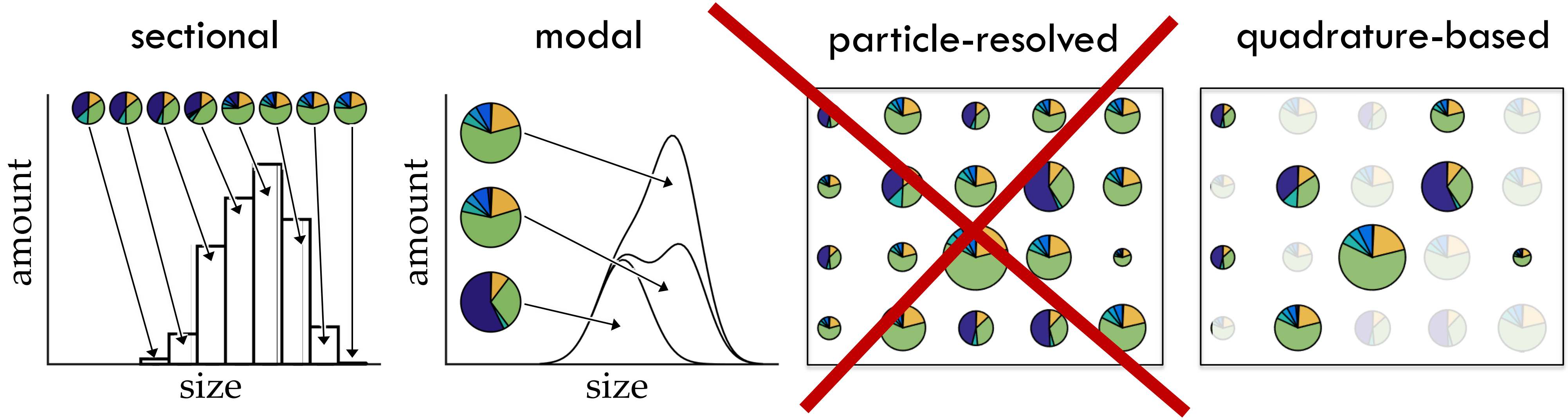
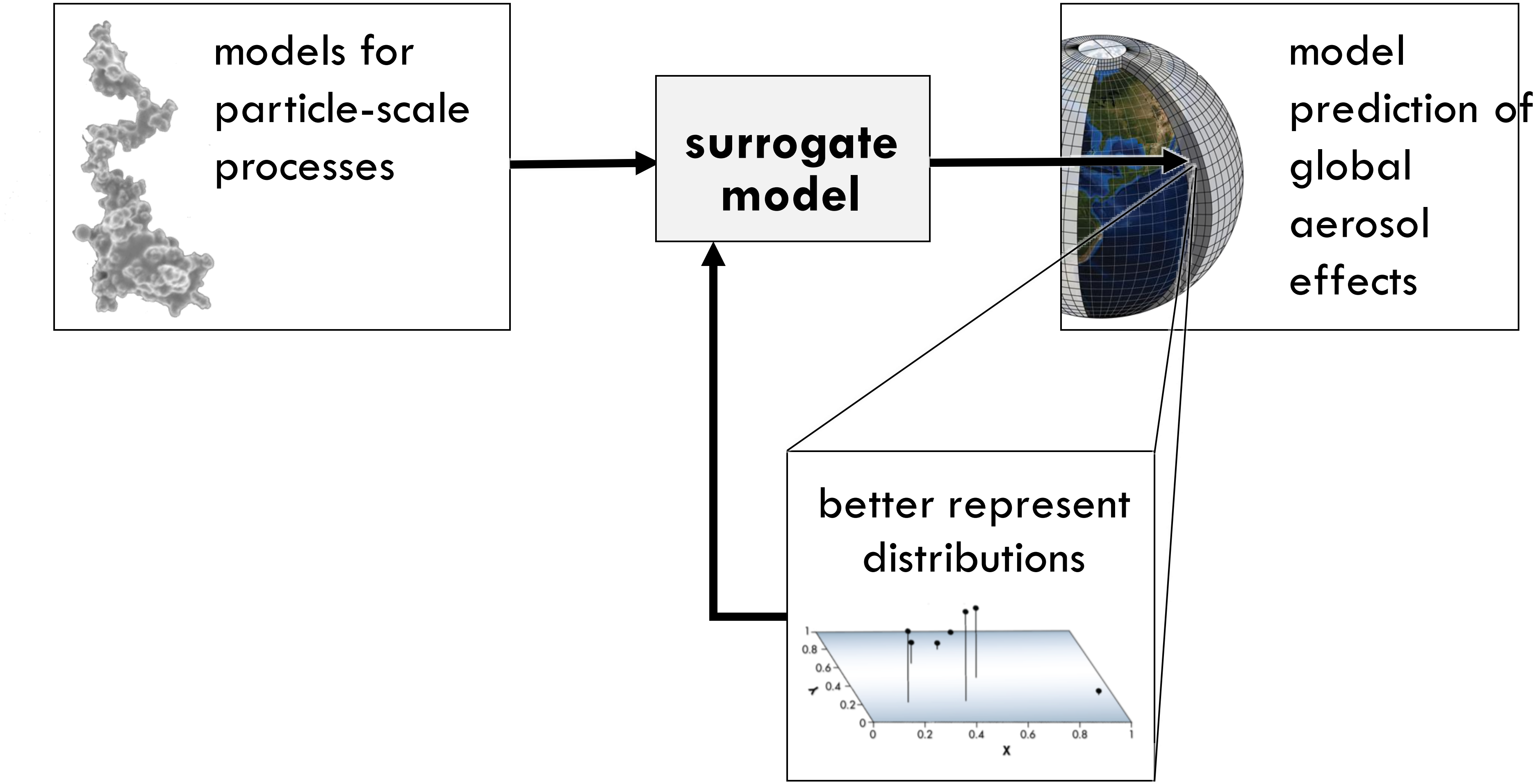
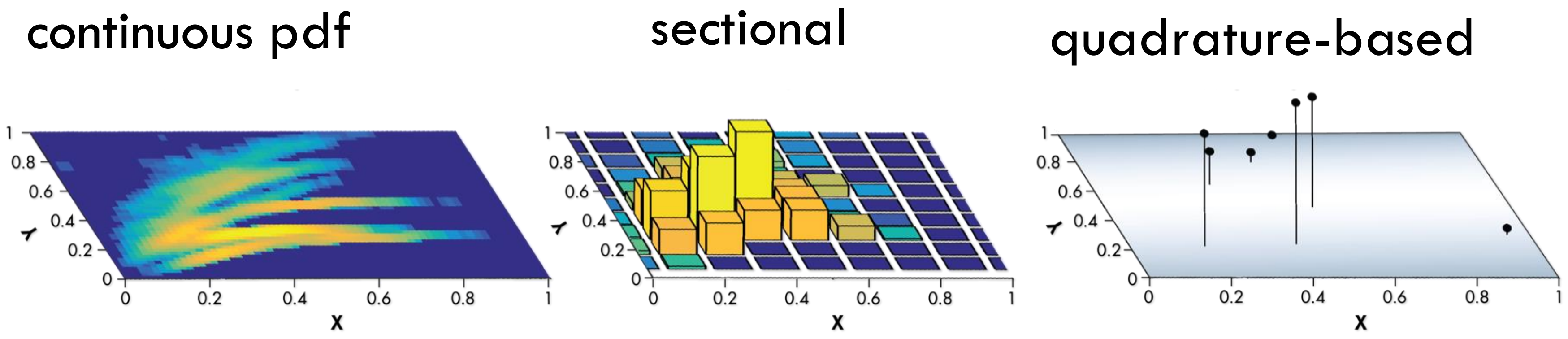


Path forward: combine surrogate models with quadrature-based aerosol scheme



Quadrature-based aerosol model is a balance between accuracy and computational efficiency



Fierce and McGraw, JGR 2017

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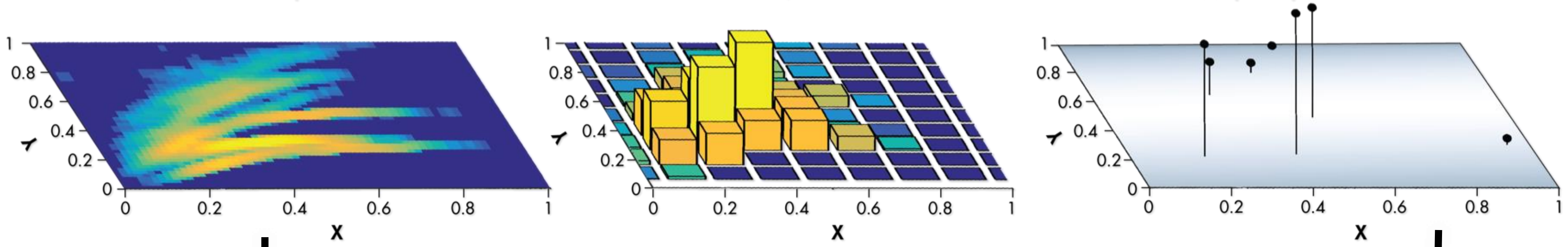
continuous pdf　　　sectional　　　quadrature-based



$$\mu_k = \int_0^\infty \int_0^\infty \dots \int_0^\infty n(x_1, x_2, \dots, x_A)\,\phi_k(x_1, x_2, \dots, x_A)\, dx_1\, dx_2 \dots dx_A \approx \sum_i^N N_i\,\phi_k(x_{1,i}, x_{2,i}, \dots, x_{A,i})$$

Fierce and McGraw, JGR 2017

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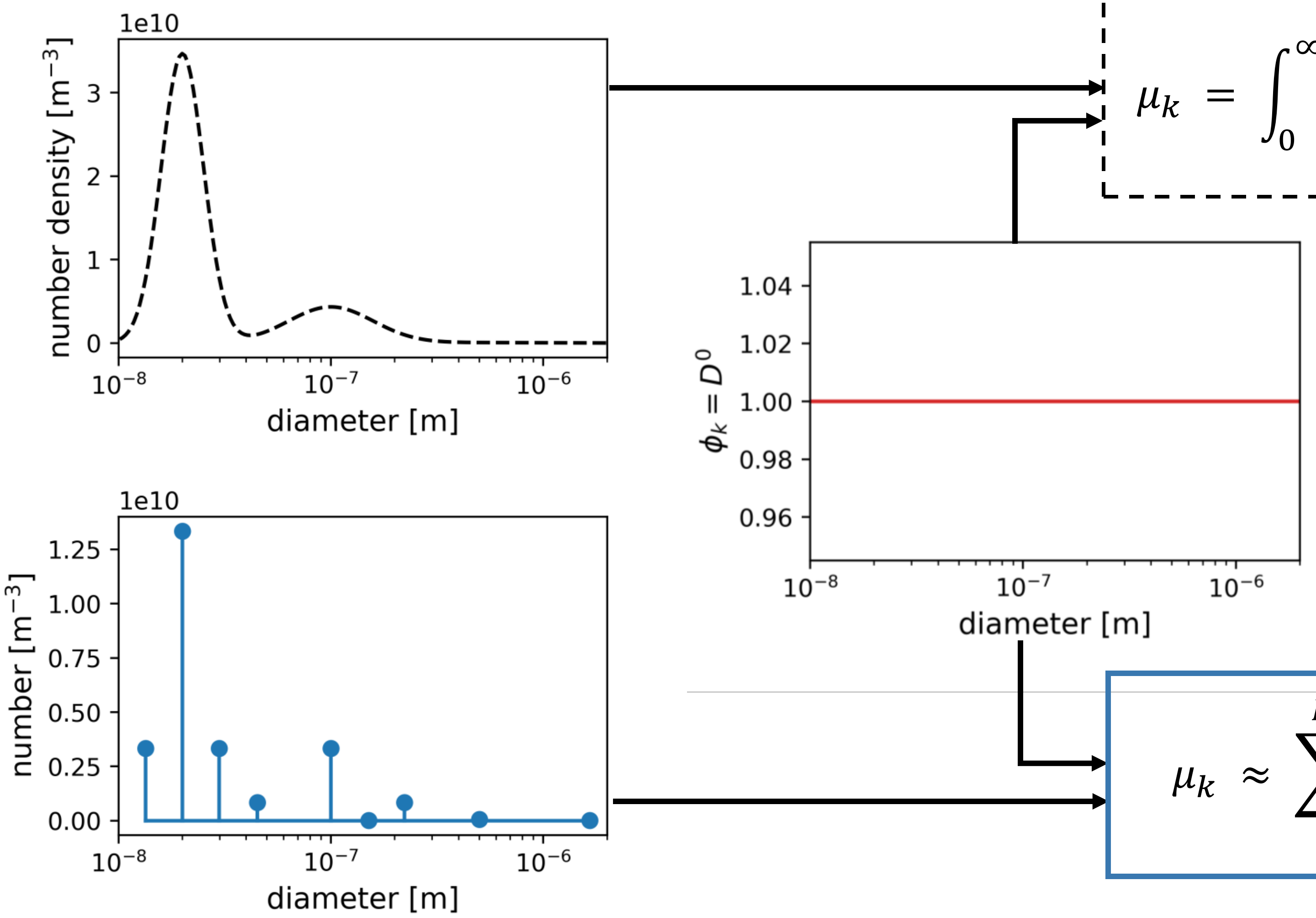
Quadrature points accurately approximate moments of the underlying aerosol distribution



$$\mu_k = \int_0^\infty n(D)\phi_k(D)\,dD$$

$$\mu_k \approx \sum_i^N N_i \phi_k(D_i)$$

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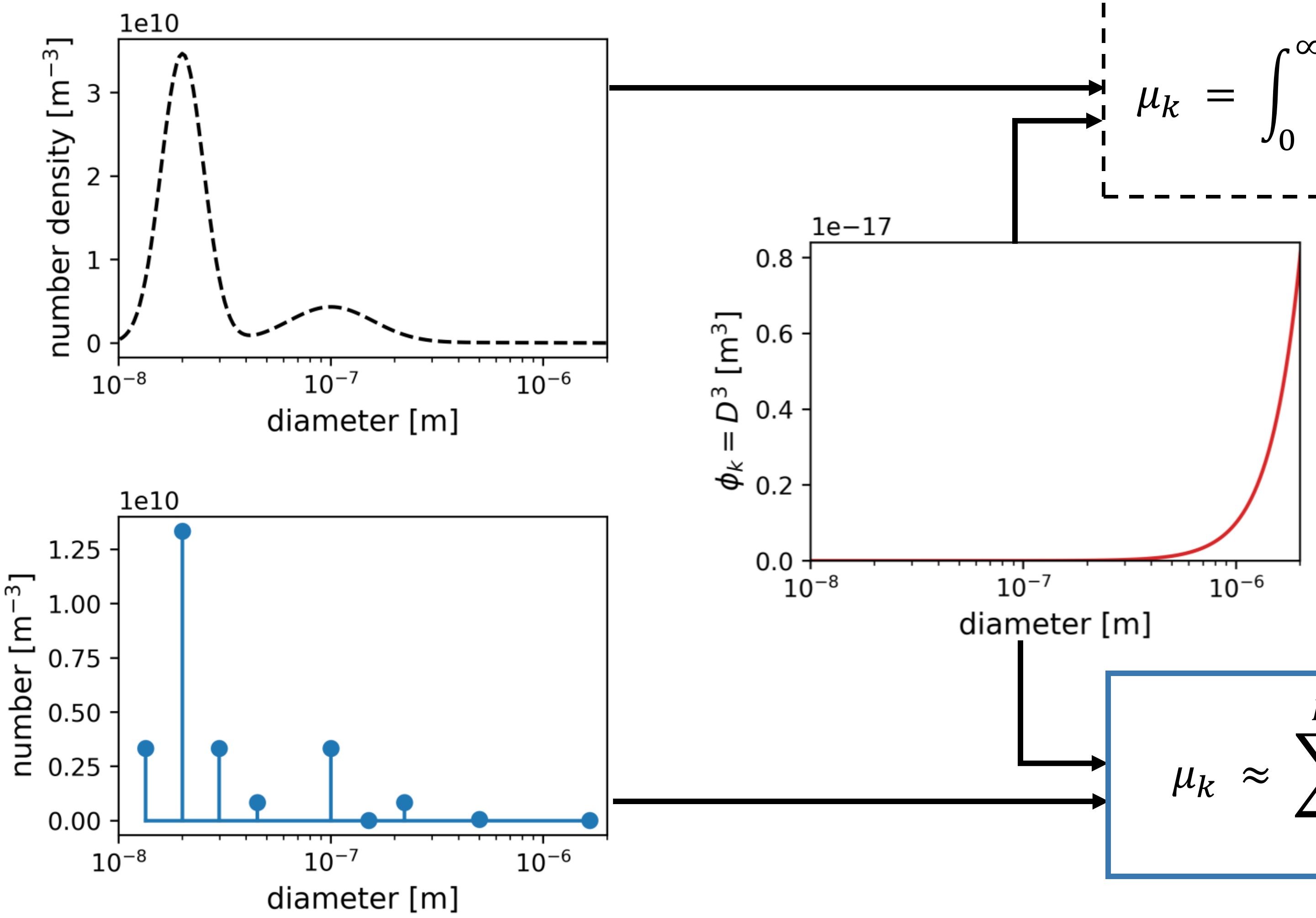

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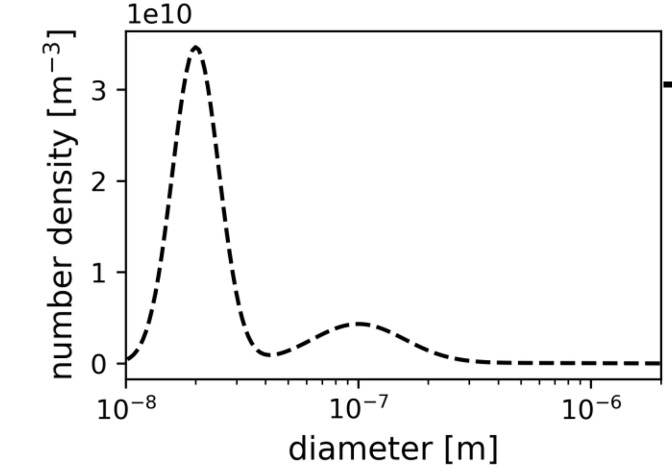
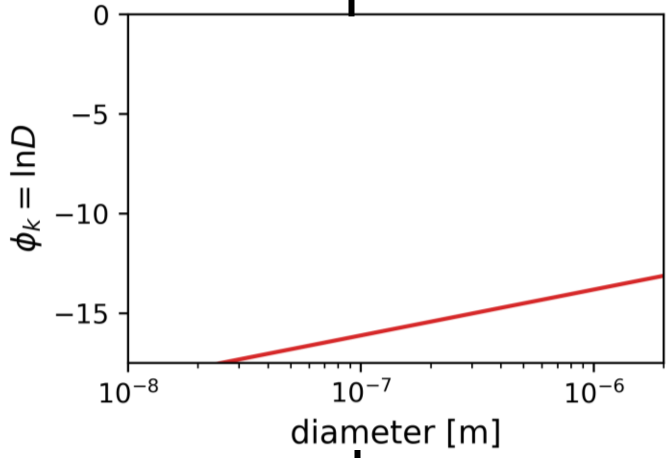
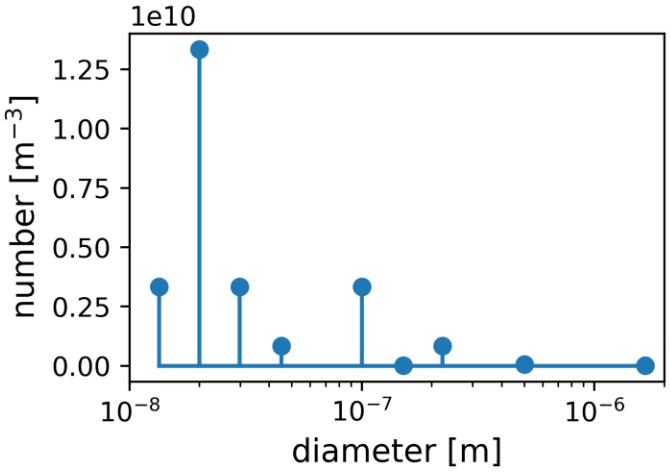


