An Unaccounted Pathway for Rapid Aging of Atmospheric Soot

Alexei Khalizov, Ella Ivanova, Egor Demidov, Ali Hasani, Jeffrey Curtis, Nicole Riemer, Gennady Gor

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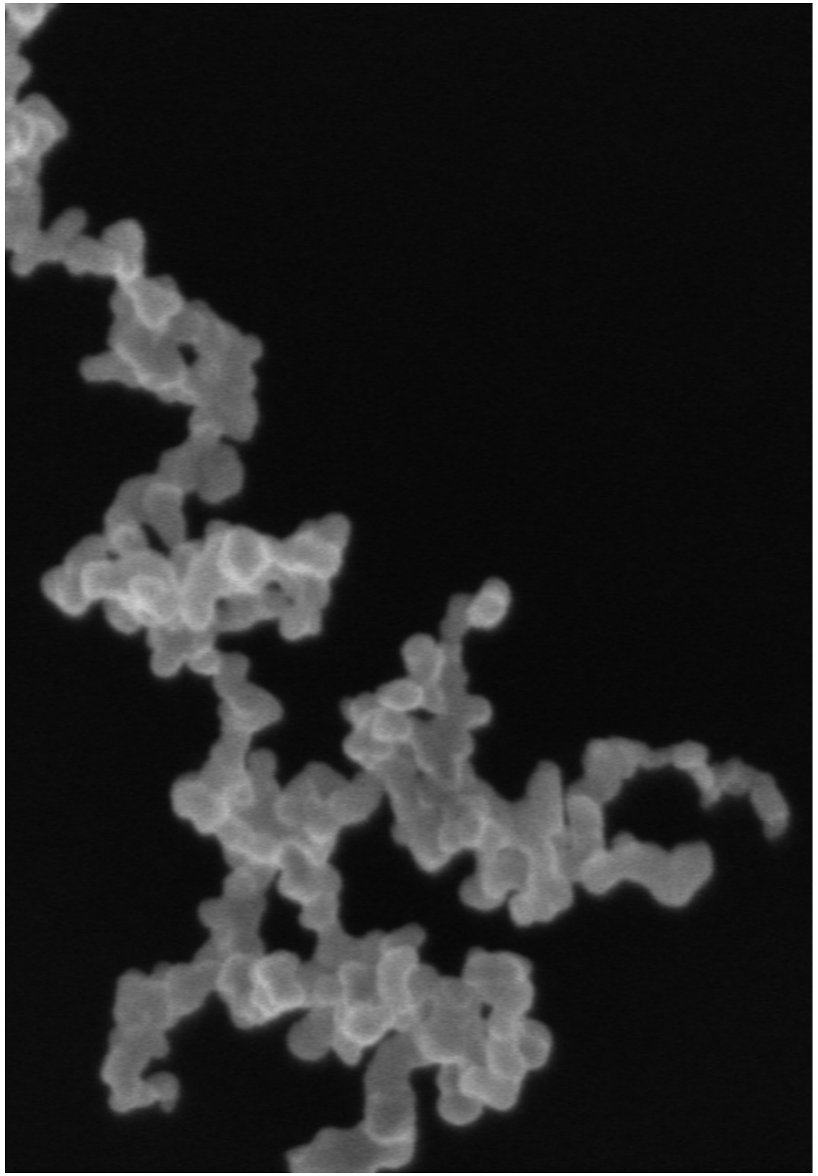
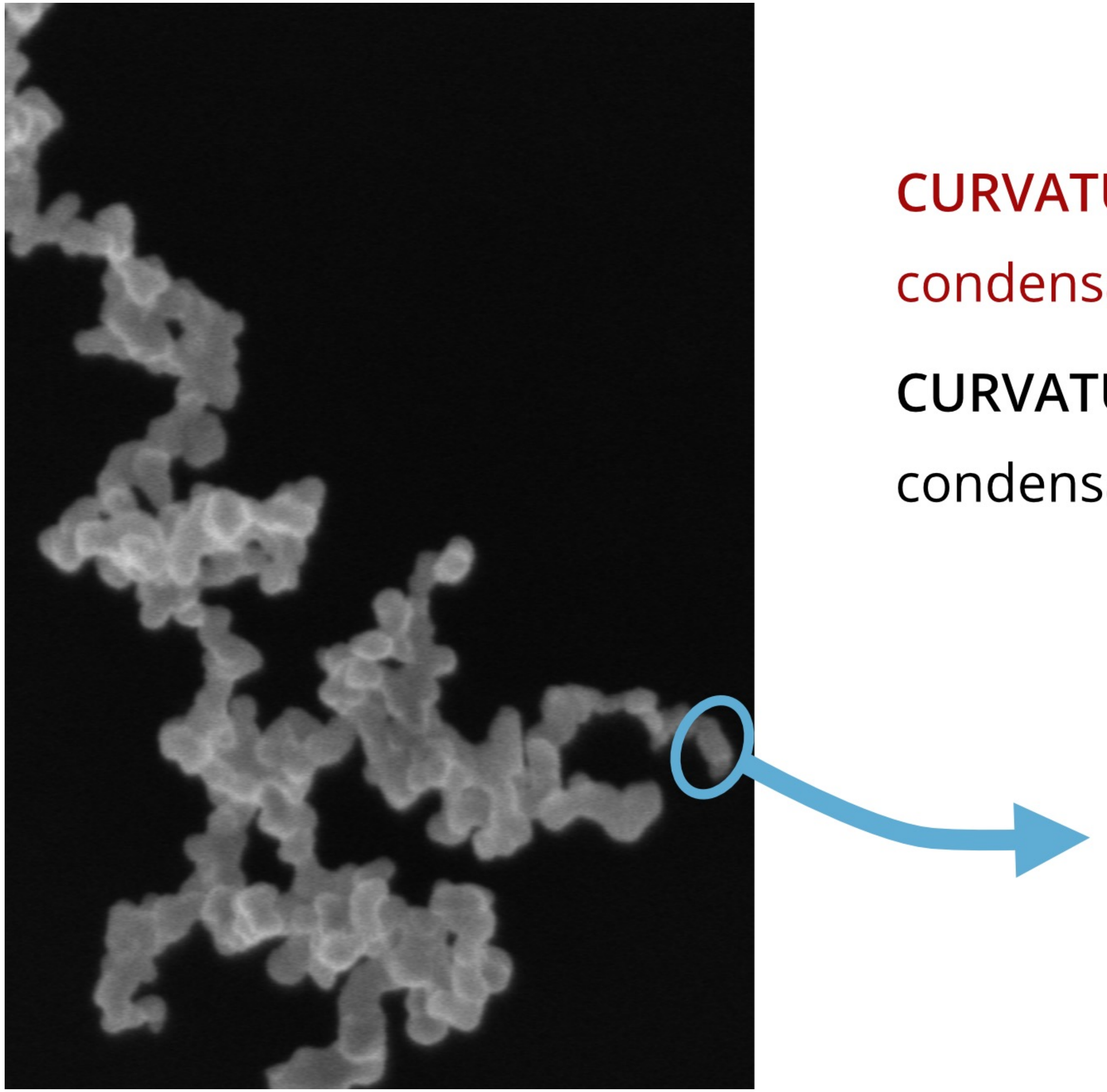
Fractal nature of soot



CURVATURE > 0

condensation resisted

Fractal nature of soot

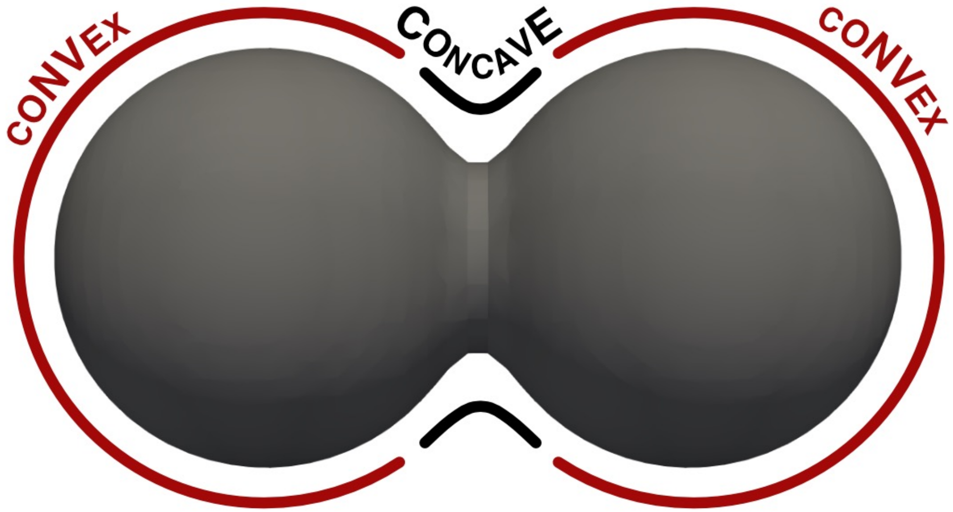


CURVATURE > 0

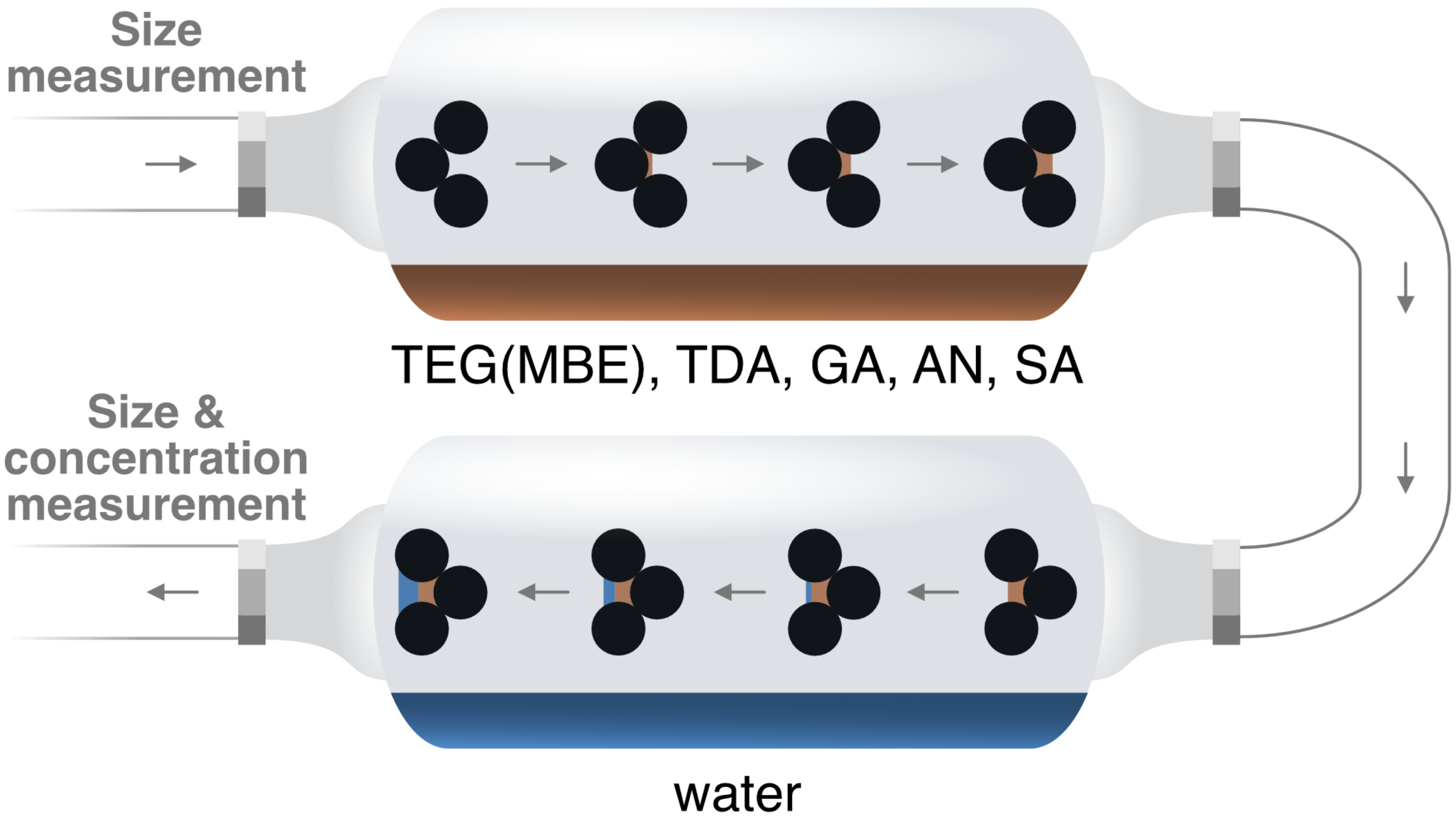
condensation resisted

CURVATURE < 0

condensation promoted



Experiment – aging of soot by capillary processing



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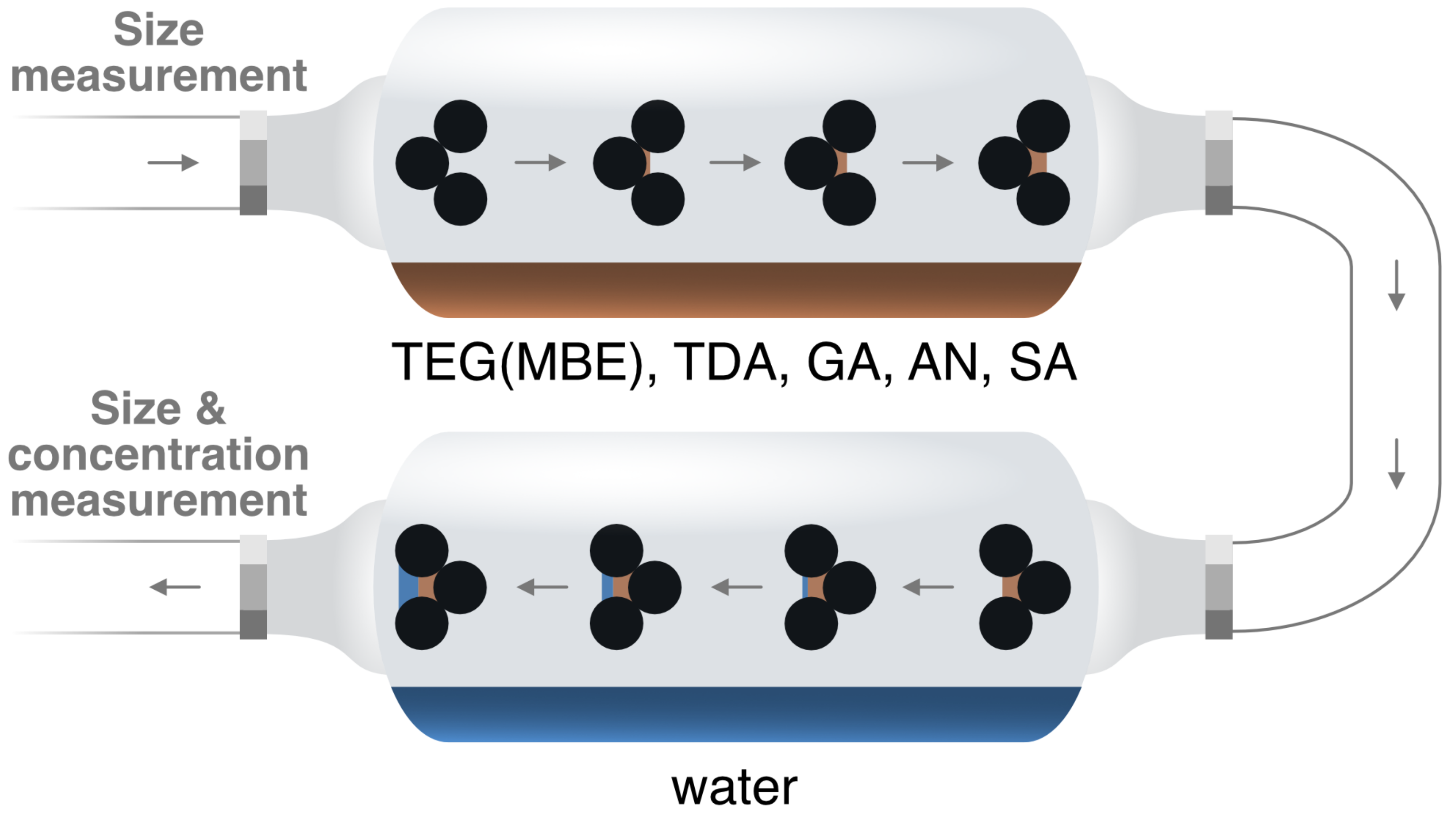