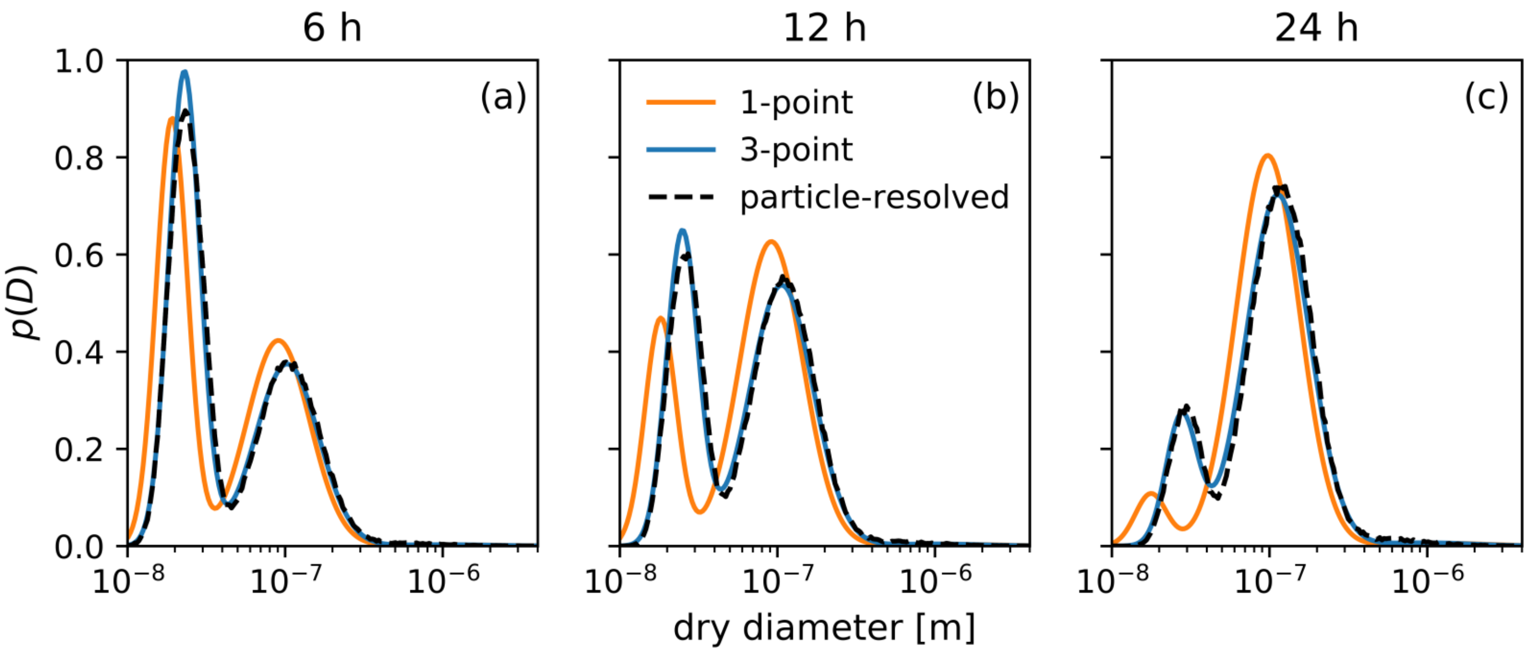
$$\mu_k = \int_0^\infty n(D)\phi_k(D)\, dD$$





$$\mu_k \approx \sum_i^N N_i \phi_k(D_i)$$

Quadrature points accurately approximate moments of the underlying aerosol distribution

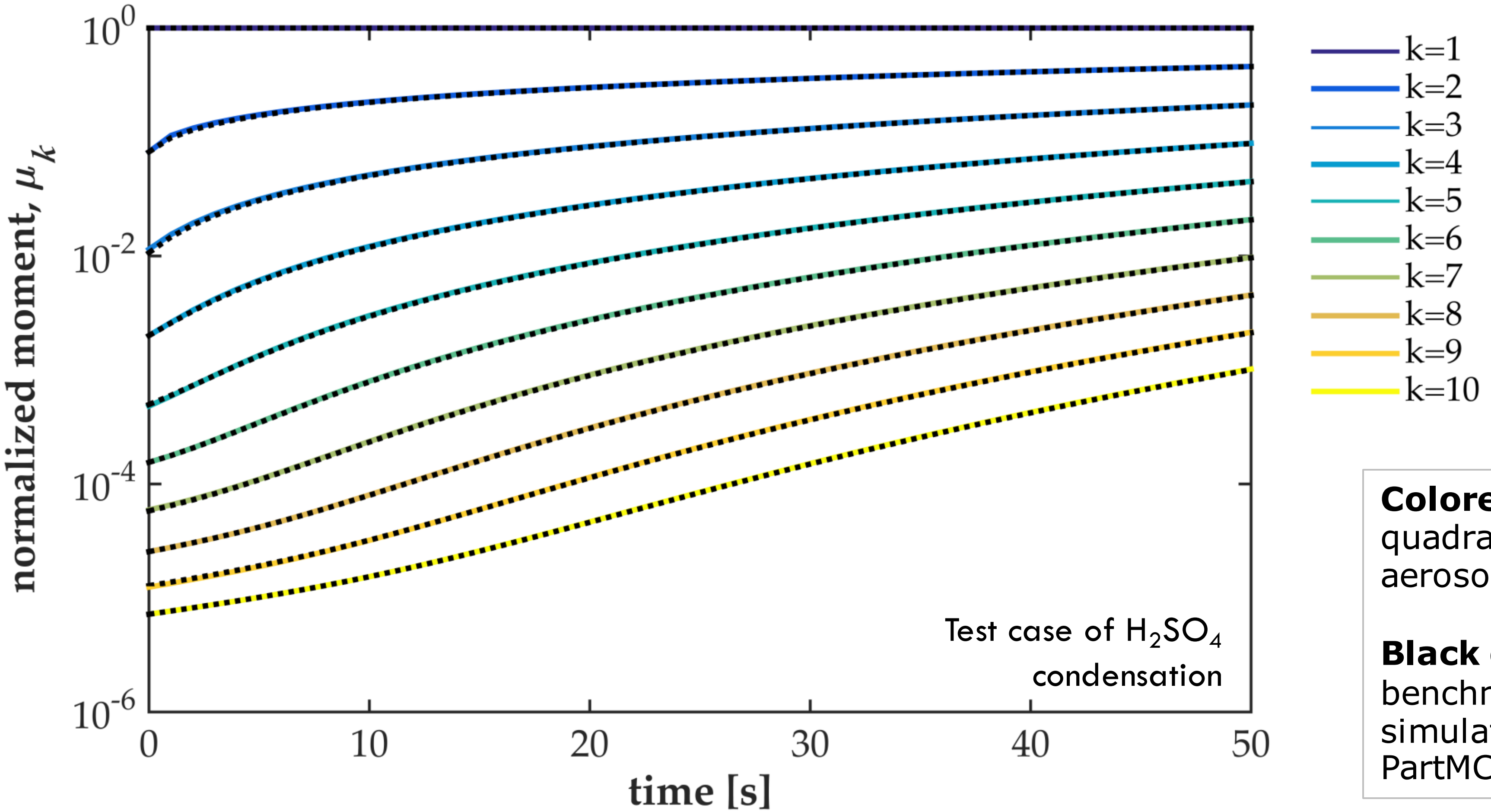


Distributions reconstructed from the moments

Test case of coagulation and emission

Fierce and McGraw (in prep)

Quadrature points accurately approximate moments of the underlying aerosol distribution



Colored lines: quadrature-based aerosol model

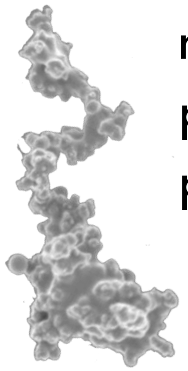
Black dashed: benchmark simulation with PartMC-MOSAIC

Fierce and McGraw (in prep)

How well do reduced aerosol schemes represent particle properties? **Not very well.**

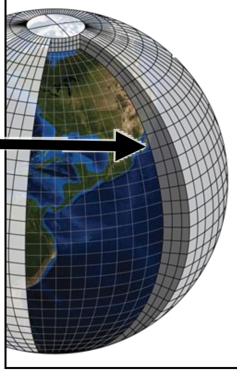
Can we do better? **Yes!**

Conclusion: the bridge between scales is a missing link in understanding aerosol effects



models for particle-scale processes

need to bridge scales by better representing particle heterogeneity



model prediction of global aerosol effects

laboratory studies to inform particle-scale models

field observations to evaluate and constrain global-scale models

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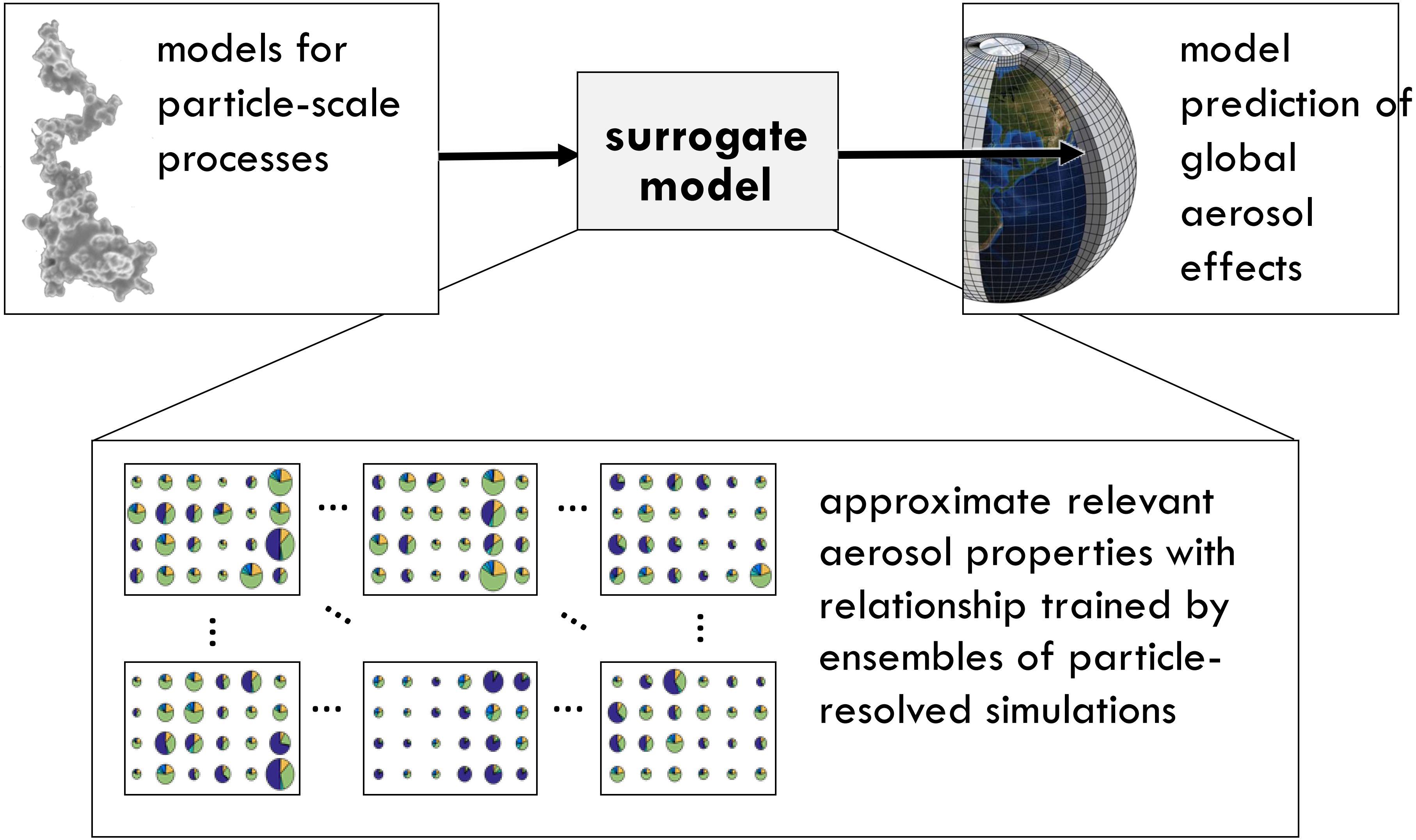
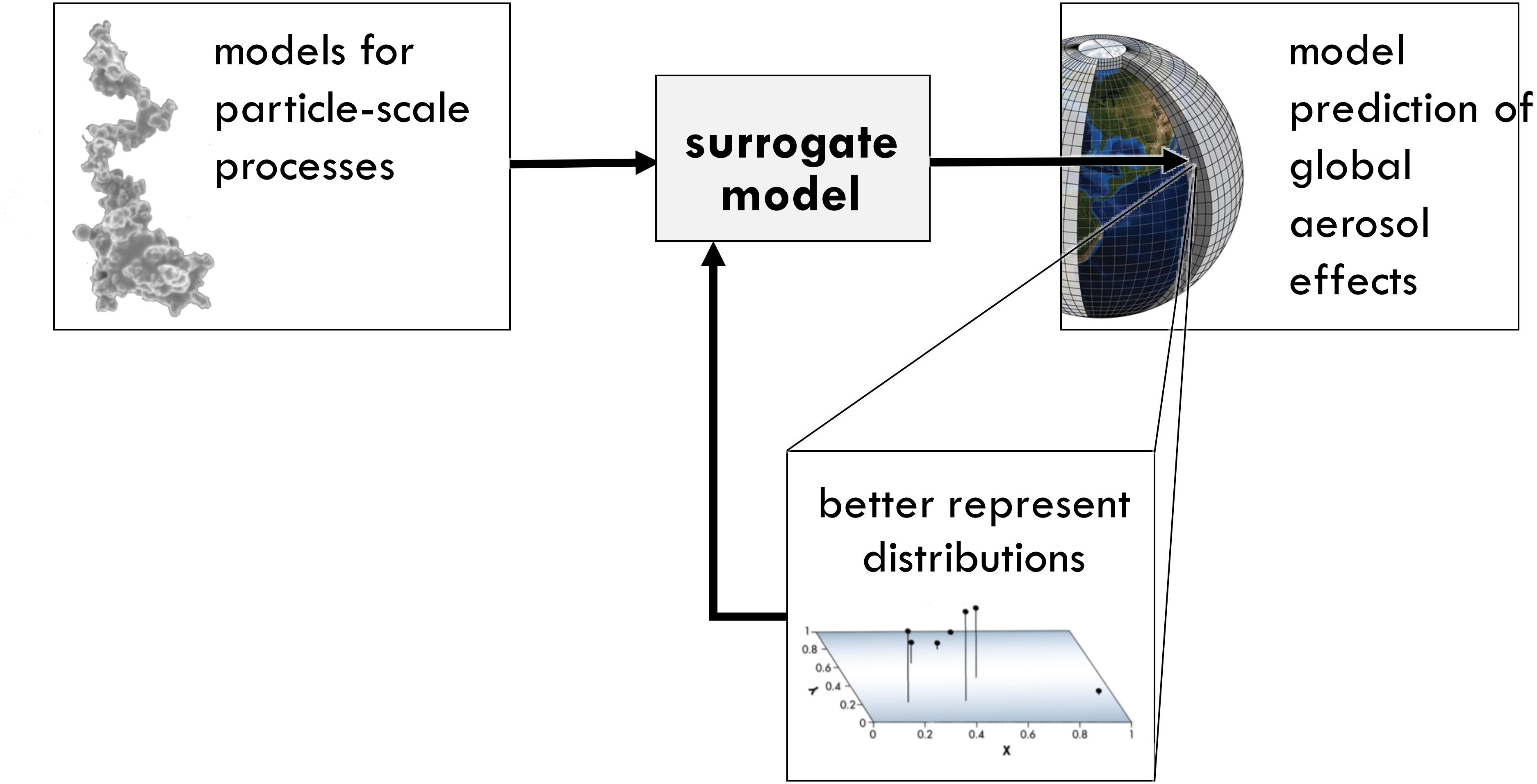
Path forward: surrogate models to bridge particle-scale and global-scale



Path forward: surrogate models to bridge particle-scale and global-scale



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Thank you!

Acknowledgements L. Fierce is funded by PNNL's Laboratory Development Research Program and the Atmospheric System Research and Earth System Modeling programs of the US Department of Energy.

U.S. DEPARTMENT OF ENERGY

Office of Science

Please contact me about opportunities for graduate students: laura.fierce@pnnl.gov

- Office of Science Graduate Student Research (SCGSR) Program: science.osti.gov/wdts/scgsr
- Student internships: careers.pnnl.gov

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Reminder: join us at lunch today for to brainstorm verification tests for aerosol models

Please provide your input in this short survey!

Questions? Ideas?

Contact AMBRS team!

laura.fierce@pnnl.gov

murphy.benjamin@epa.gov

nriemer@Illinois.edu

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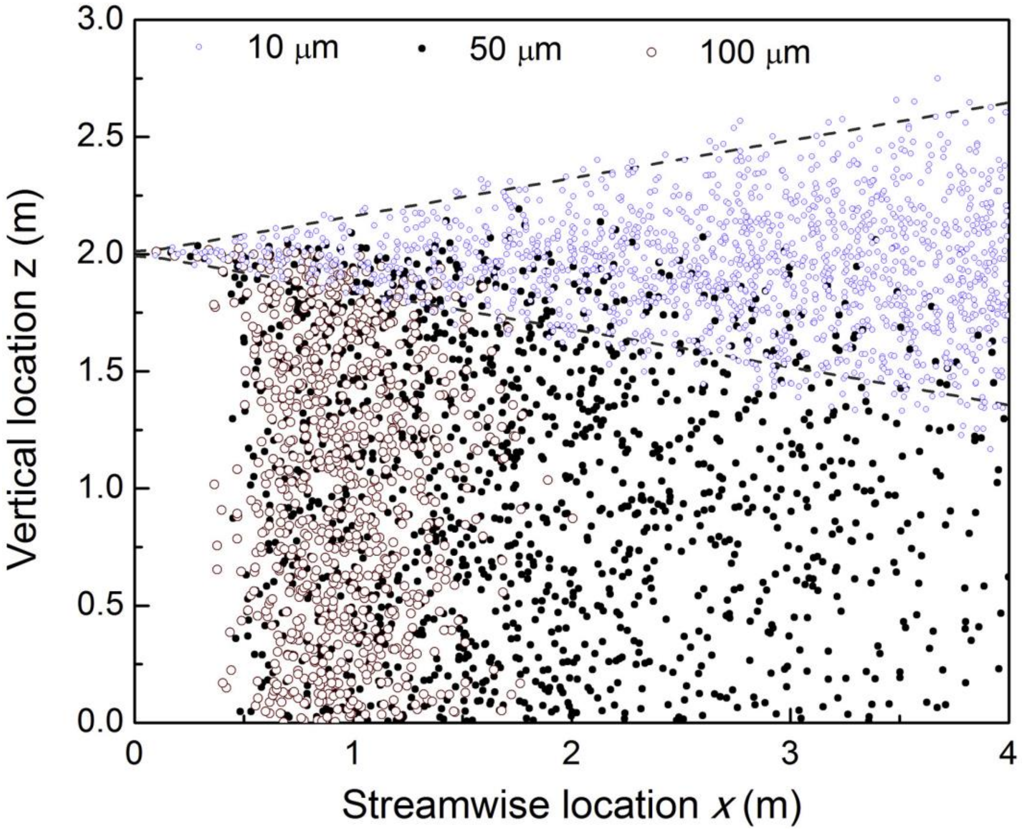
Inspiration: discrete random walk model



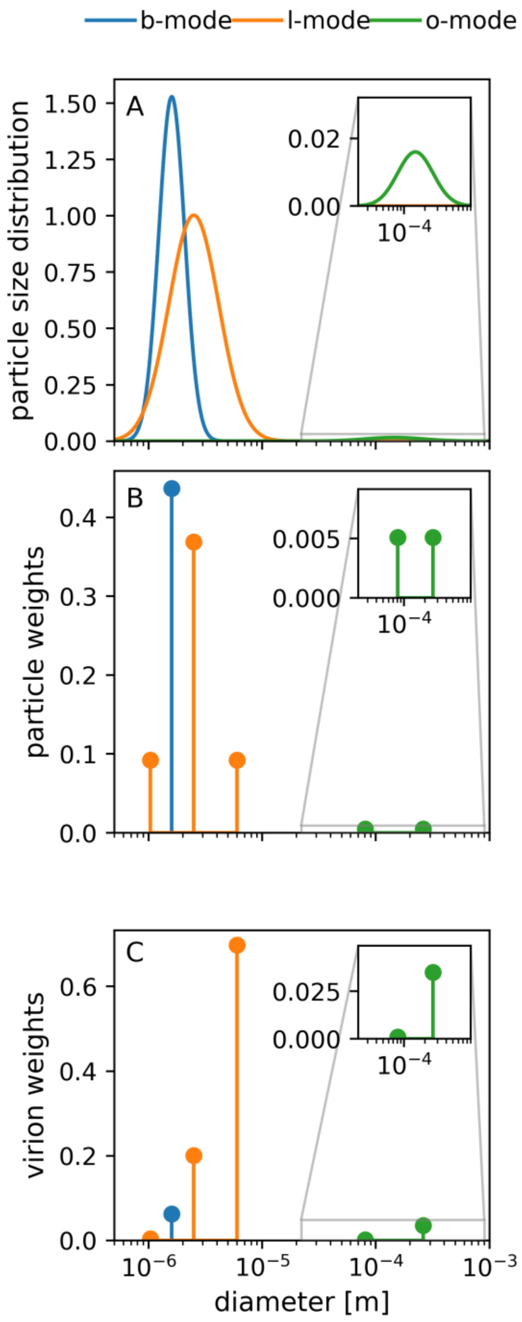
Problem: Need large ensembles to represent uncertainty in relevant processes but tracking thousands of Monte Carlo model of particle evolution and dispersion is expensive!

Solution: Quadrature-based model of Respiratory Aerosol and Droplets (QuaRAD)

Wei, Jianjian, and Yuguo Li. "Enhanced spread of expiratory droplets by turbulence in a cough jet." *Building and Environment* 93 (2015): 86-96.

Efficient quadrature-based[1] representation

- Continuous respiratory aerosol size distribution (top) from **Morawska group**[2] is represented by efficient quadrature approximation (middle)
- Virion weights (bottom) computed from quadrature points and measurements of pathogen loading with respect to particle size for influenza from **Milton group**[3].



1. McGraw, R. (1997). Description of aerosol dynamics by the quadrature method of moments. *Aerosol Science and Technology*, *27*(2), 255-265.
2. Johnson, G. R., et al. "Modality of human expired aerosol size distributions." *Journal of Aerosol Science* 42.12 (2011): 839-851.
3. Milton, D. K., et al. "Influenza virus aerosols in human exhaled breath: particle size, culturability, and effect of surgical masks." *PLoS pathogens* 9.3 (2013): e1003205.

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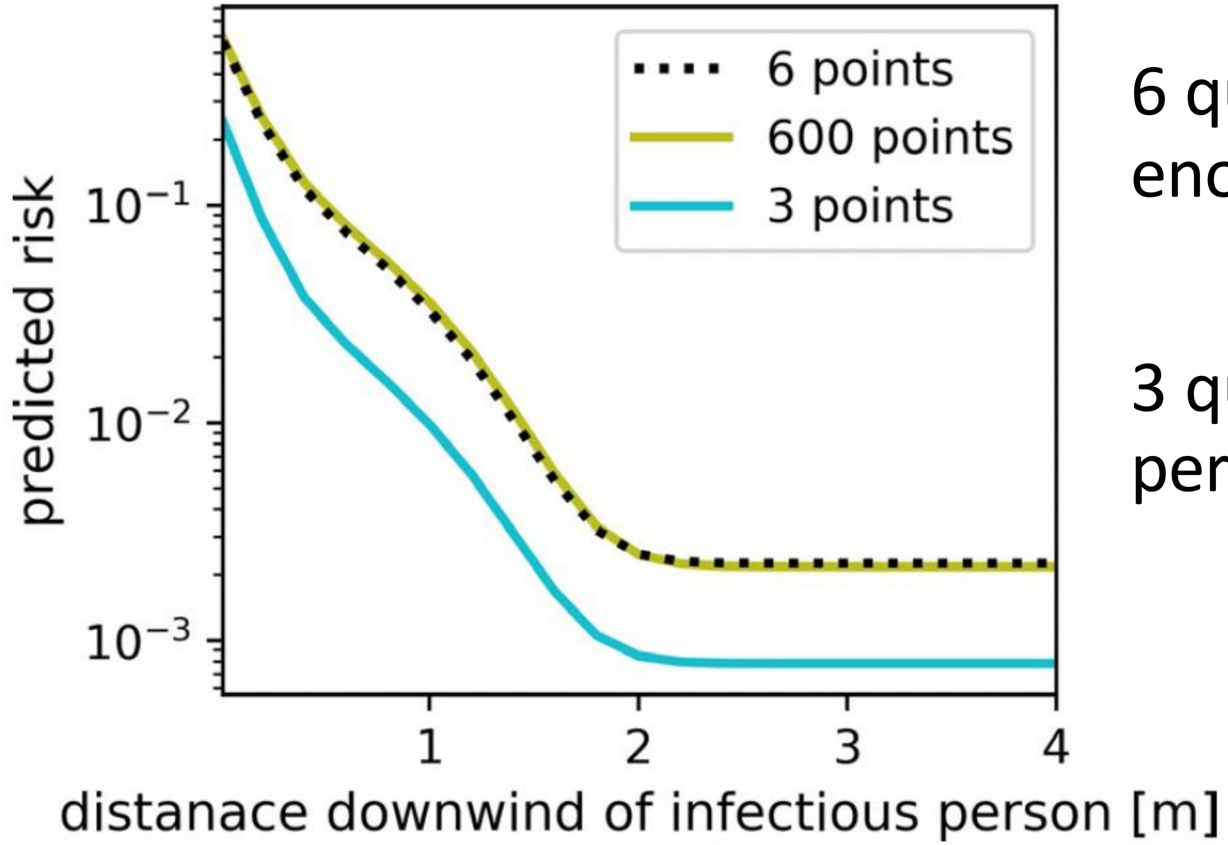
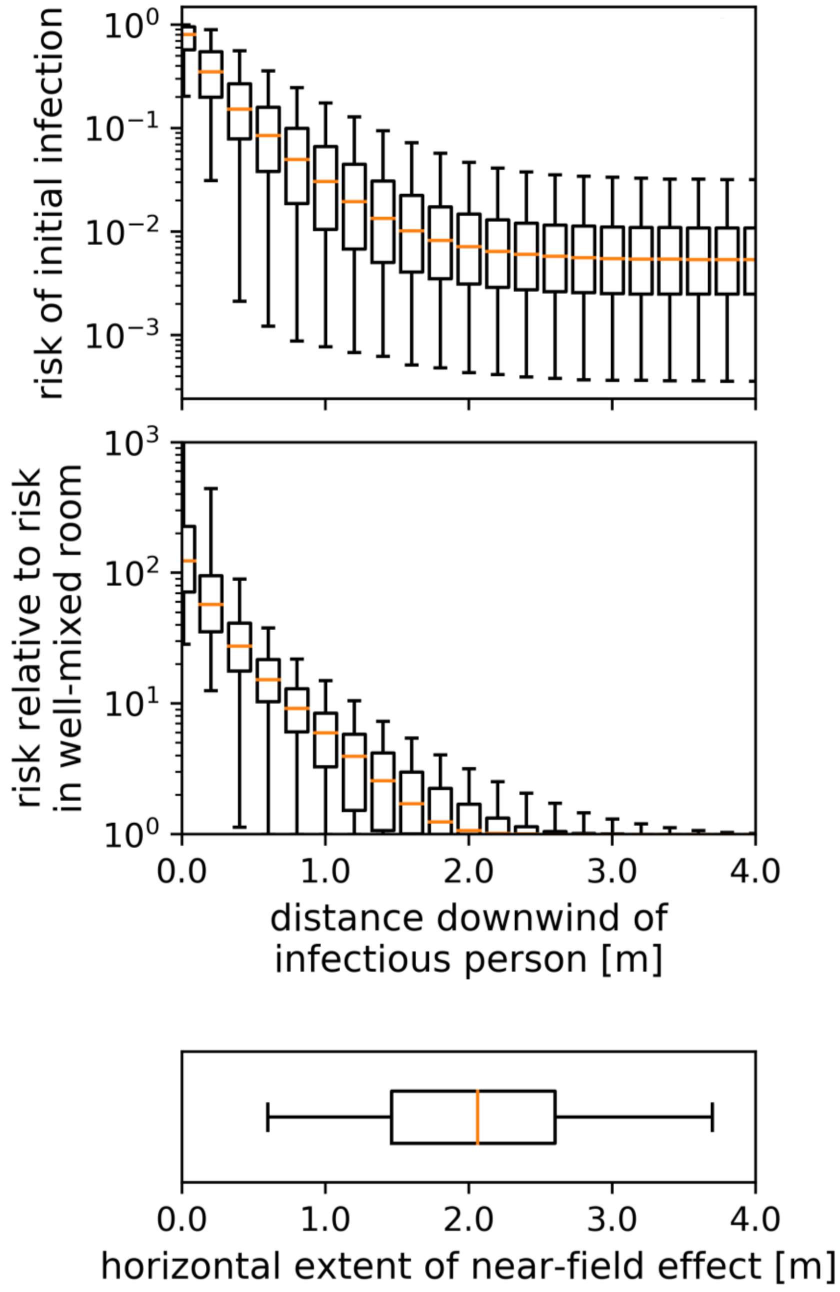
Increasing number of quadrature points does not improve accuracy.



6 quadrature points is enough

3 quadrature points (1 per mode) is not.



Risk of transmission enhanced by orders of magnitude near infectious person

Horizontal extent of near-field effect is highly variable, with median value of ~2 m.

Sensitivity analysis revealed variability in horizontal extent is driven by variability in expiration velocity.

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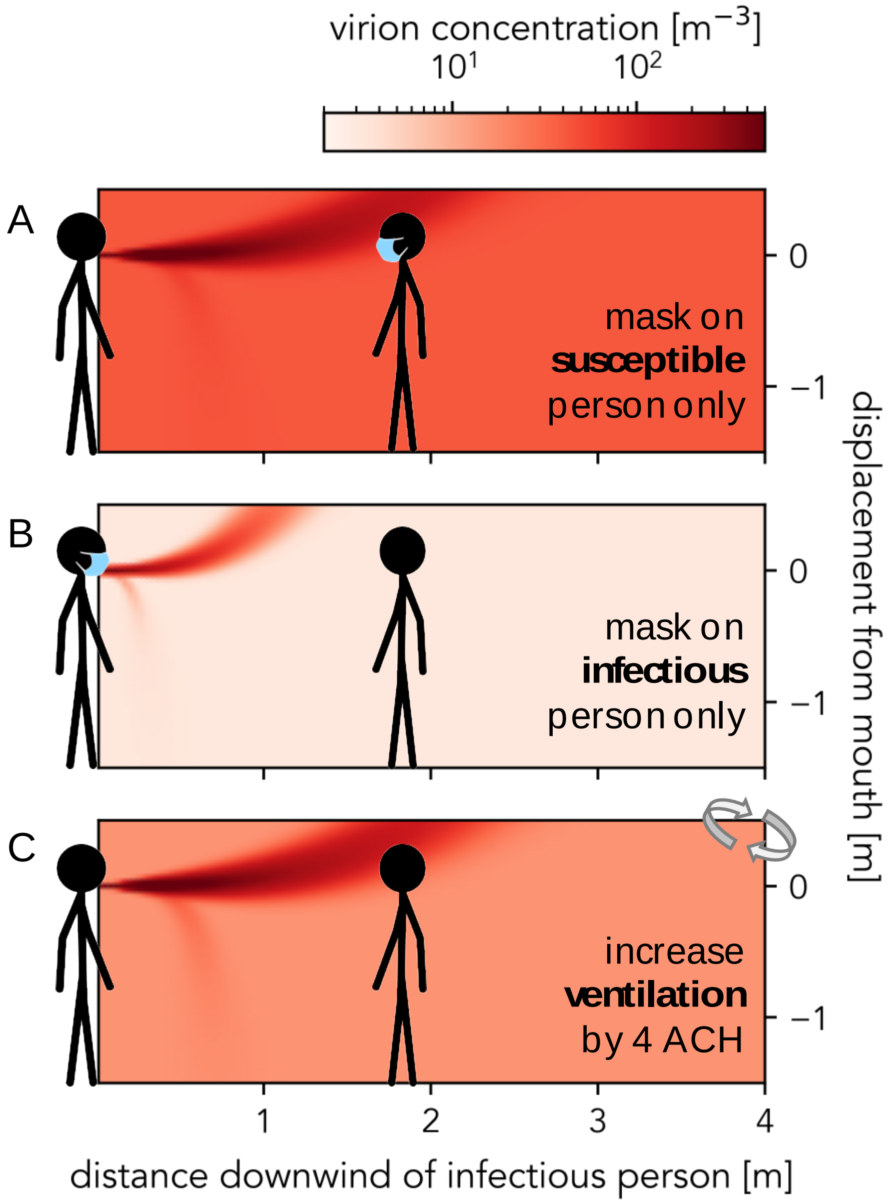
Framework for evaluating layered controls on airborne transmission



Fierce, L., Robey, A. J., & Hamilton, C. (2022). High efficacy of layered controls for reducing exposure to airborne pathogens. *Indoor air*, *32*(2), e12989.

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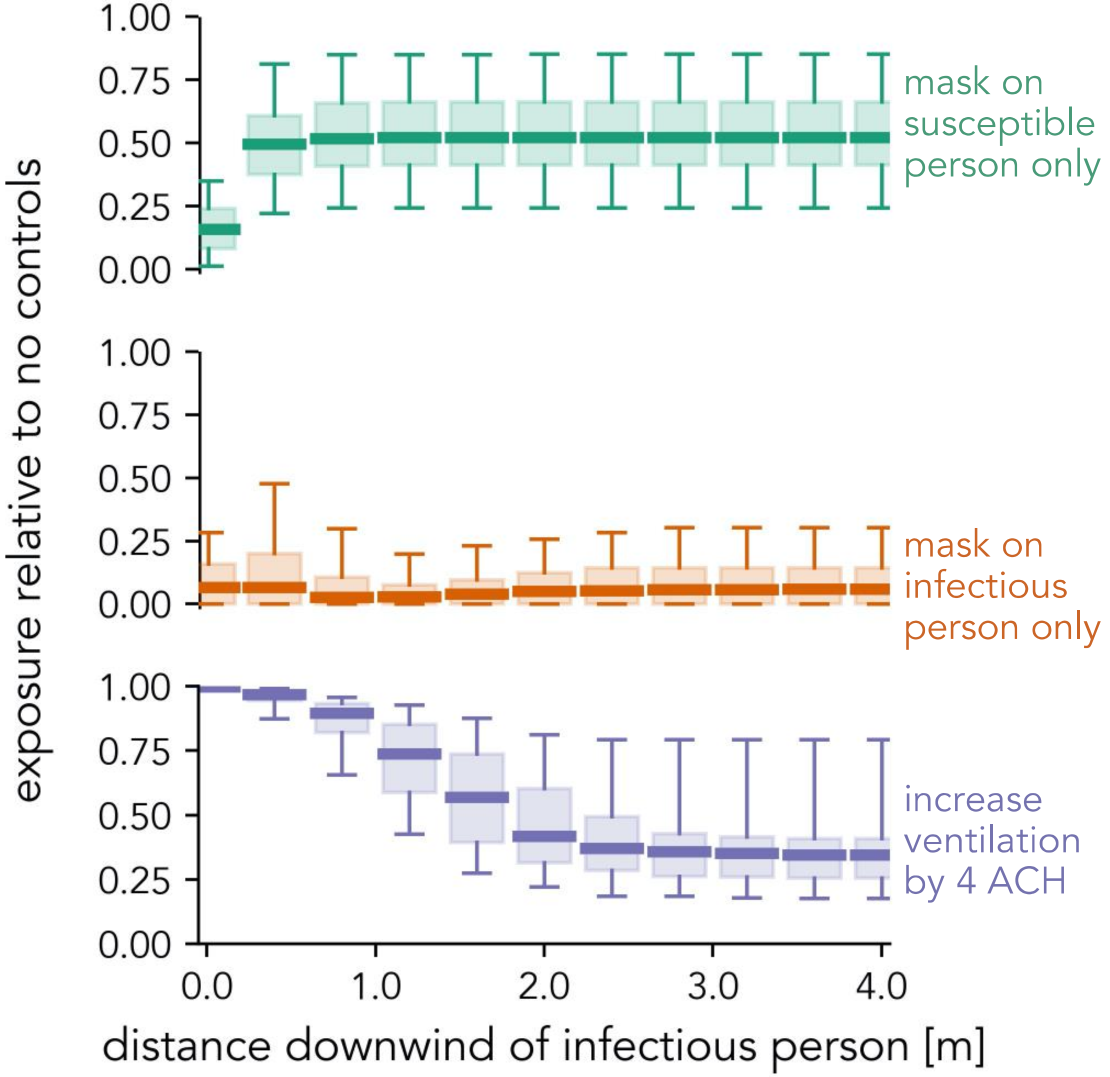
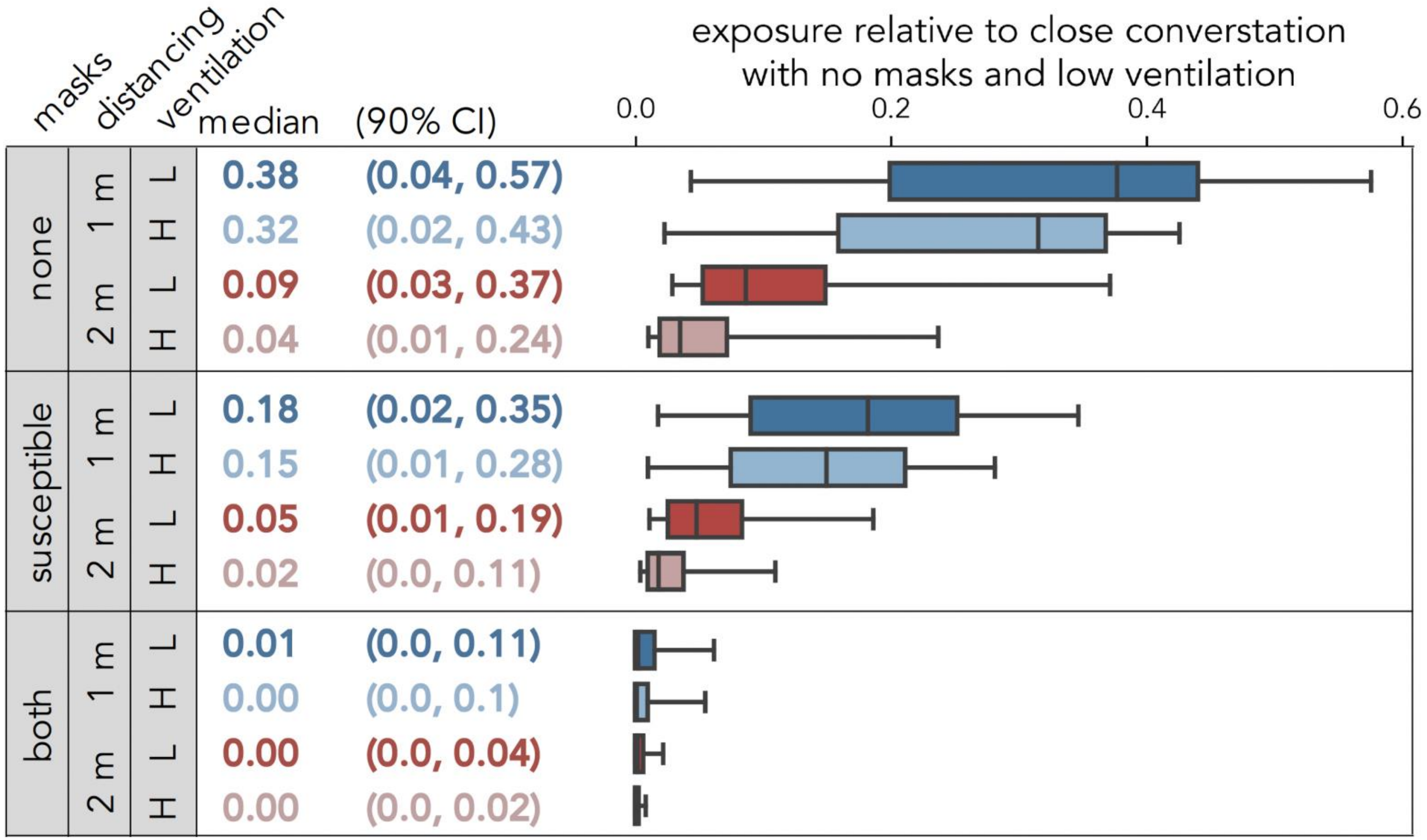


Fierce, L., Robey, A. J., & Hamilton, C. (2022). High efficacy of layered controls for reducing exposure to airborne pathogens. *Indoor air*, *32*(2), e12989.