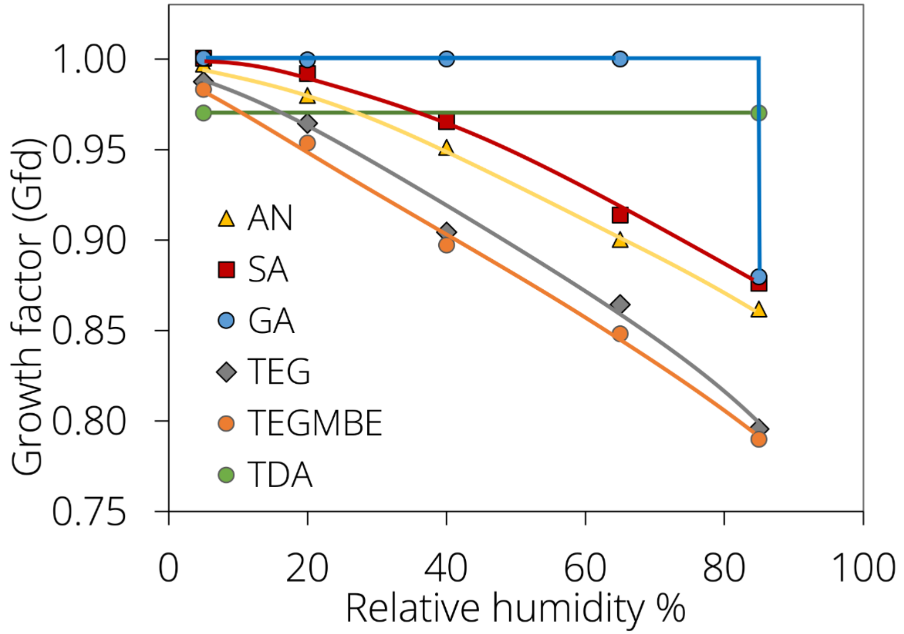

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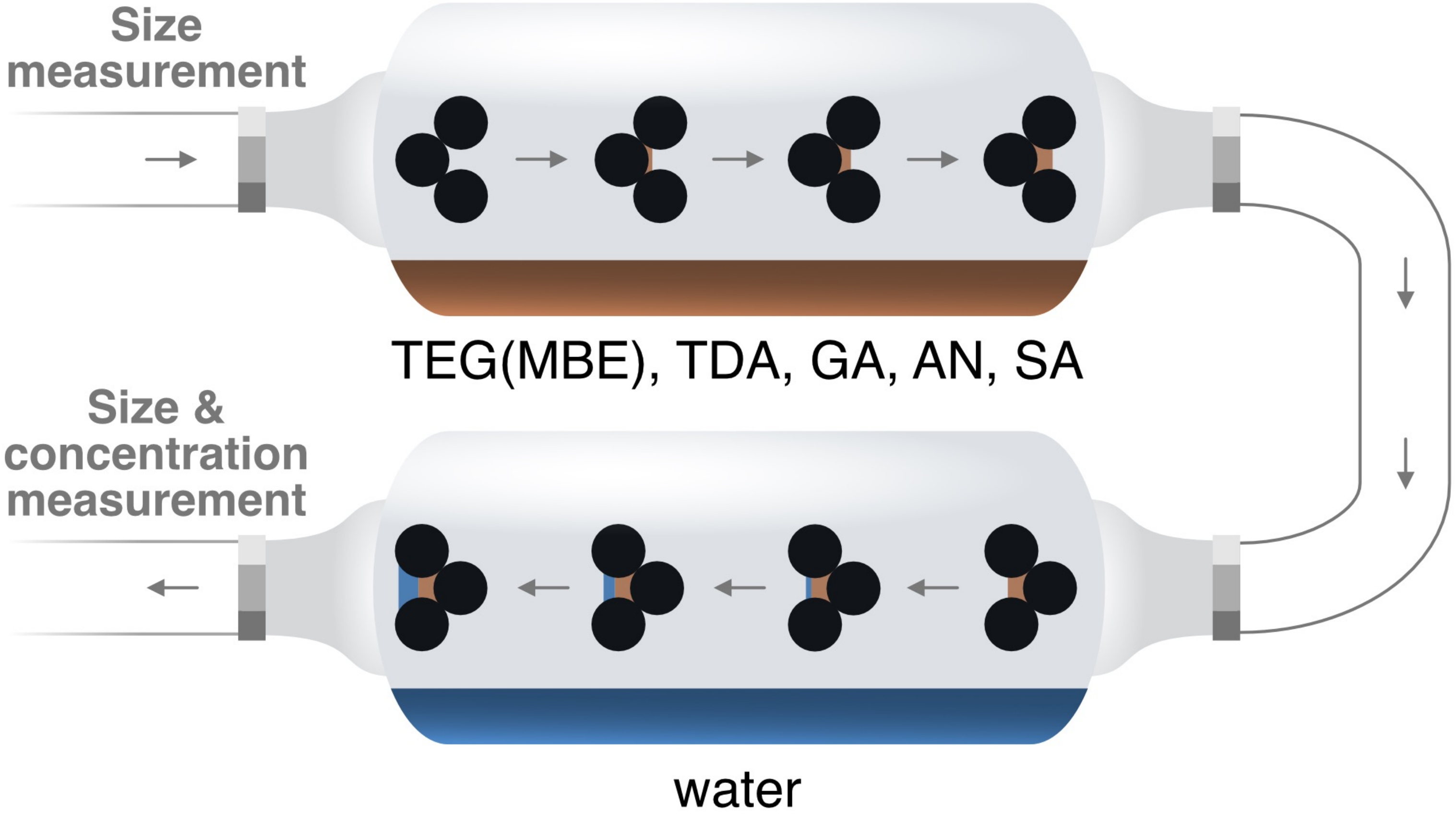
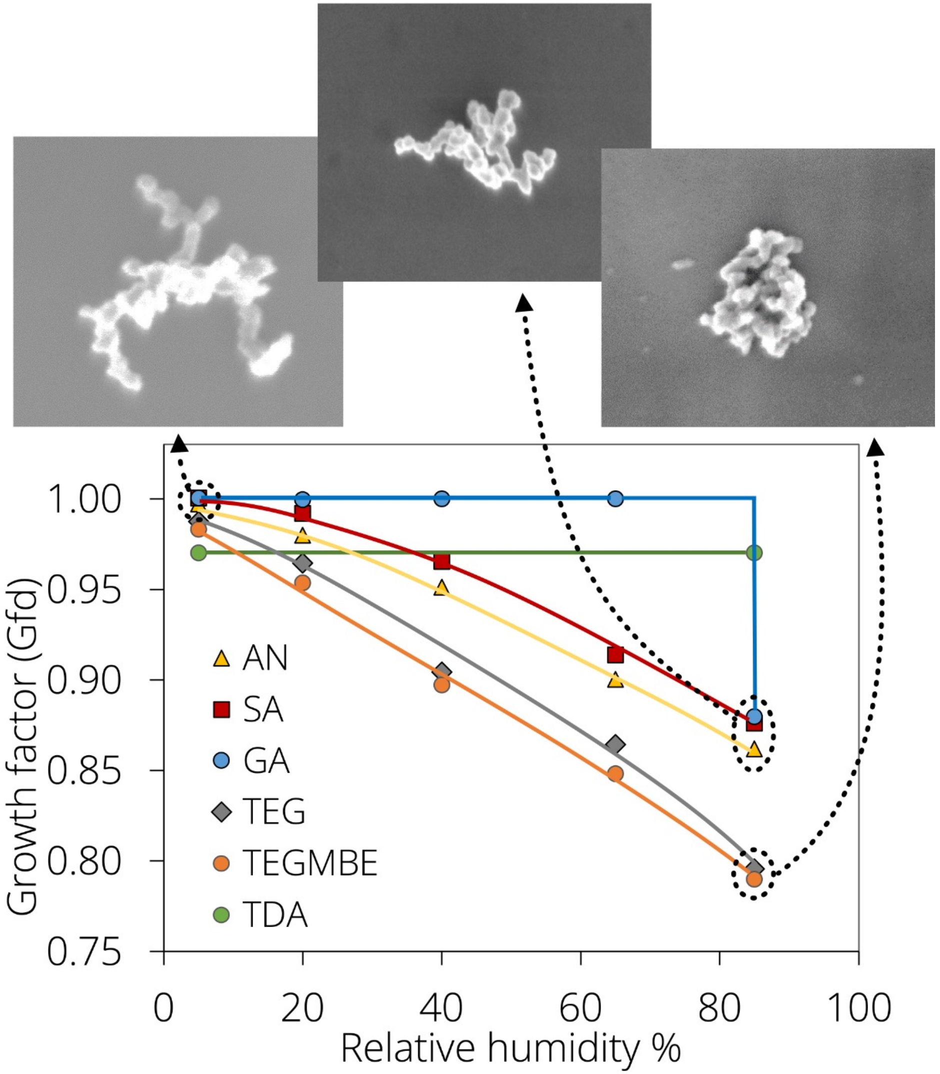
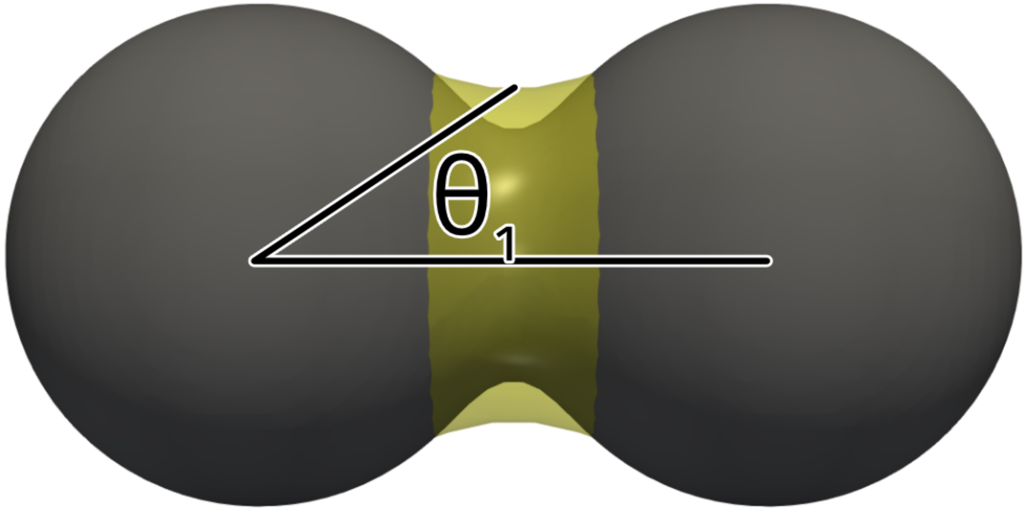




Step 1: capillary condensation of trace chemical



$$\frac{dV}{dt} \propto p_{\mathrm{sat}} \left(s - \exp\left[\frac{1}{2} \ell_{\mathrm{K}} \kappa \right] \right)$$