Exposure reduced most effectively through layered controls



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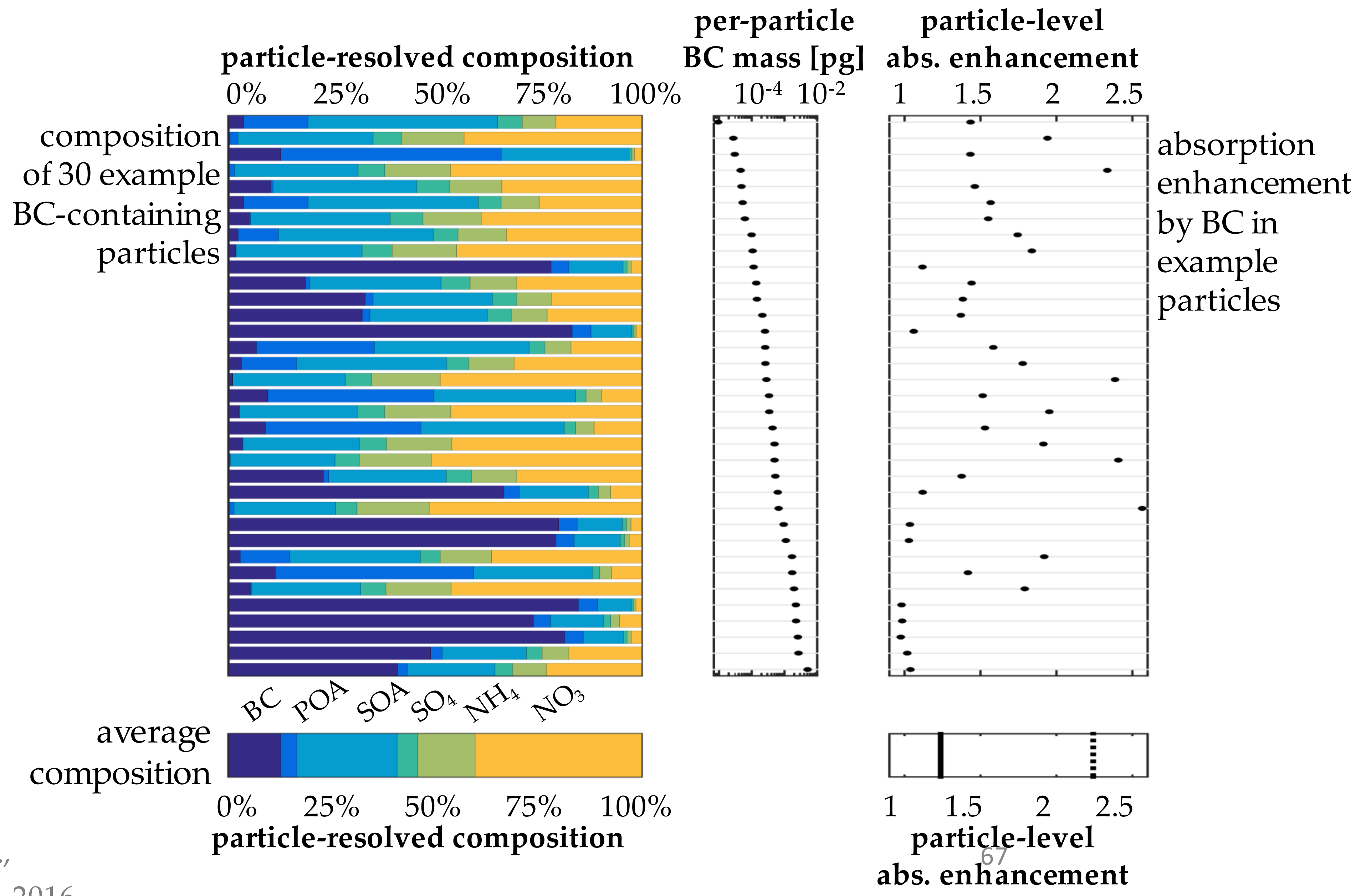
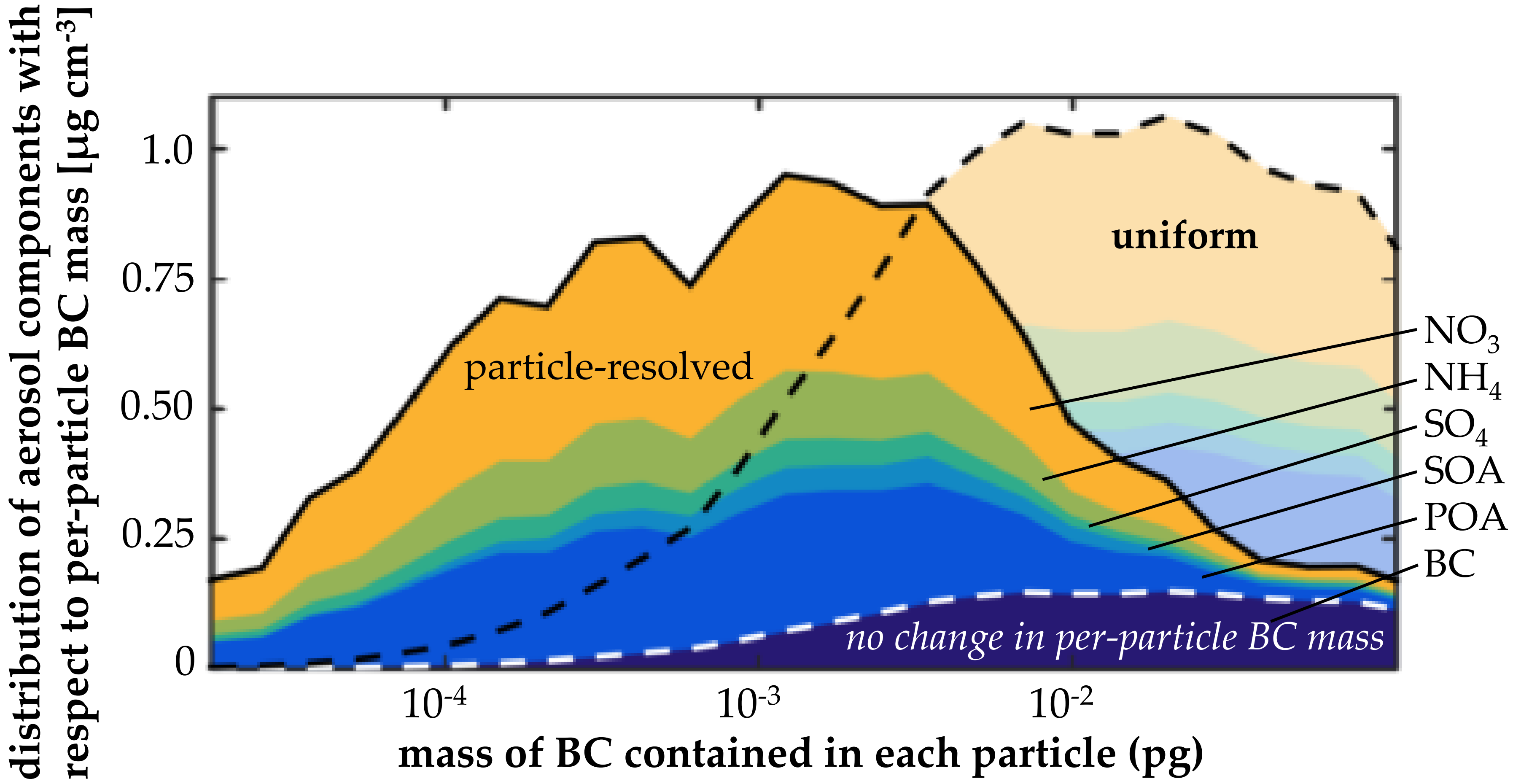


Fierce et al., *Nat. Comm.* 2016



Fierce et al.,
Nat. Comm. 2016

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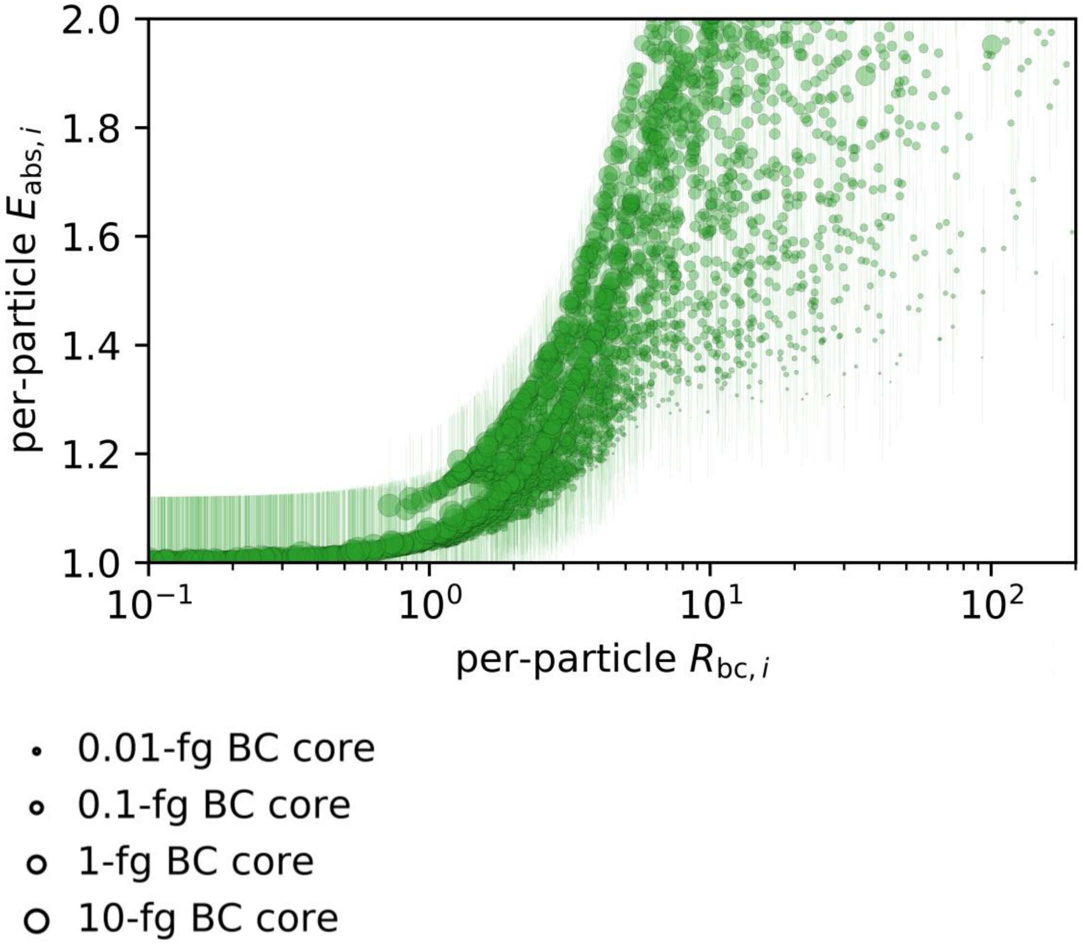
Particle-resolved model reveals variability in per-particle absorption enhancement



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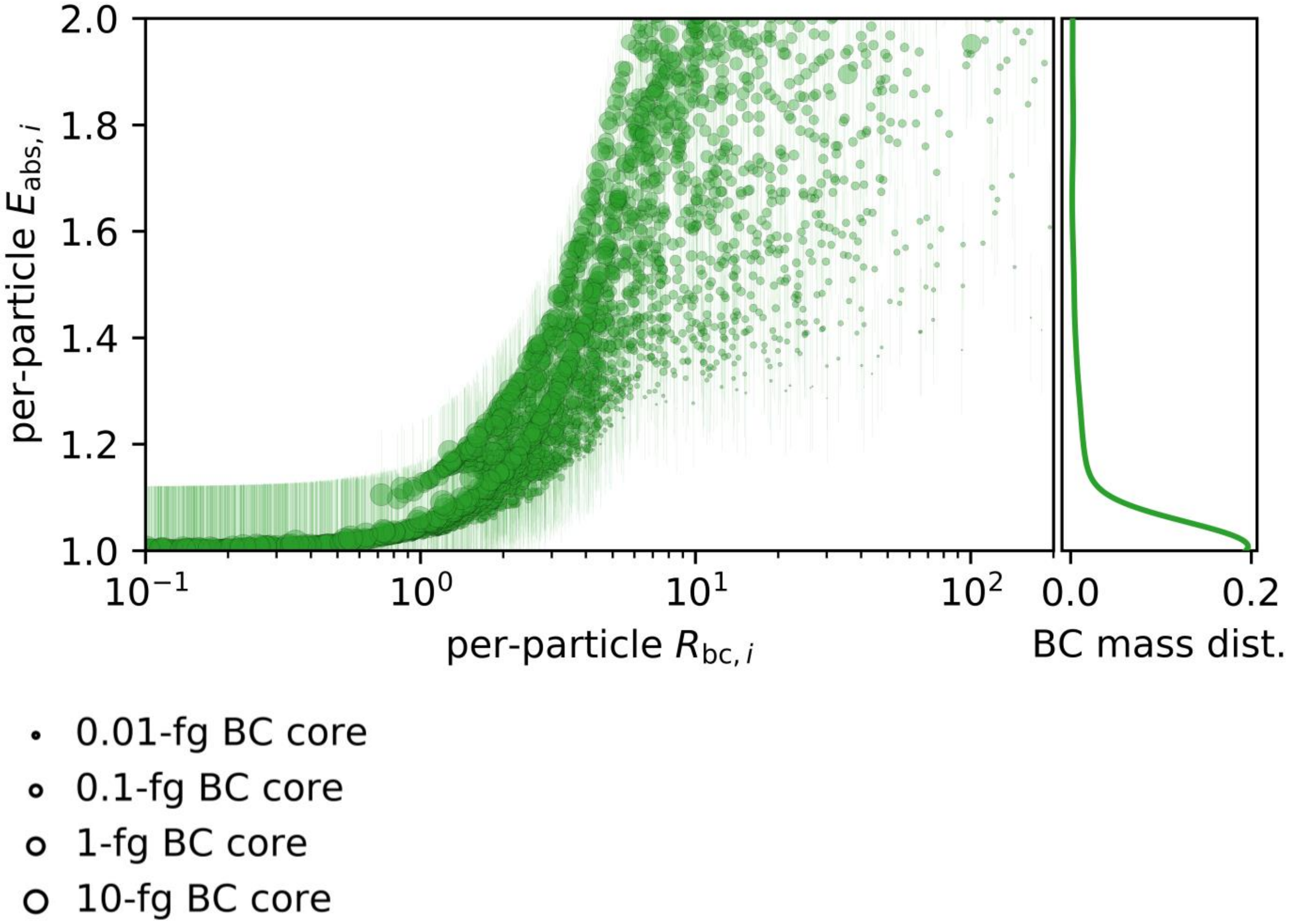

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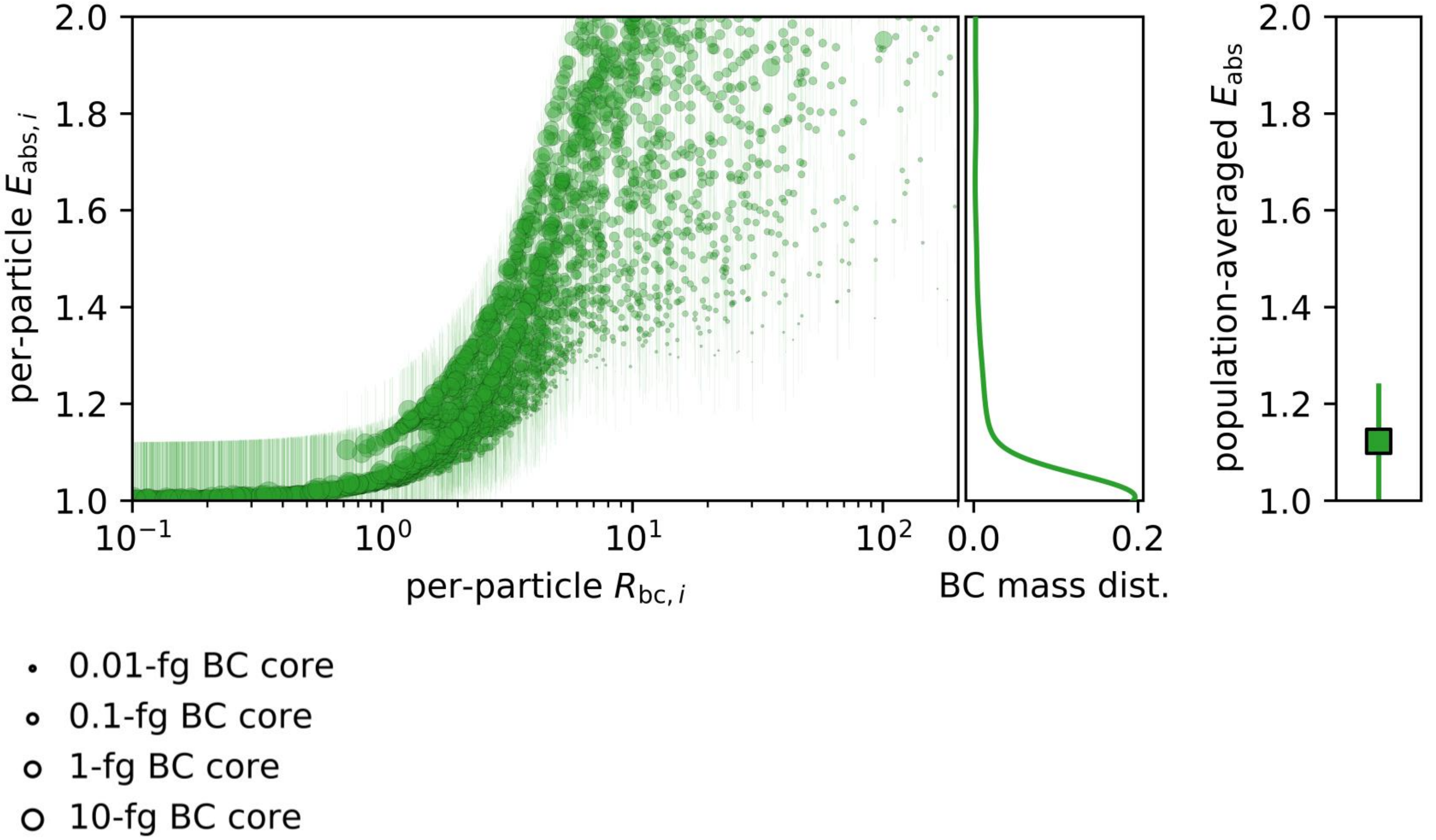