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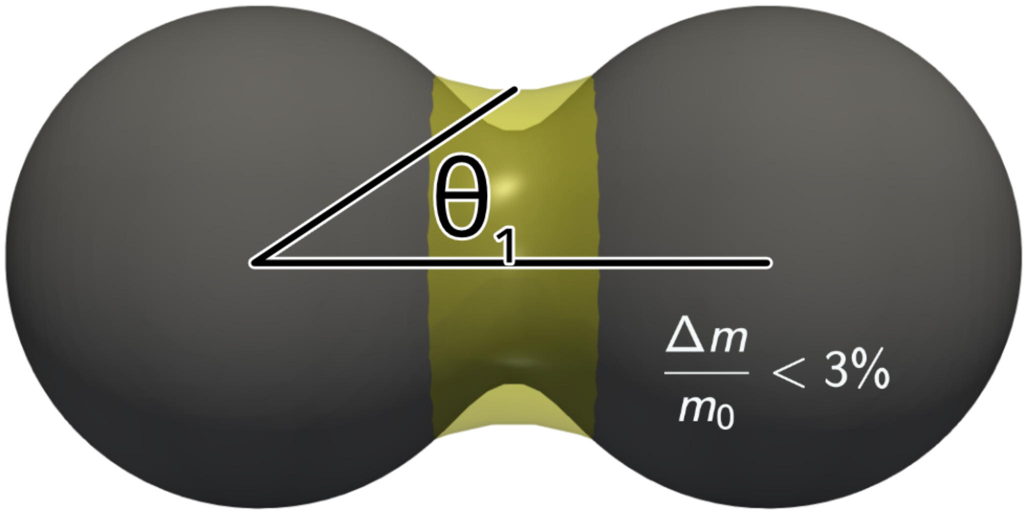
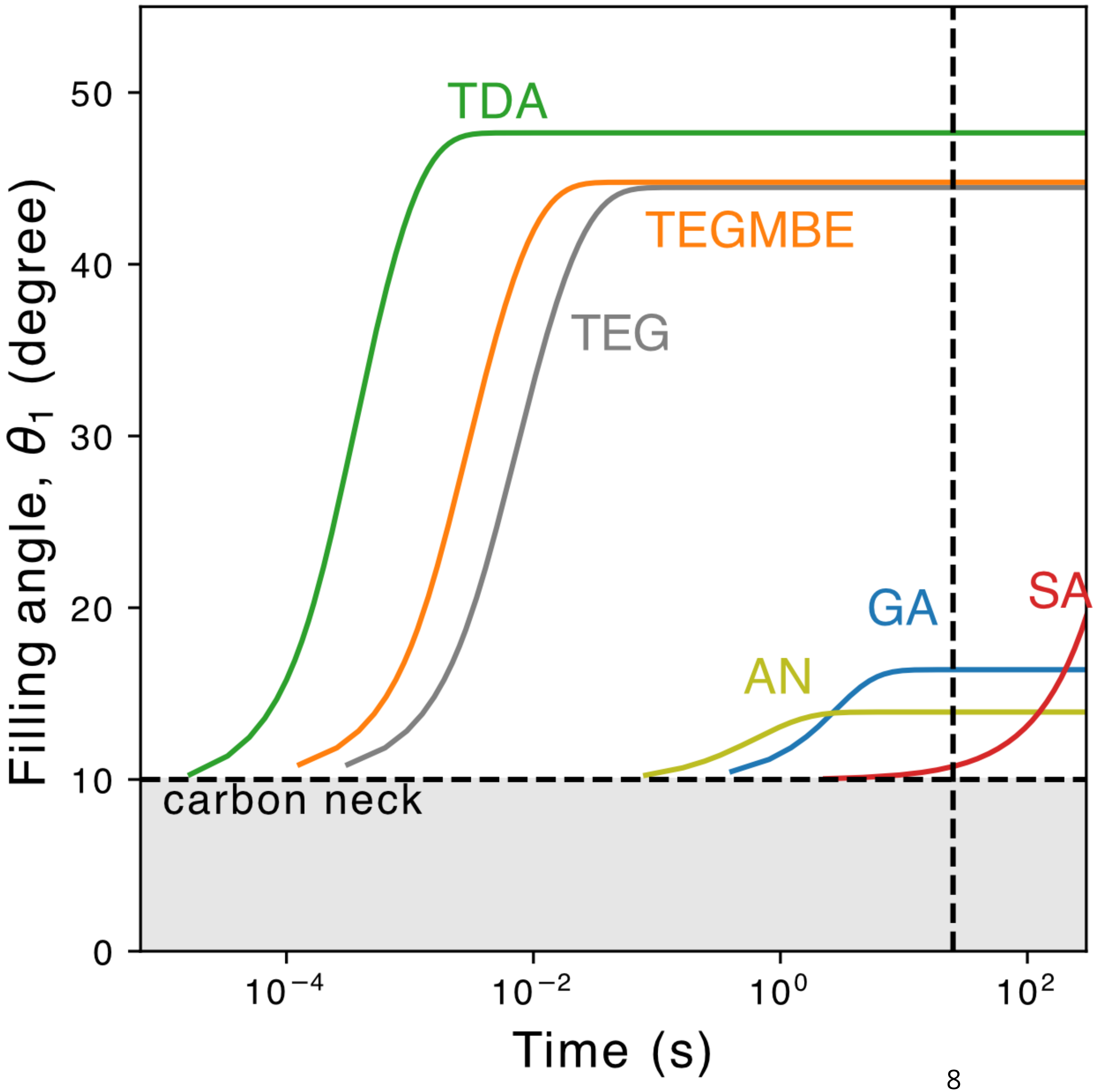
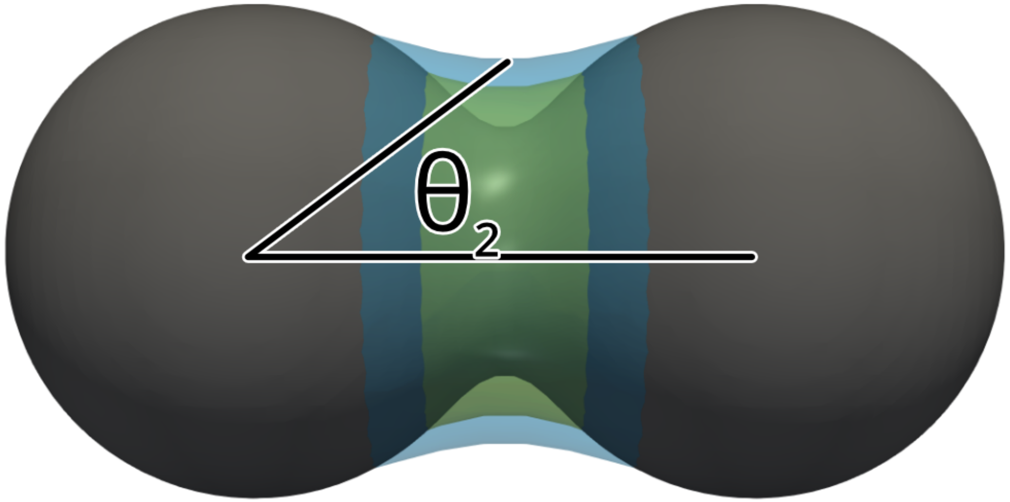


$$\frac{dV}{dt} \propto p_{\text{sat}} \left(s - \exp\left[\frac{1}{2} \ell_K \kappa \right] \right)$$

- No measurable mass gain
- Slight compaction - indicator of condensation



Step 2: water vapor absorption



$$p^{\mathrm{RH}} = p(V_{\mathrm{H_2O}}) \exp\left[\frac{1}{2}\ell_{\mathrm{K}}\kappa\right]$$