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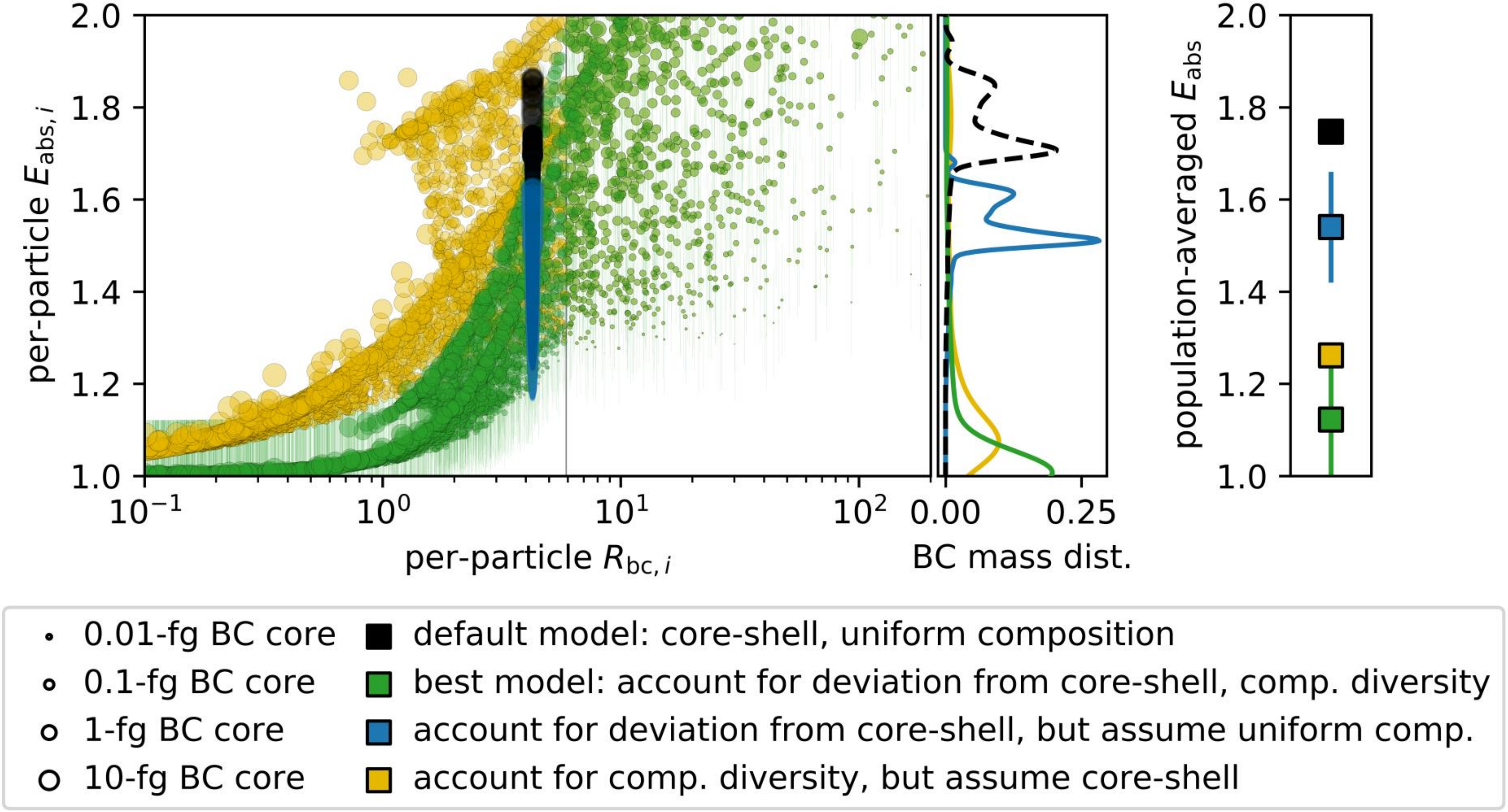
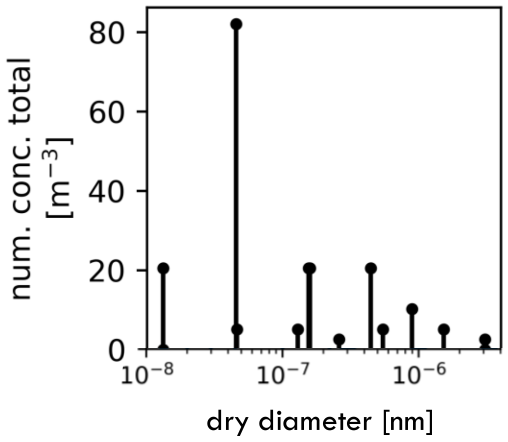


Path forward: quadrature-based aerosol scheme in large-scale atmospheric models

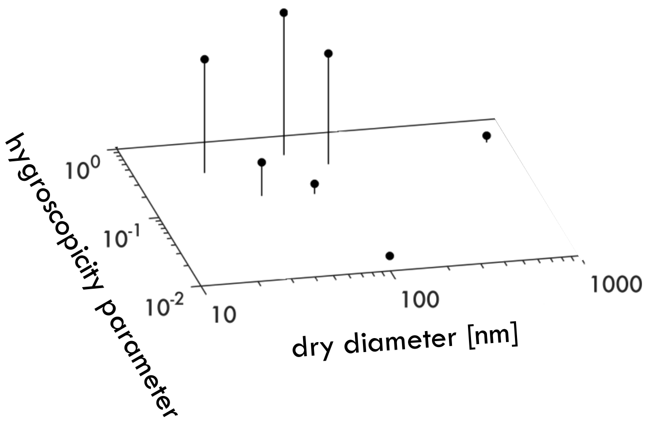
Univariate quadrature scheme

- Framework to efficiently represent aerosol size distributions, but simplified composition
- Implementation can be completed proces by process, without replacing scheme all at once



Multivariate quadrature scheme

- Framework to efficiently represent evolution of aerosol size-composition distributions
- Implementation in large-scale models would require complete overhaul of aerosol scheme



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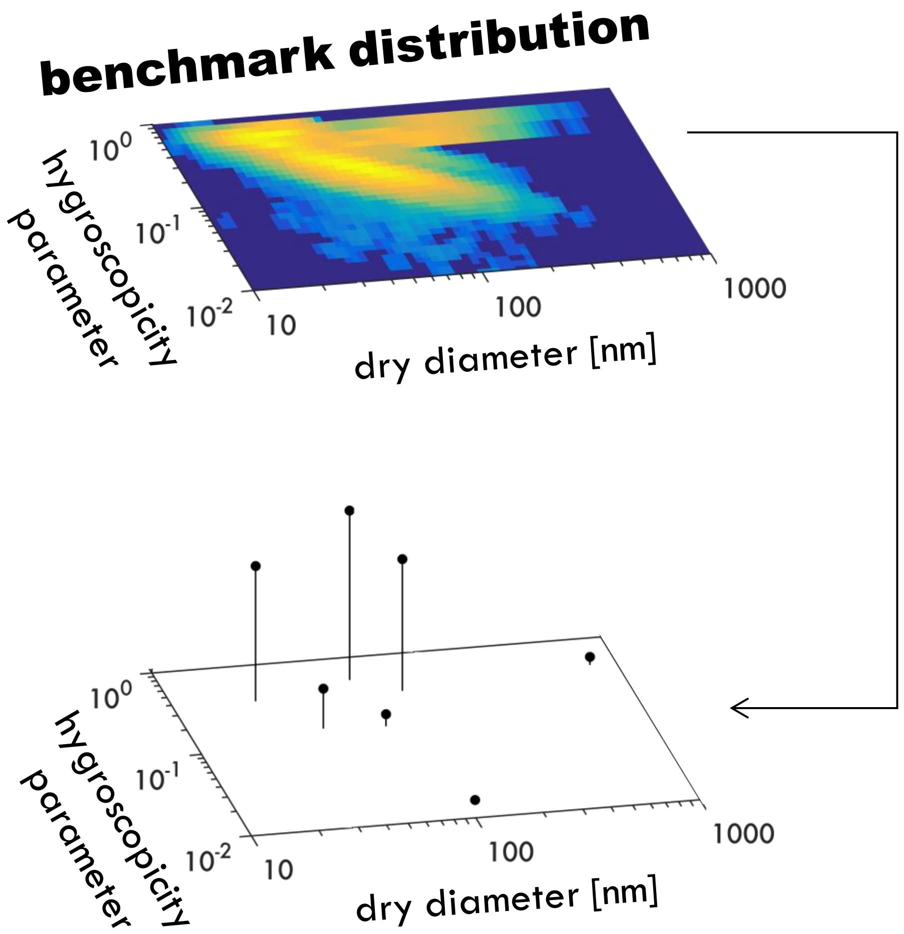
Accurate CCN spectra from quadrature



Quadrature approximation of particle size-composition distribution are constructed from multivariate moments

Fierce and McGraw, JGR 2017

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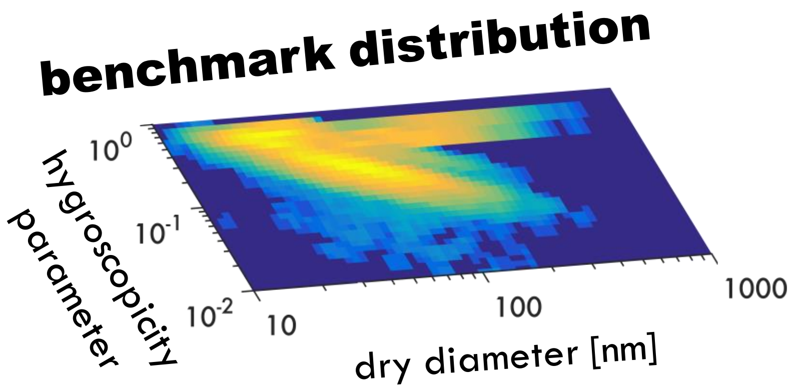
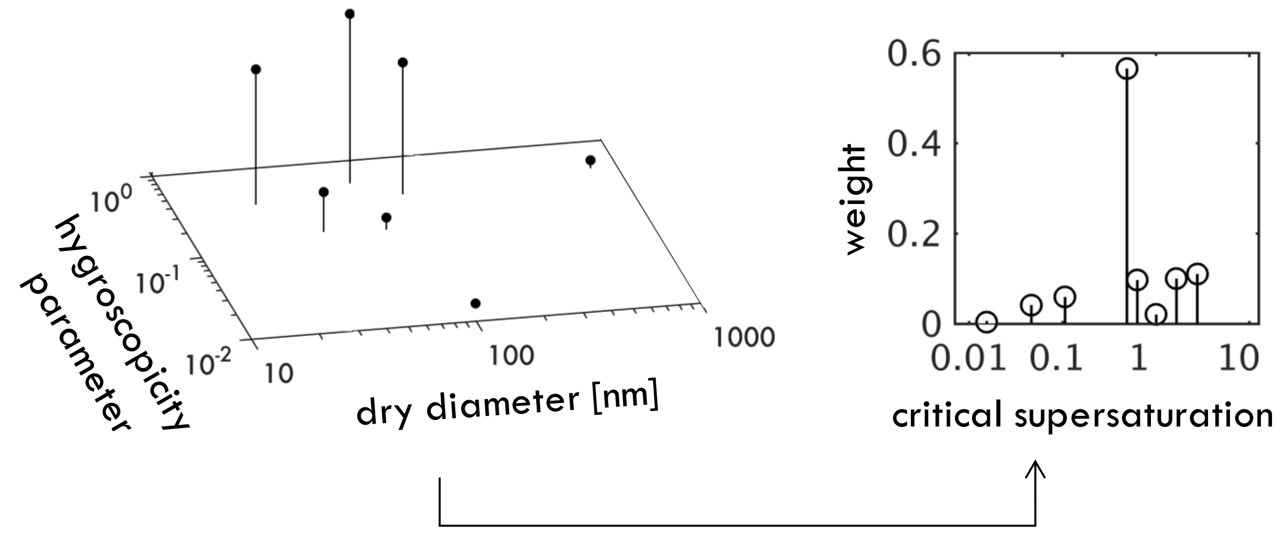
Accurate CCN spectra from quadrature



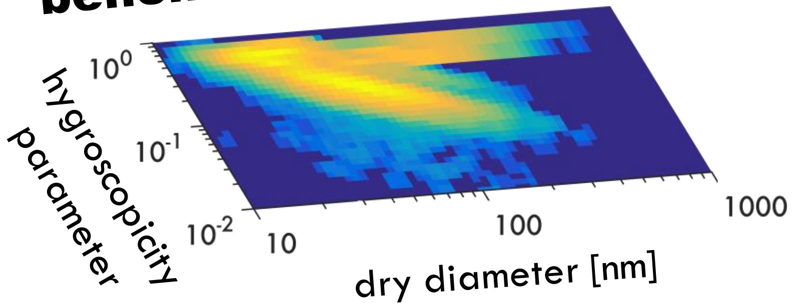


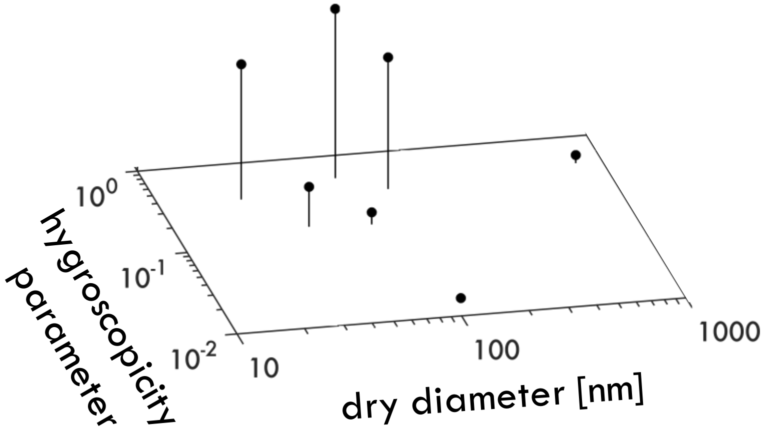
project quadrature from D_{dry}-κ space to s_c space

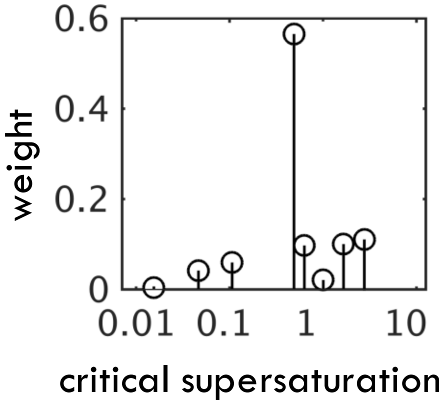
Fierce and McGraw, JGR 2017

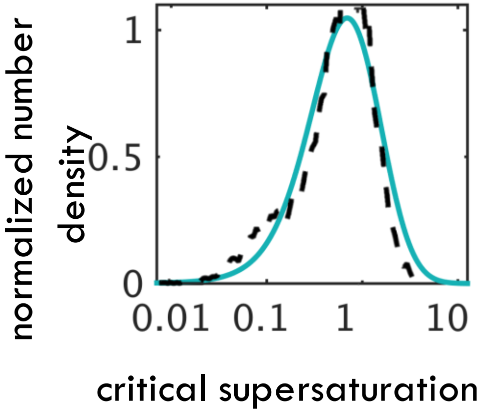
Accurate CCN spectra from quadrature

benchmark distribution







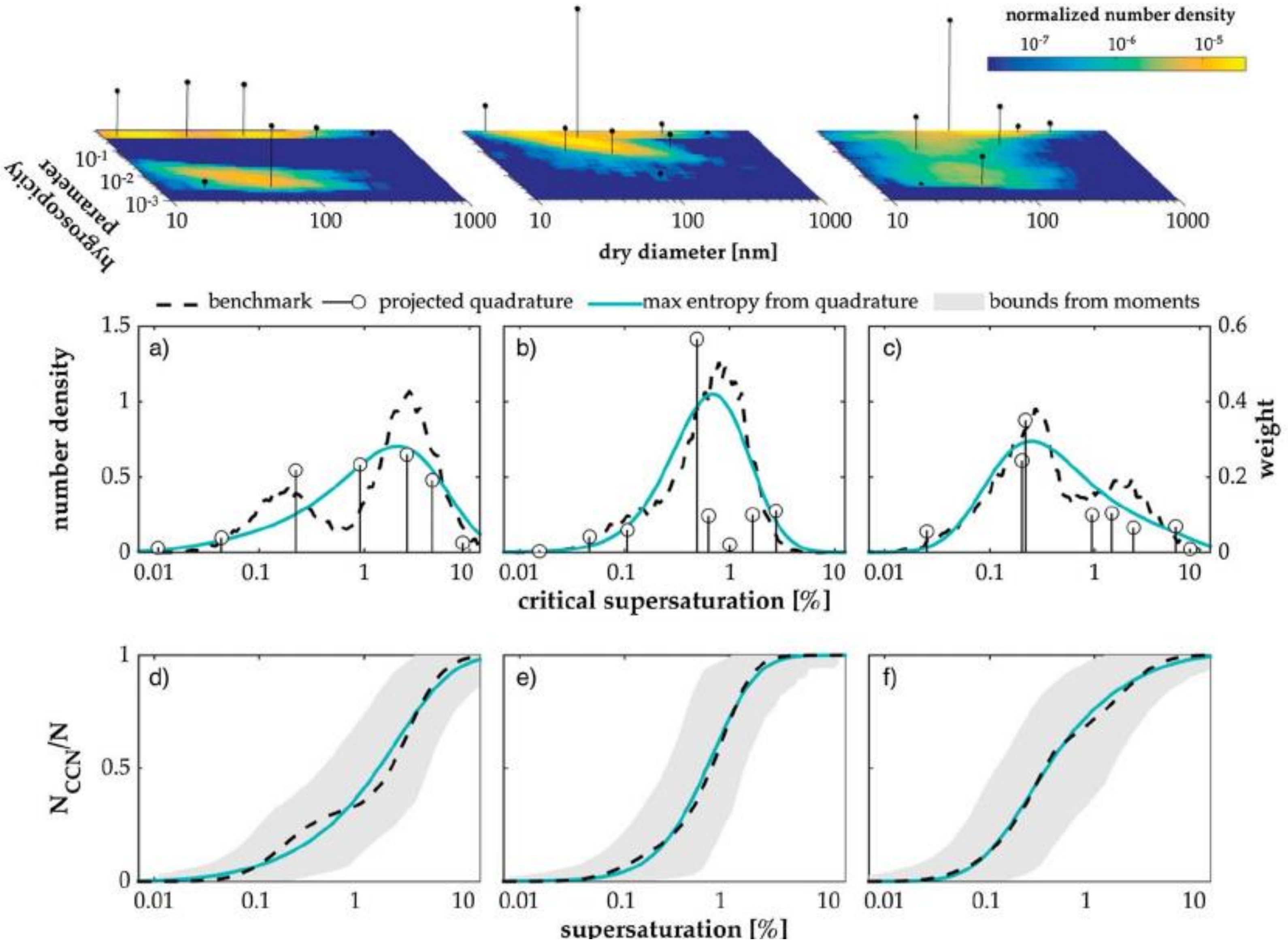


project quadrature from D_{dry}-κ space to s_c space

continuous distribution from moments with respect to s_c

Fierce and McGraw, JGR 2017

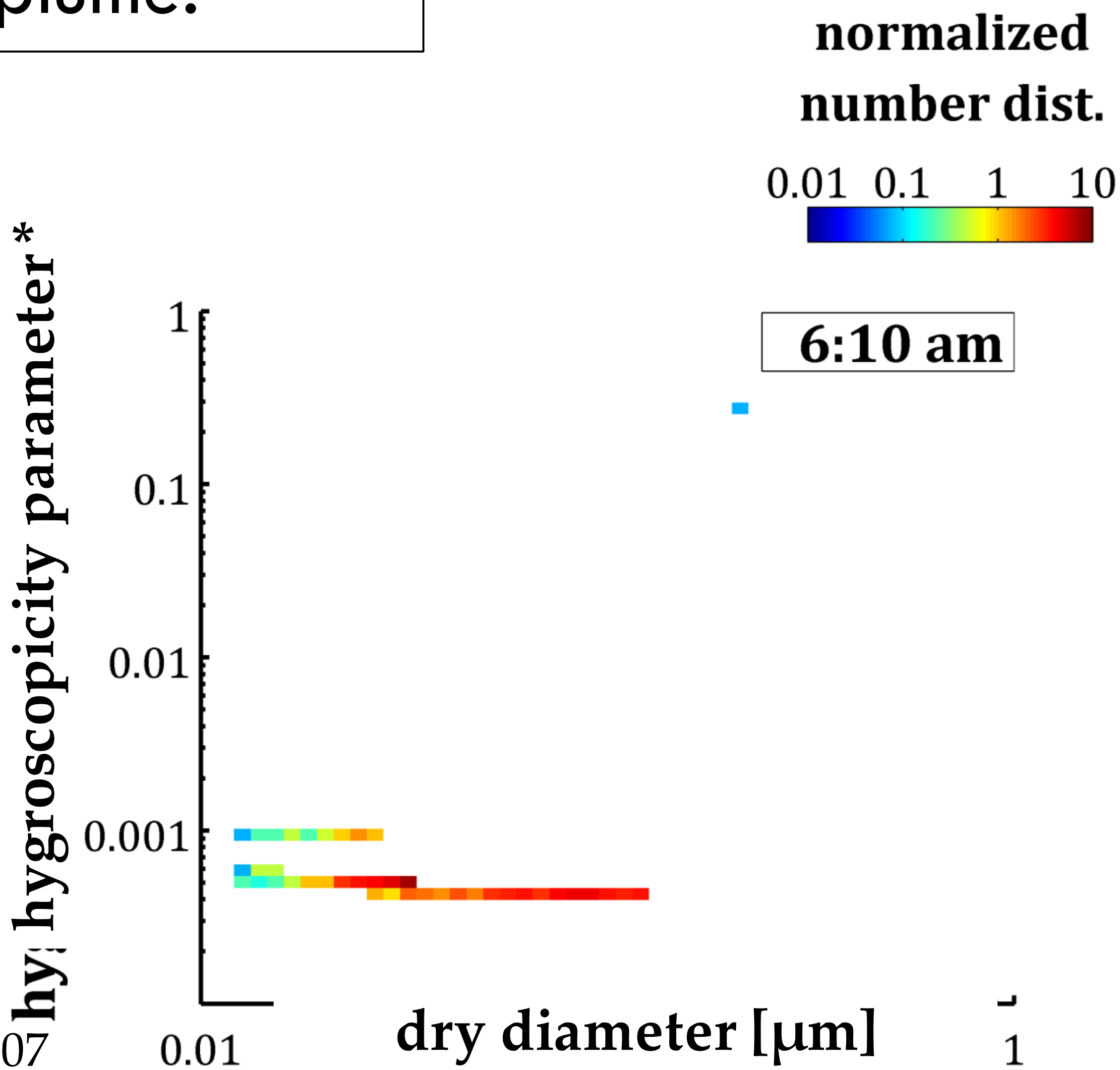
Accurate CCN spectra from quadrature



BC-containing particles evolve rapidly in a polluted plume.