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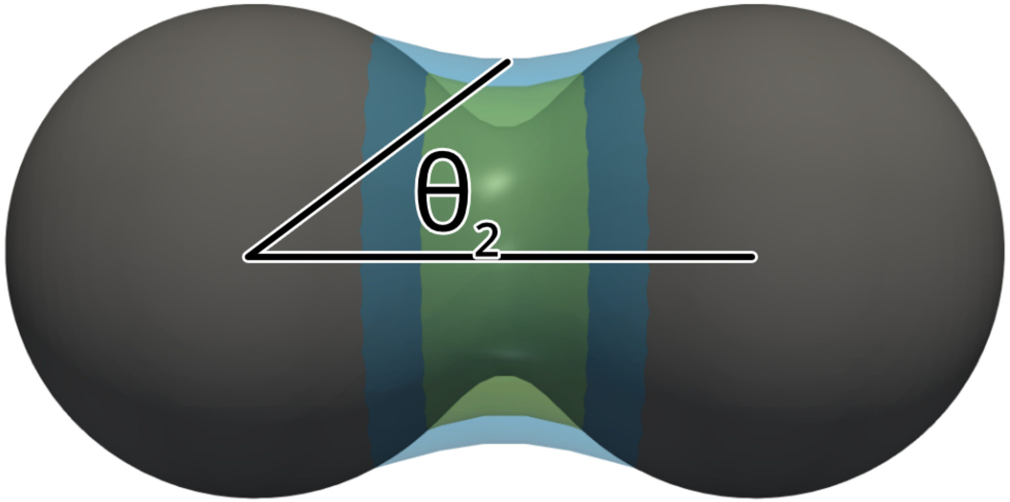
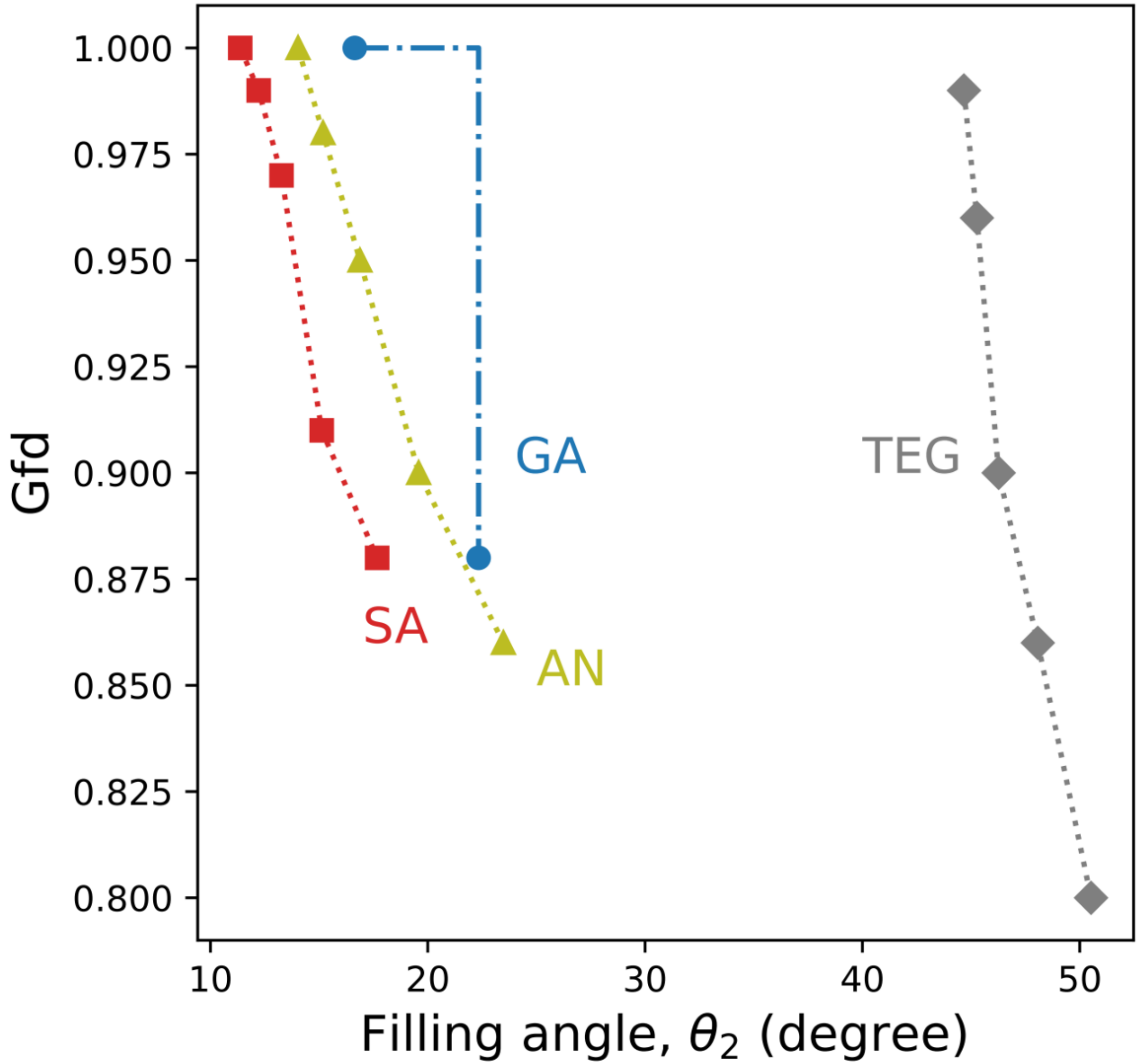


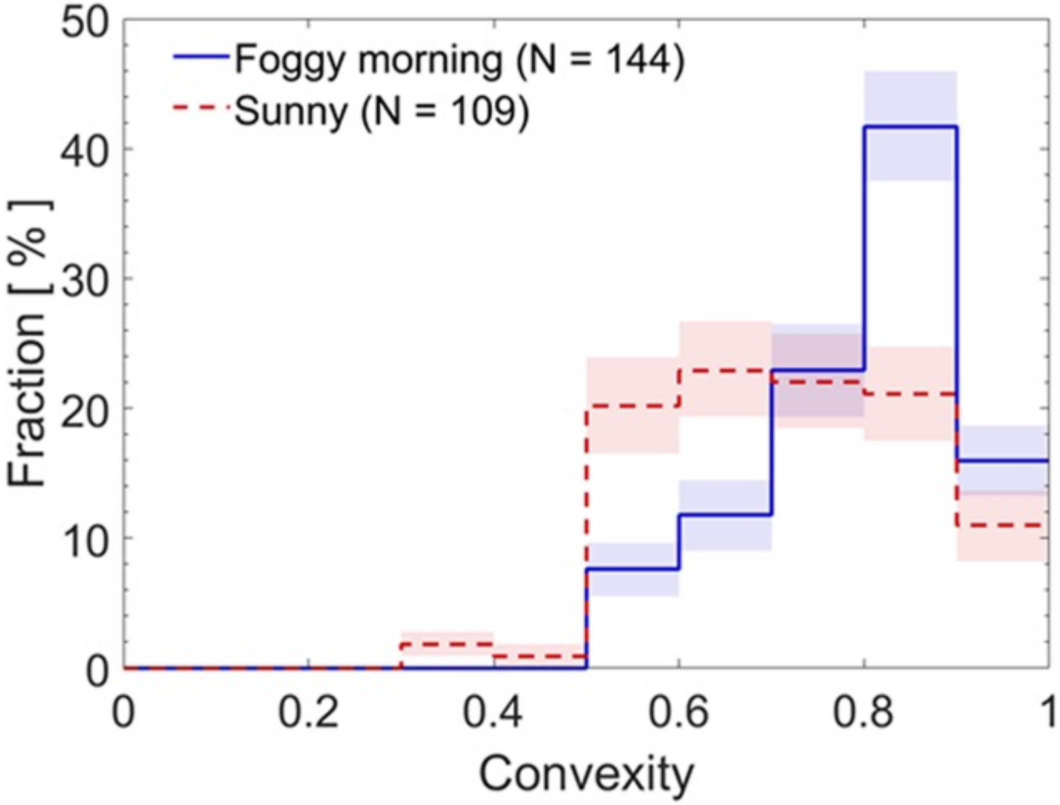
$$p^{\mathrm{RH}} = p(V_{\mathrm{H_2O}}) \exp\left[\frac{1}{2}\ell_{\mathrm{K}}\kappa\right]$$

- Compaction increasing with relative humidity
- Consistently, model predicts increasing filling angles



Assessing the role of capillary condensation in the atmosphere

- Field and chamber studies observed compact soot particles lacking visible coatings[1,2]

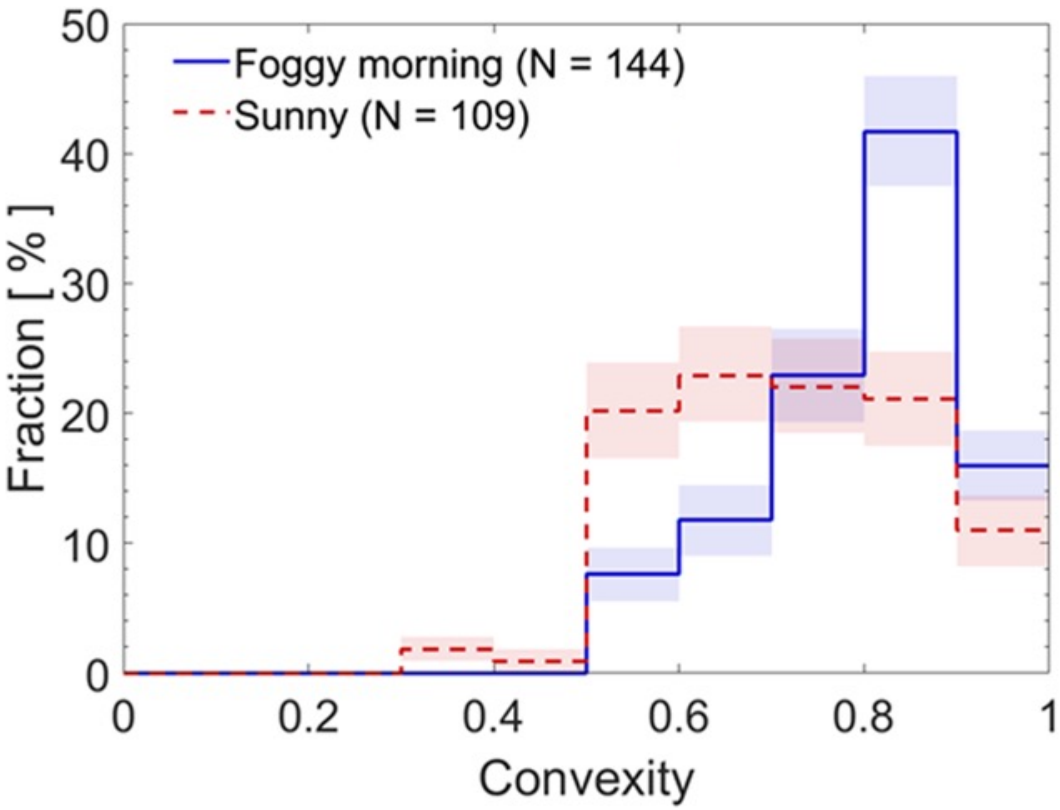


[1] Bhandari, Janarjan, et al. "Extensive soot compaction by cloud processing from laboratory and field observations." *Scientific reports* 9.1 (2019): 11824.

[2] Chen, Chao, et al. "Effect of organic coatings derived from the OH-initiated oxidation of amines on soot morphology and cloud activation." *Atmospheric Research* 239 (2020): 104905.

Assessing the role of capillary condensation in the atmosphere

- Field and chamber studies observed compact soot particles lacking visible coatings[1,2]
- We hypothesized that those results could be explained by cloud processing enabled by capillary condensation of trace chemicals



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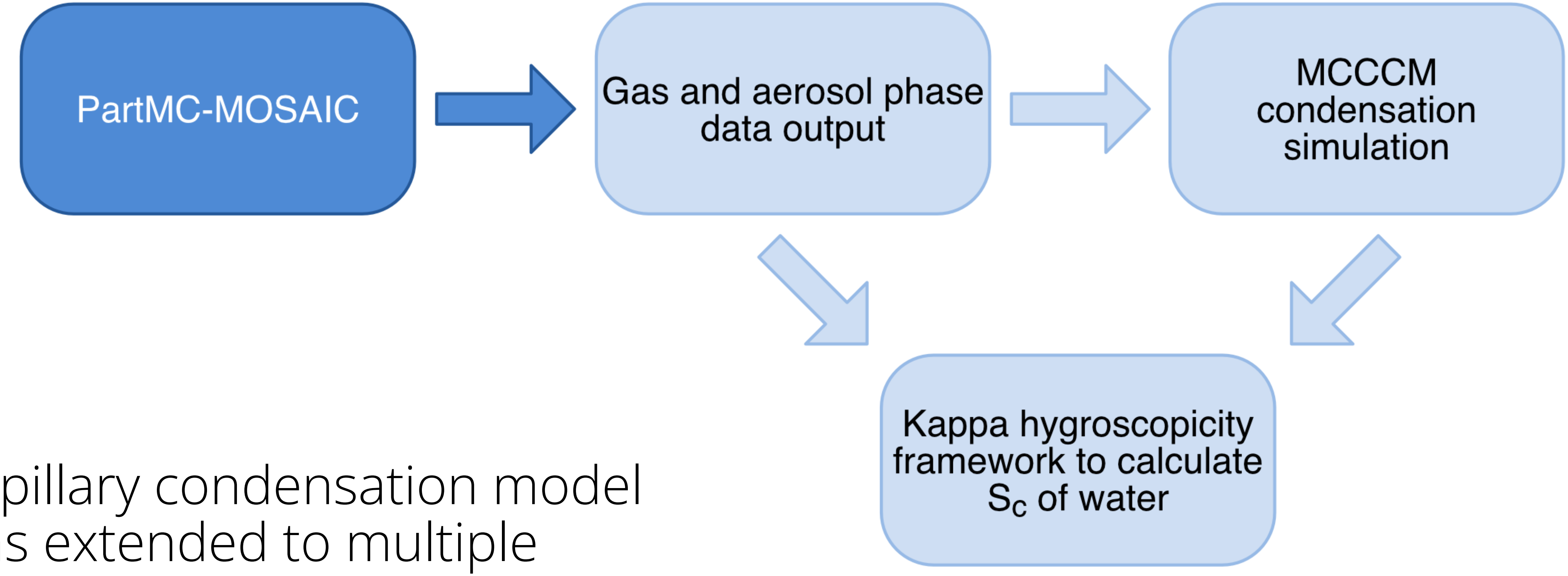
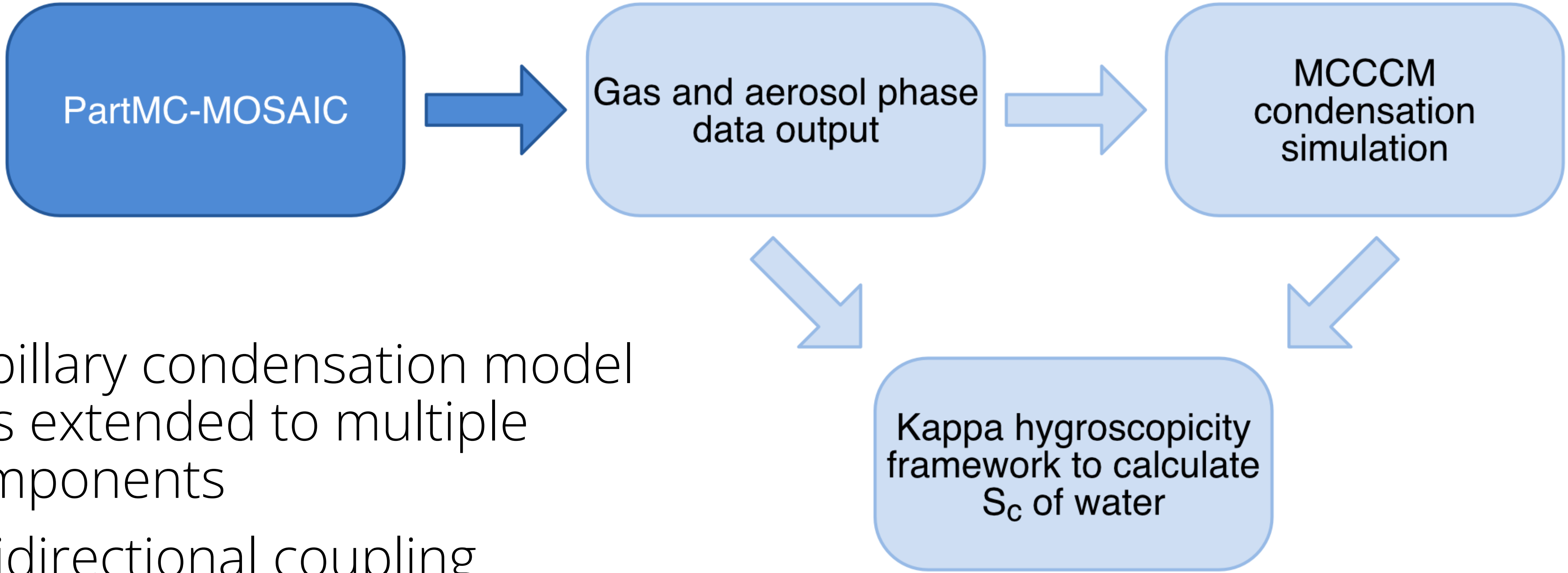
Coupling between PartMC-MOSAIC and MCCCM