Fierce et al., *JGR* 2013
Fierce et al., *ACP* 2015

Two-dimensional distribution shows changes in size and composition influencing CCN activation.



* Petters and Kreidenweis, ACP 2007

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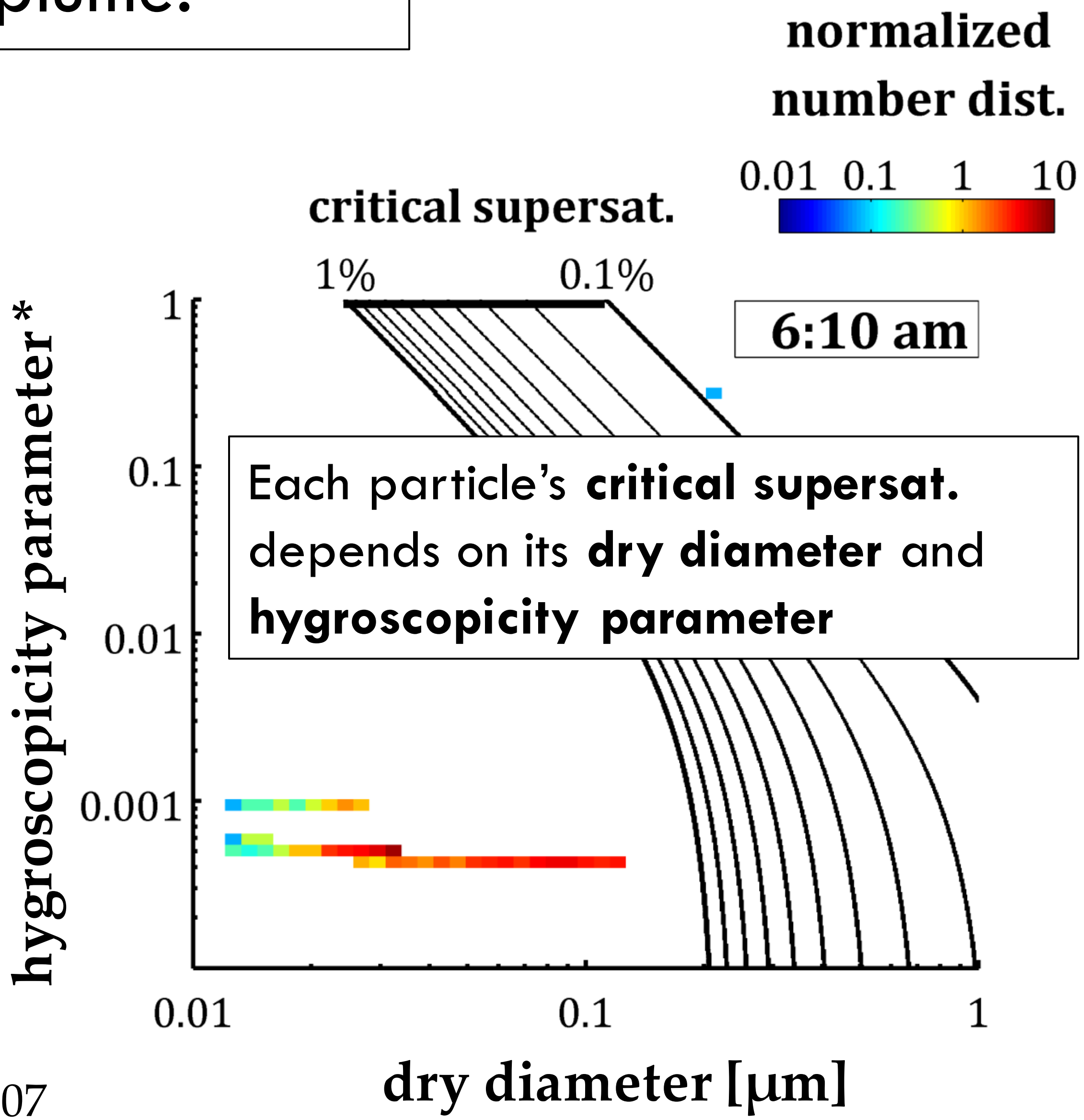

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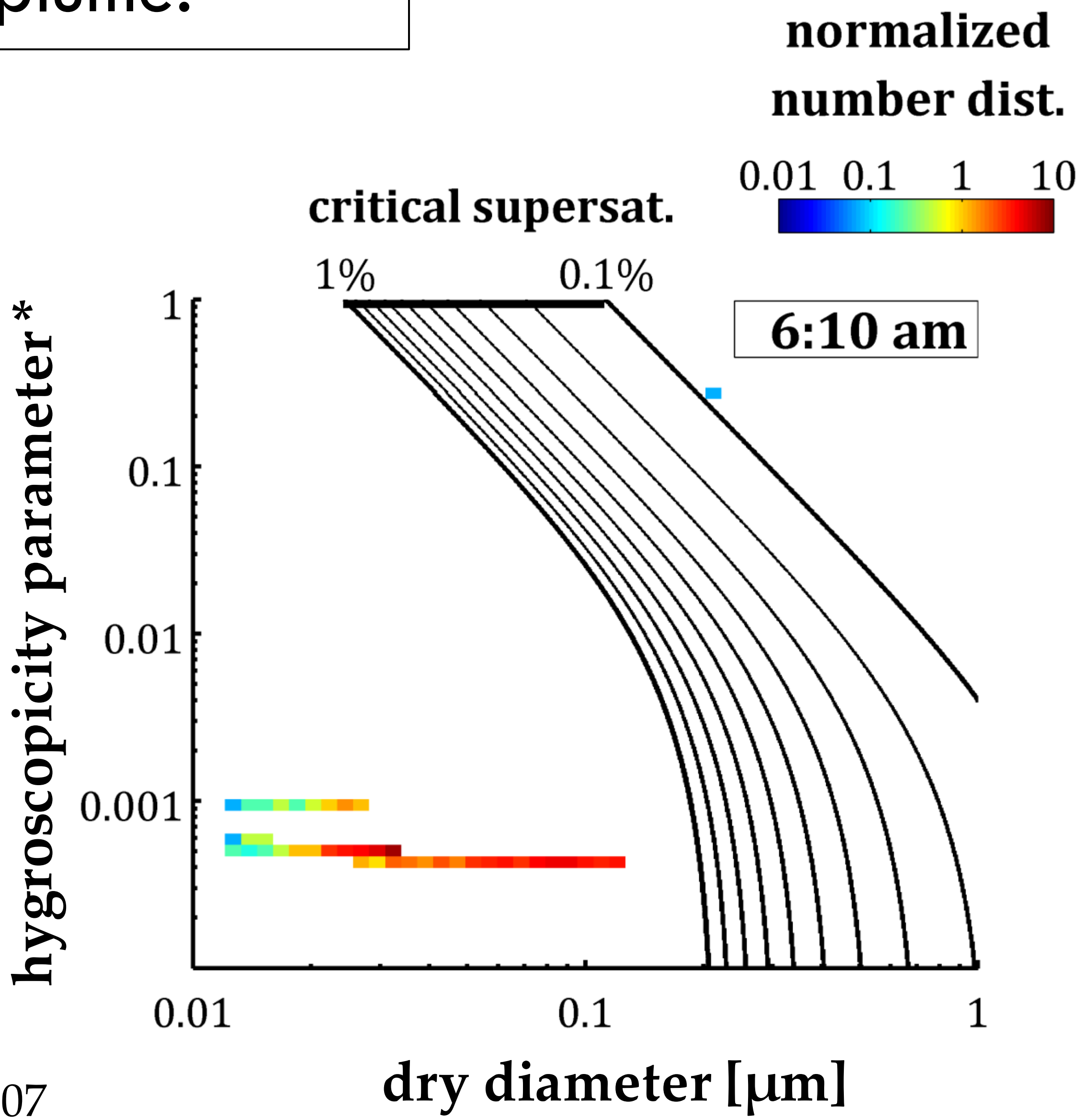

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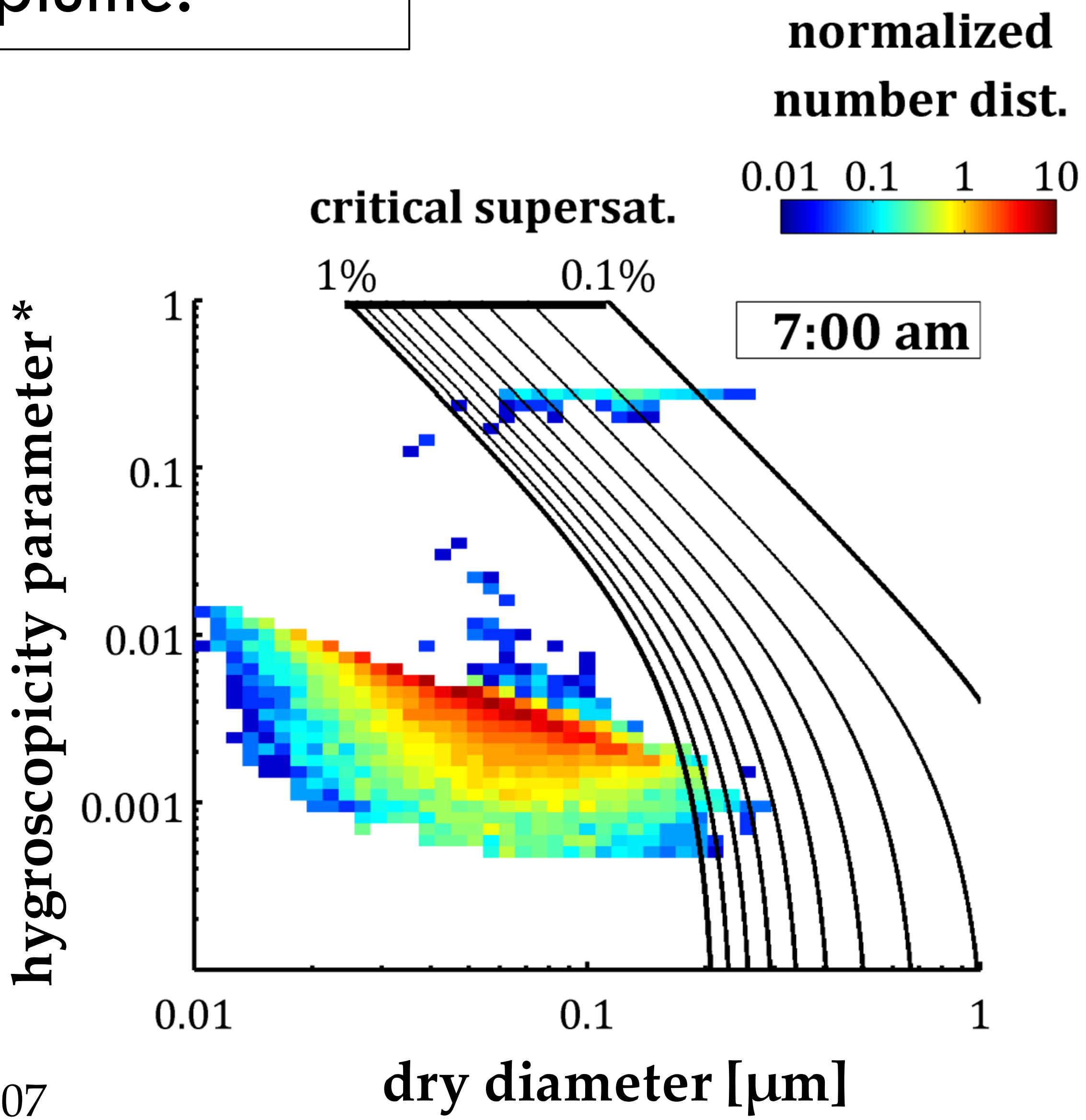

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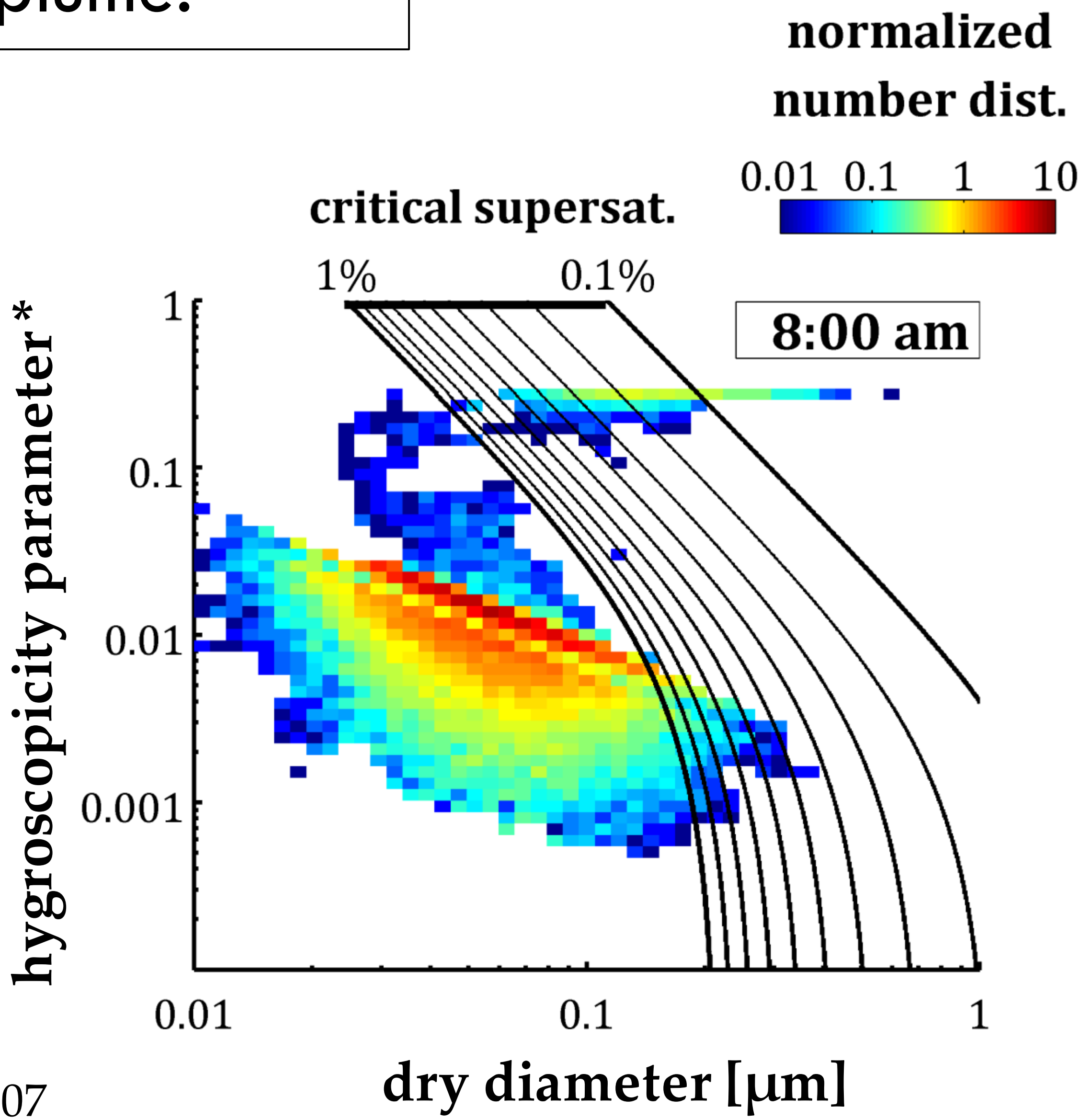

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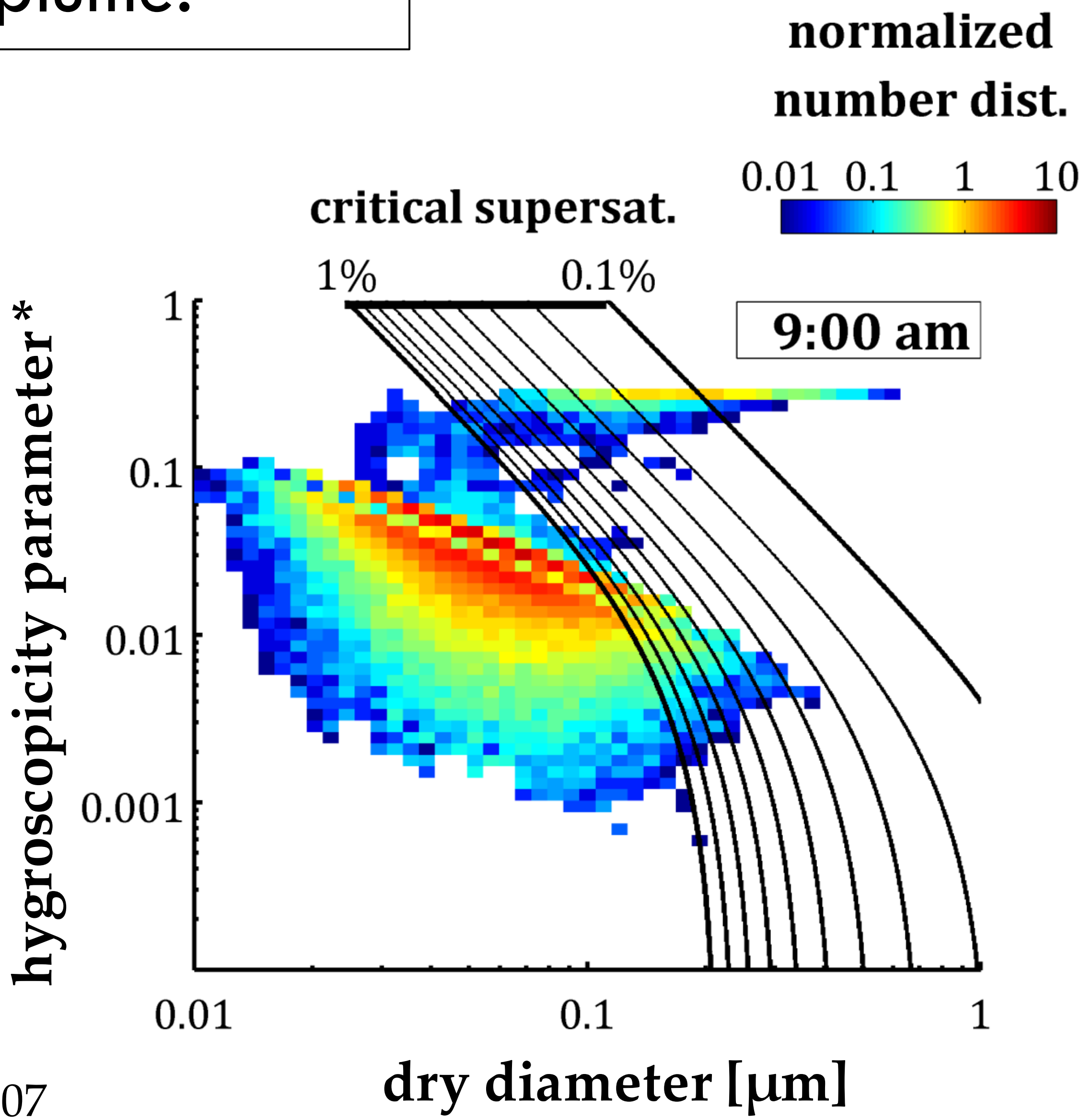

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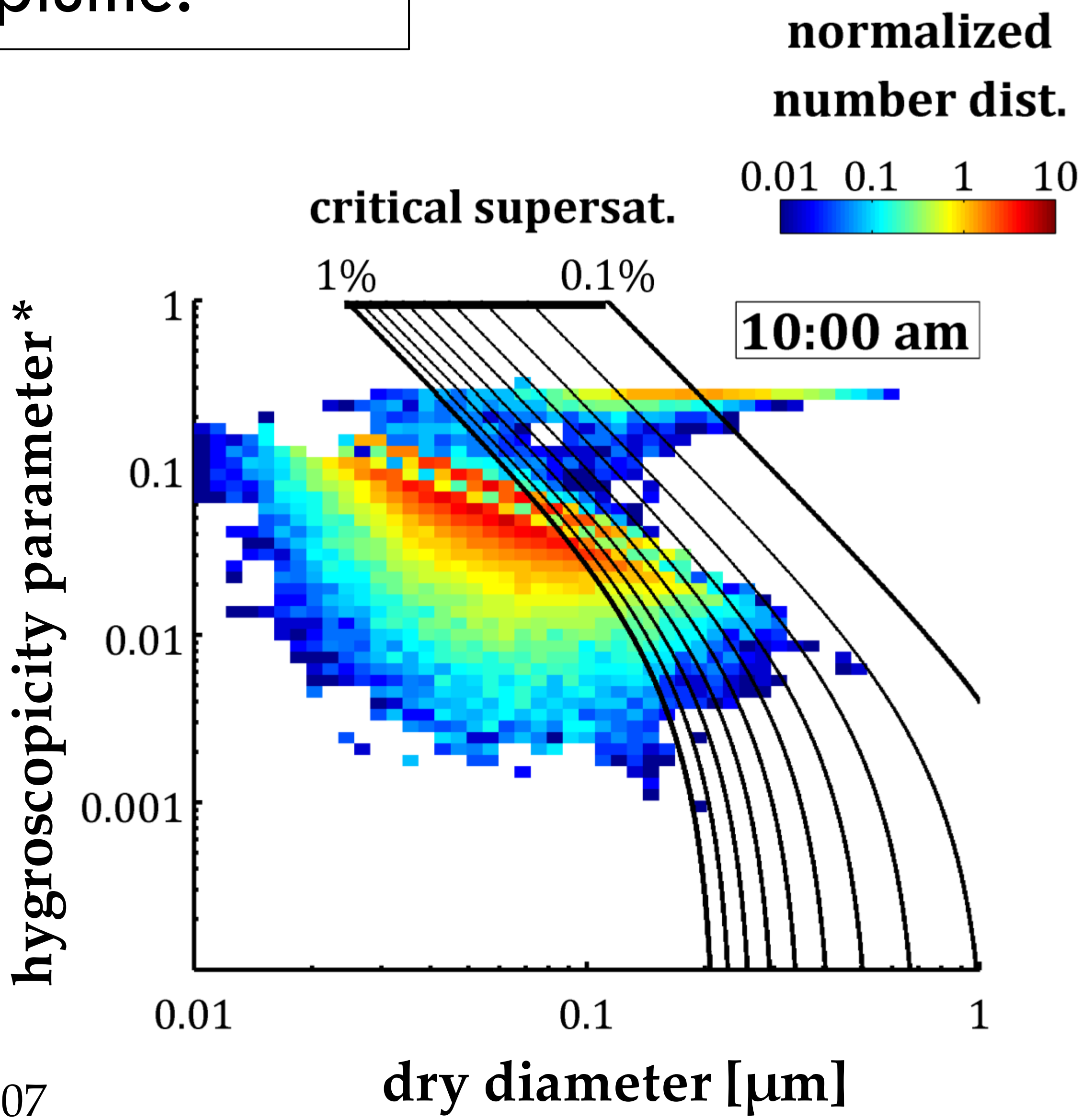

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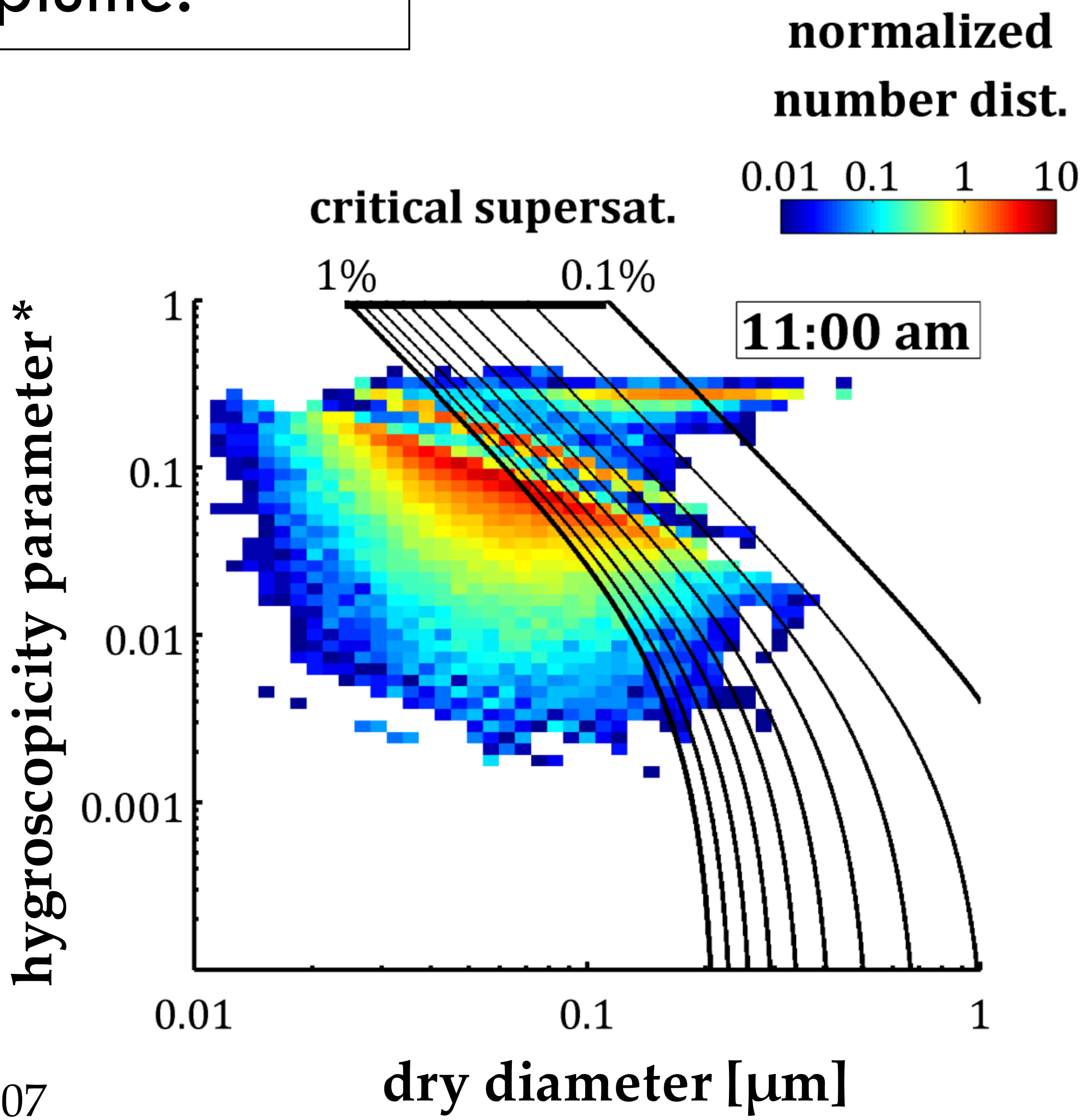

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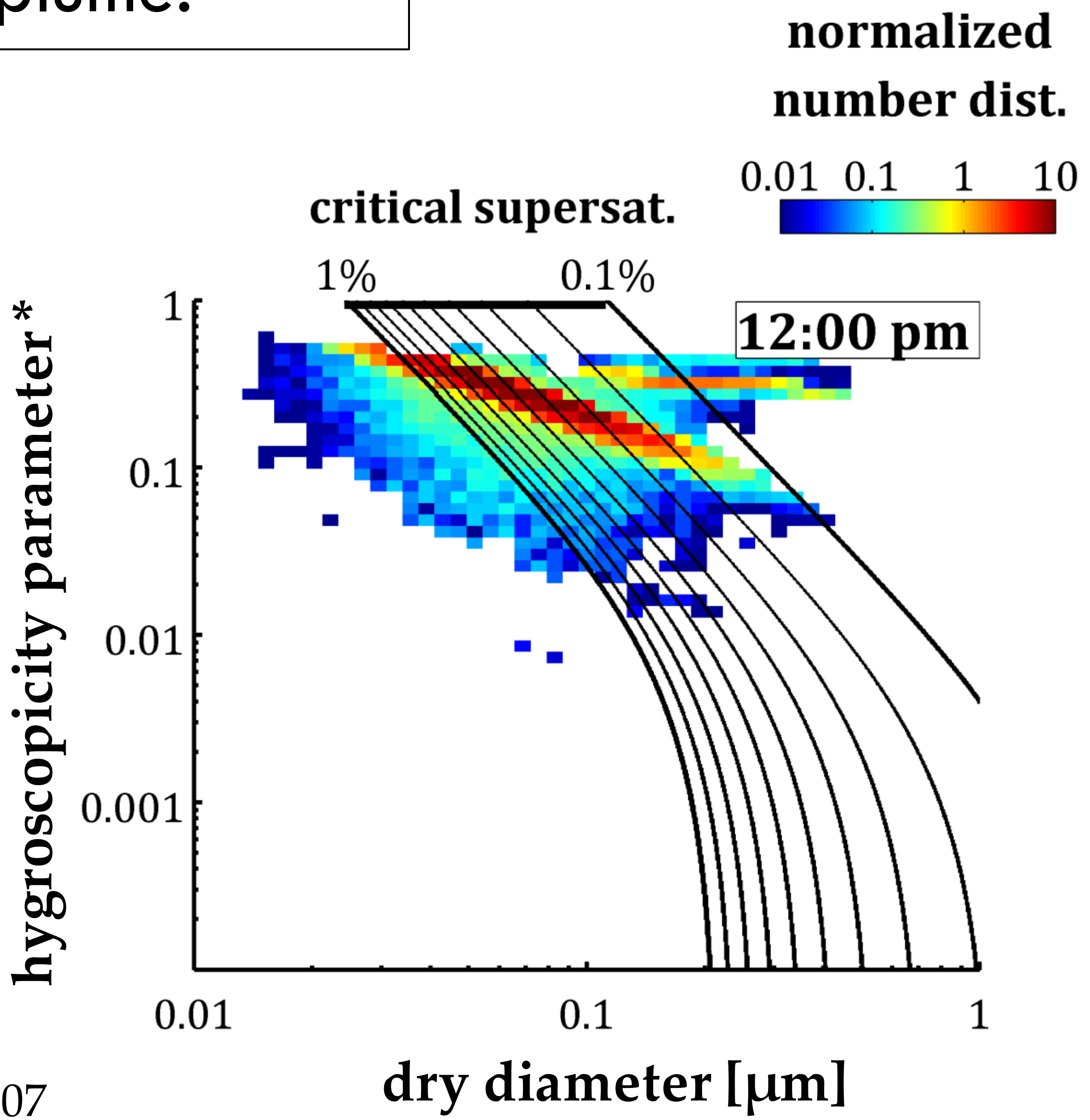

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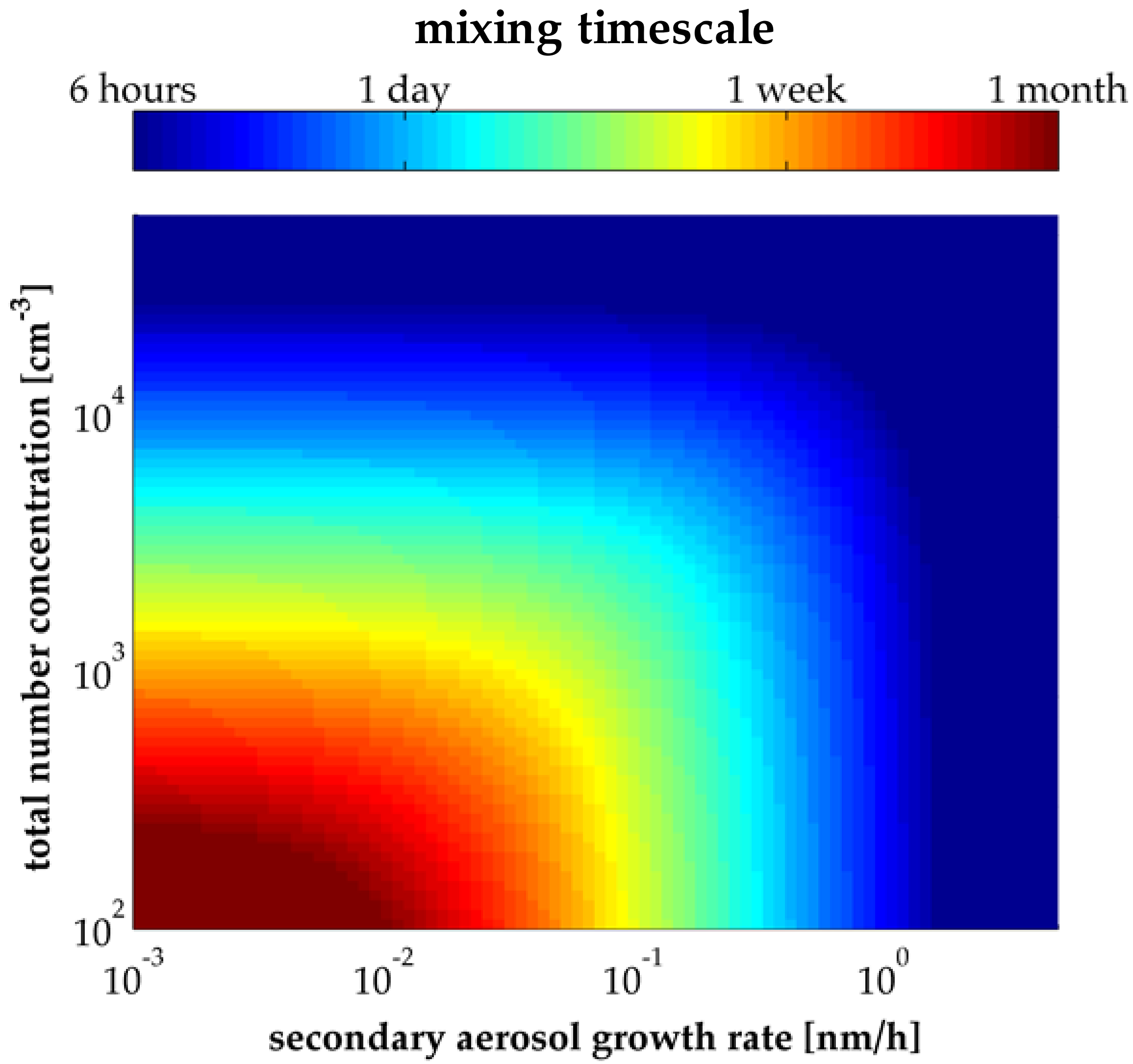
Parameterize hydrophobic to hygroscopic conversion using timescale for internal mixing



Timescale for internal mixing varies with local conditions.

Fierce et al., *BAMS* 2017

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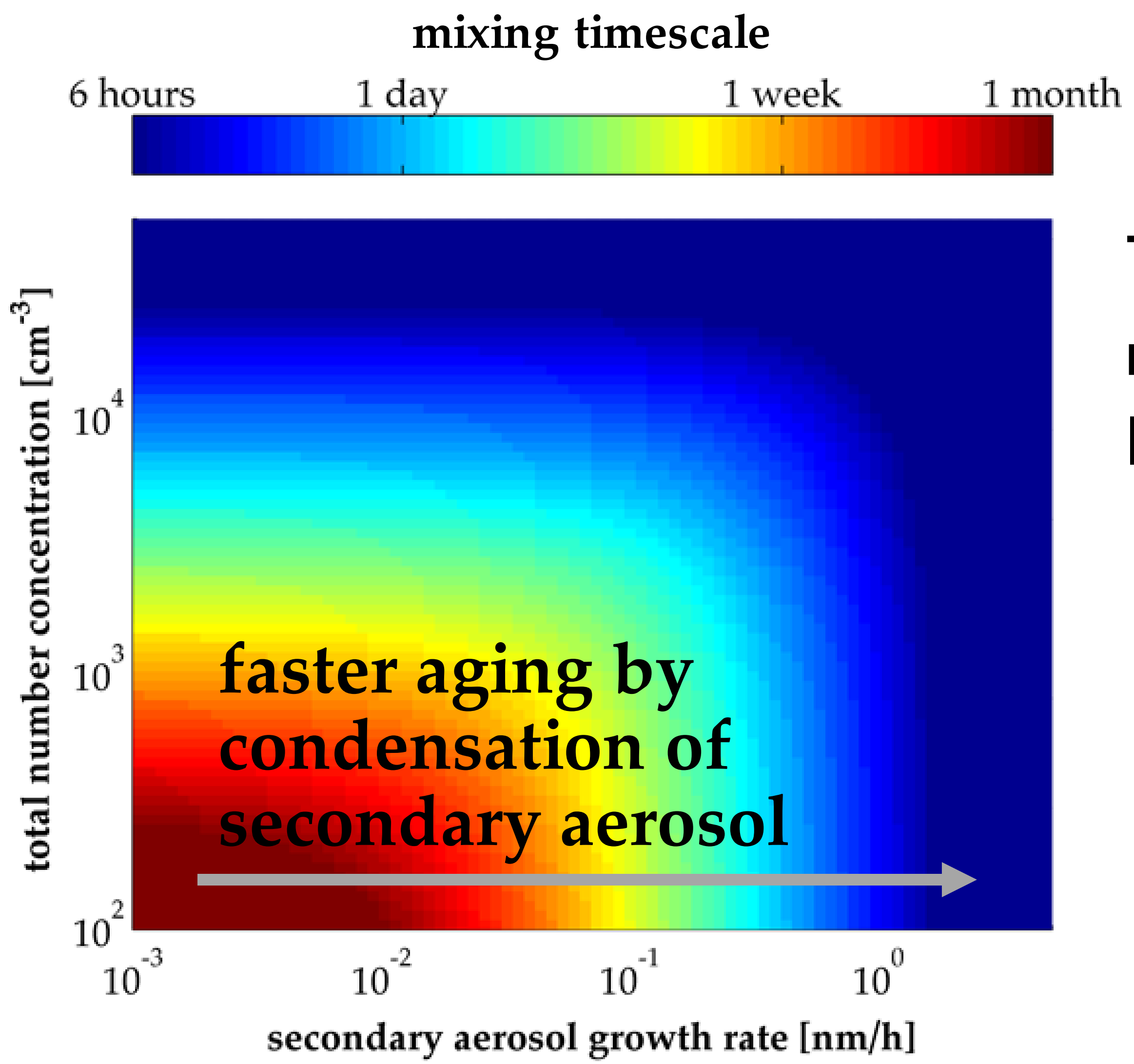

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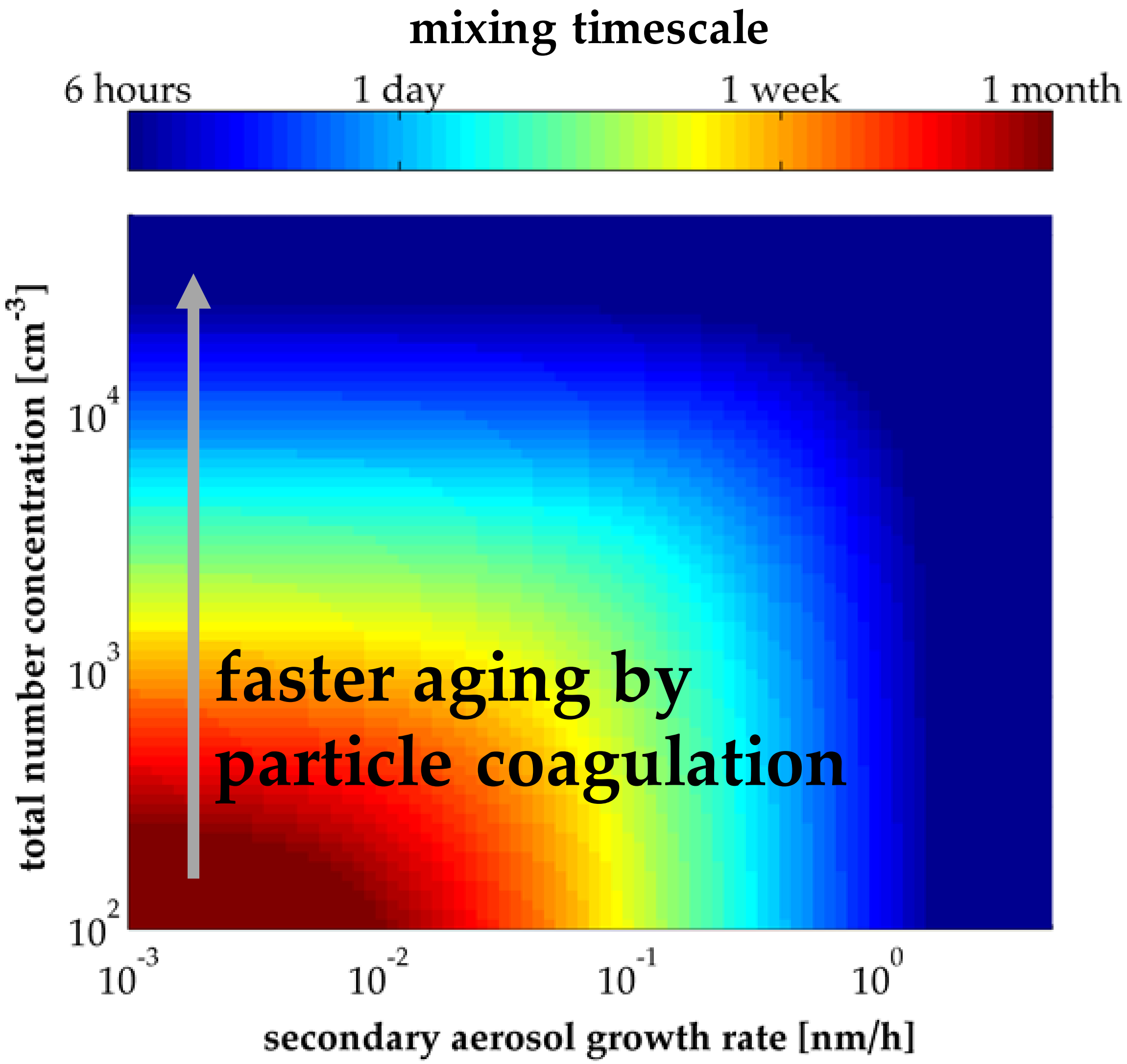

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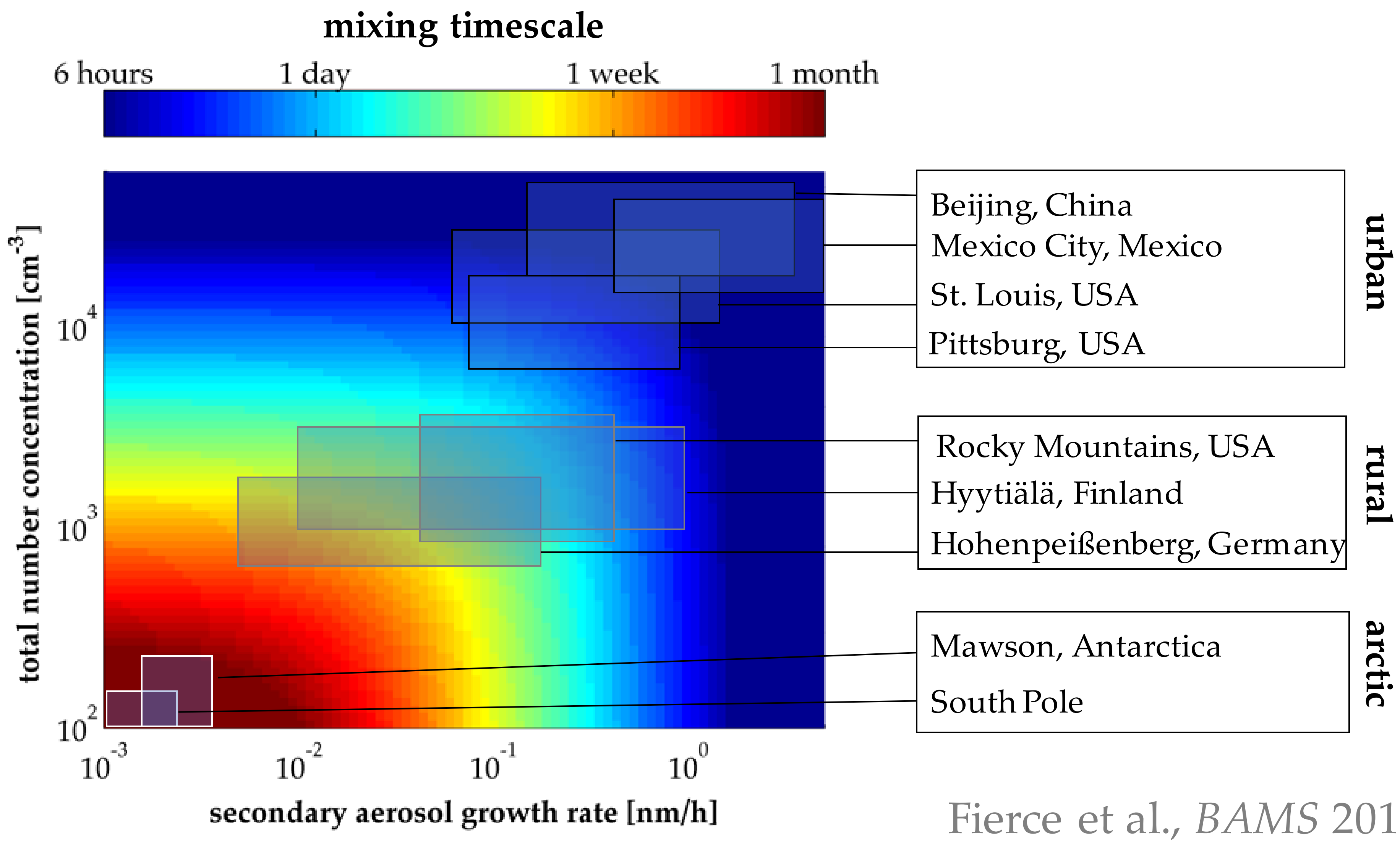
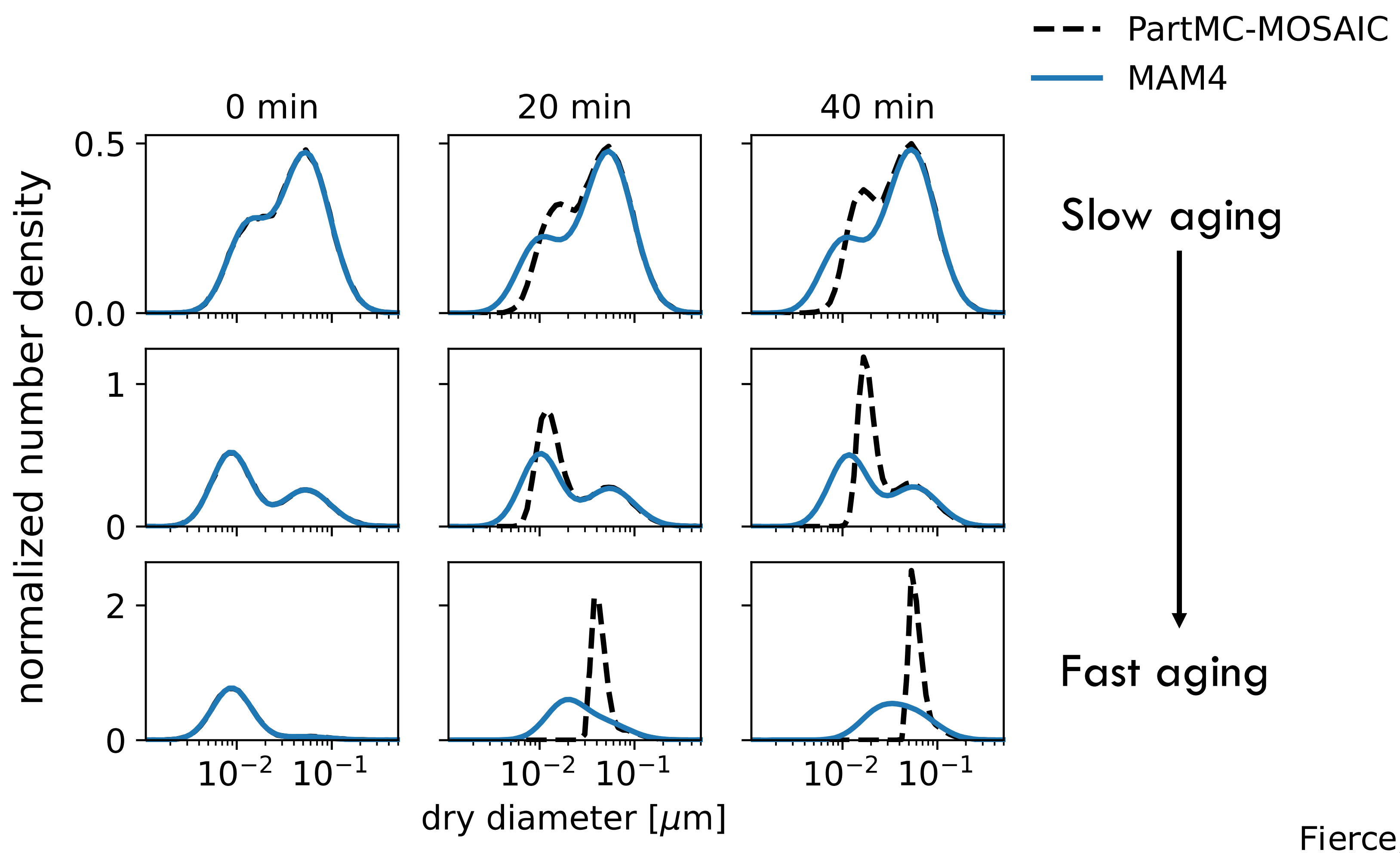


Fierce et al., *BAMS* 2017

Box models studies for evaluating reduced representation of size distributions

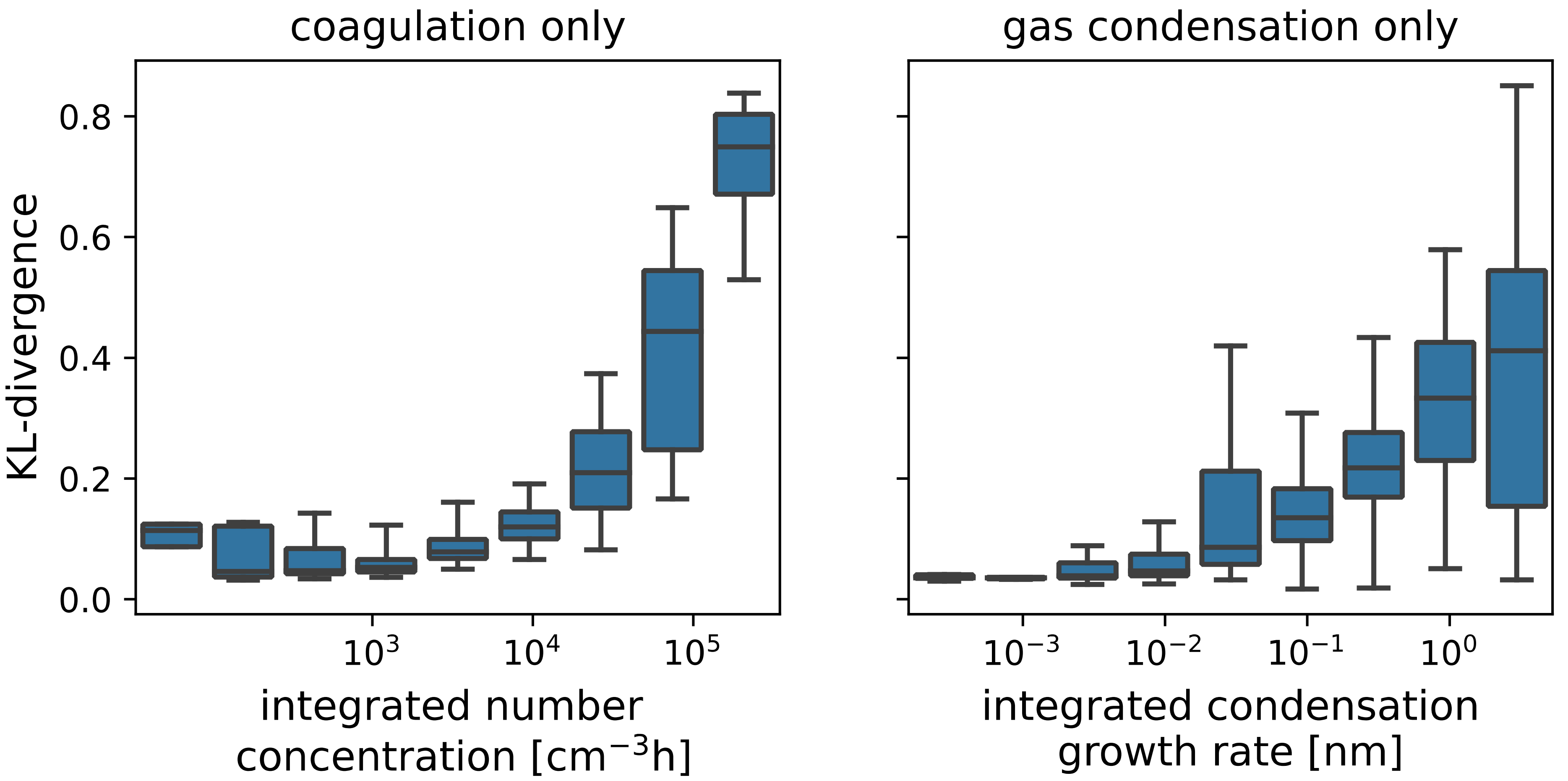


Slow aging

Fast aging

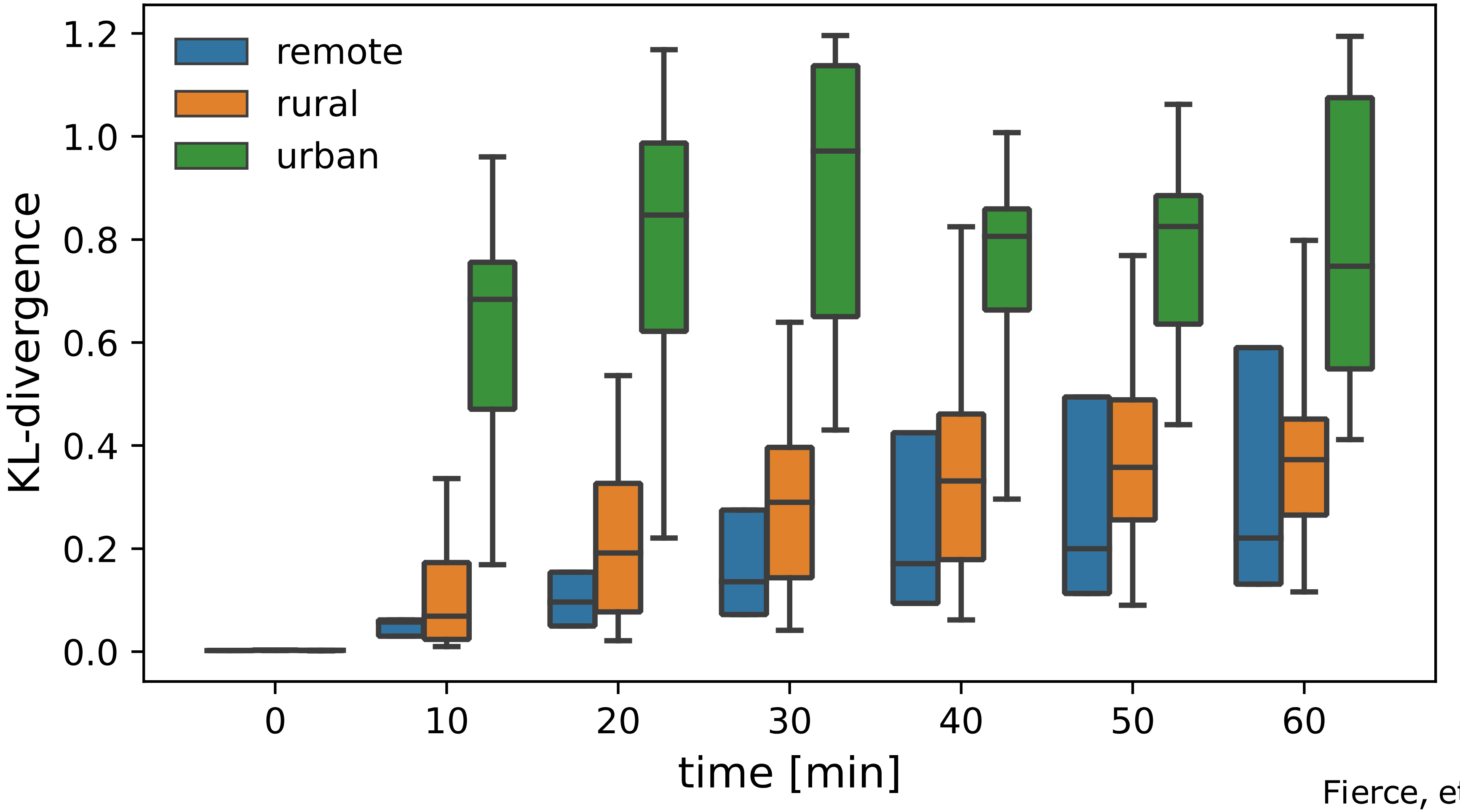
Fierce, et al. (in prep)

Across scenarios, divergence controlled by time integral over aging conditions



Fierce, et al. (in prep)

Reduced model diverges from benchmark across regimes, most quickly under polluted conditions



Fierce, et al. (in prep)

A plug for DOE training programs!

Office of Science Graduate Student Research (SCGSR) Program:

- science.osti.gov/wdts/scgsr
- Next deadline

Graduate student internships and postdoctoral positions:
careers.pnnl.gov

Reach out if you're interested: laura.fierce@pnnl.gov

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