- Capillary condensation model was extended to multiple components

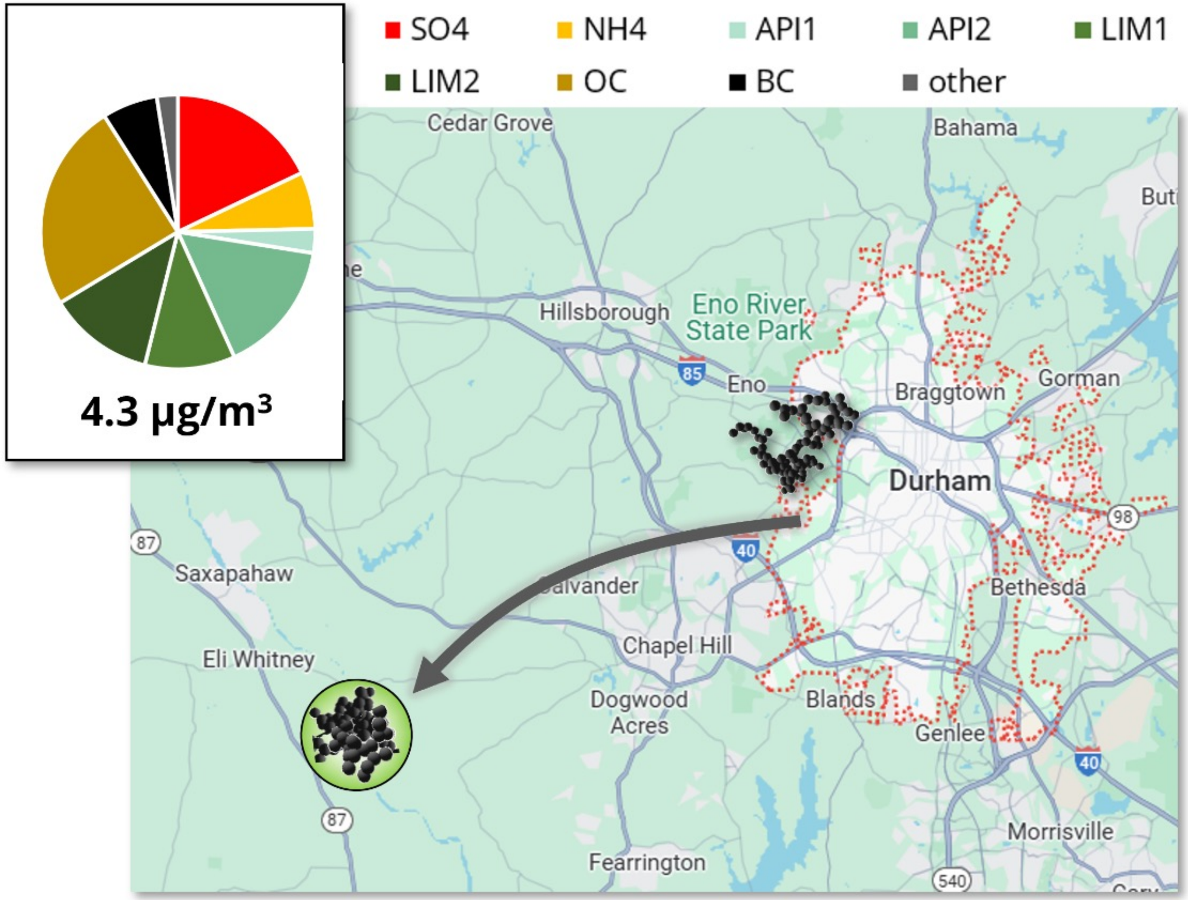
Coupling between PartMC-MOSAIC and MCCCM



- Capillary condensation model was extended to multiple components
- Unidirectional coupling between MCCCM and Part-MC for capillary condensation in atmospheric scenarios

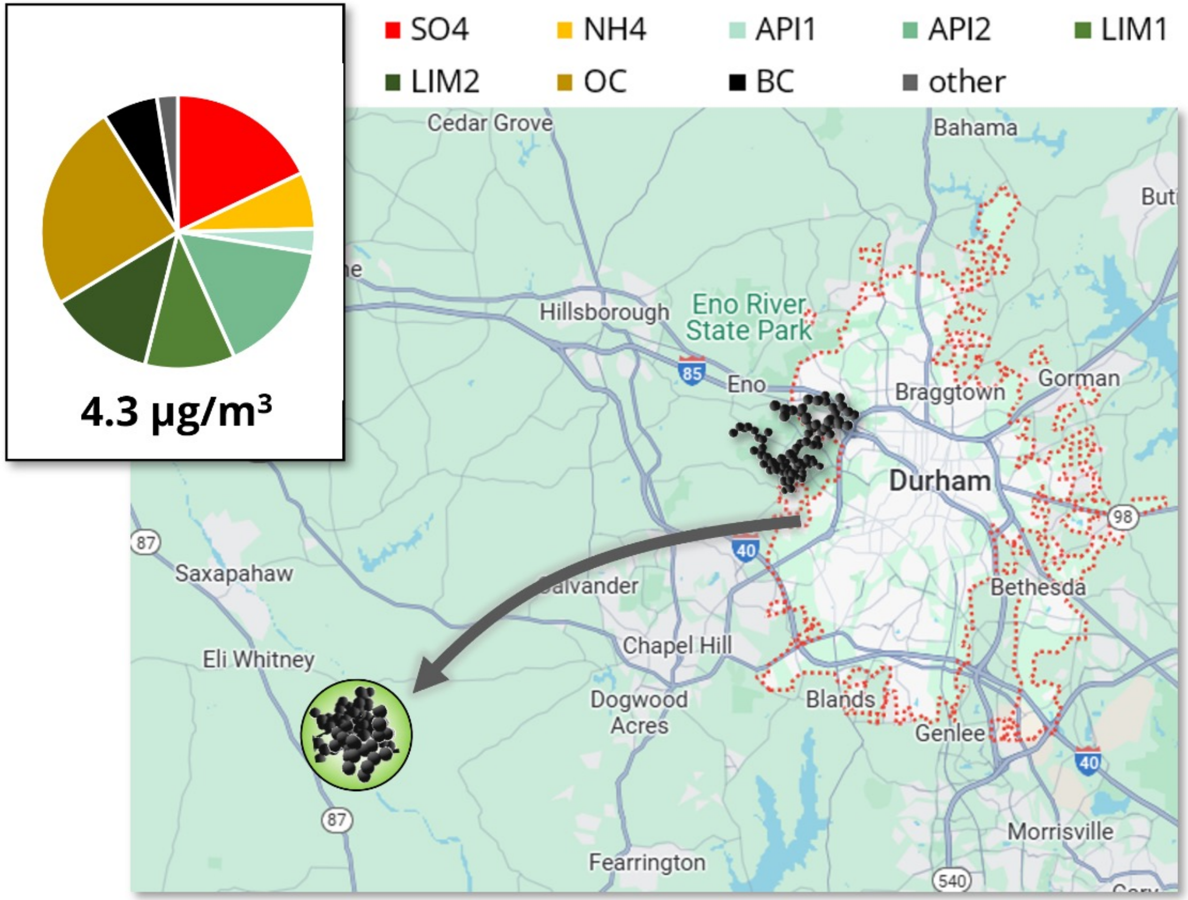
Implications for atmospheric processing

- PartMC-MOSAIC rural scenario
  - Soot emitted in an urban area at night
  - Travels over a forested area



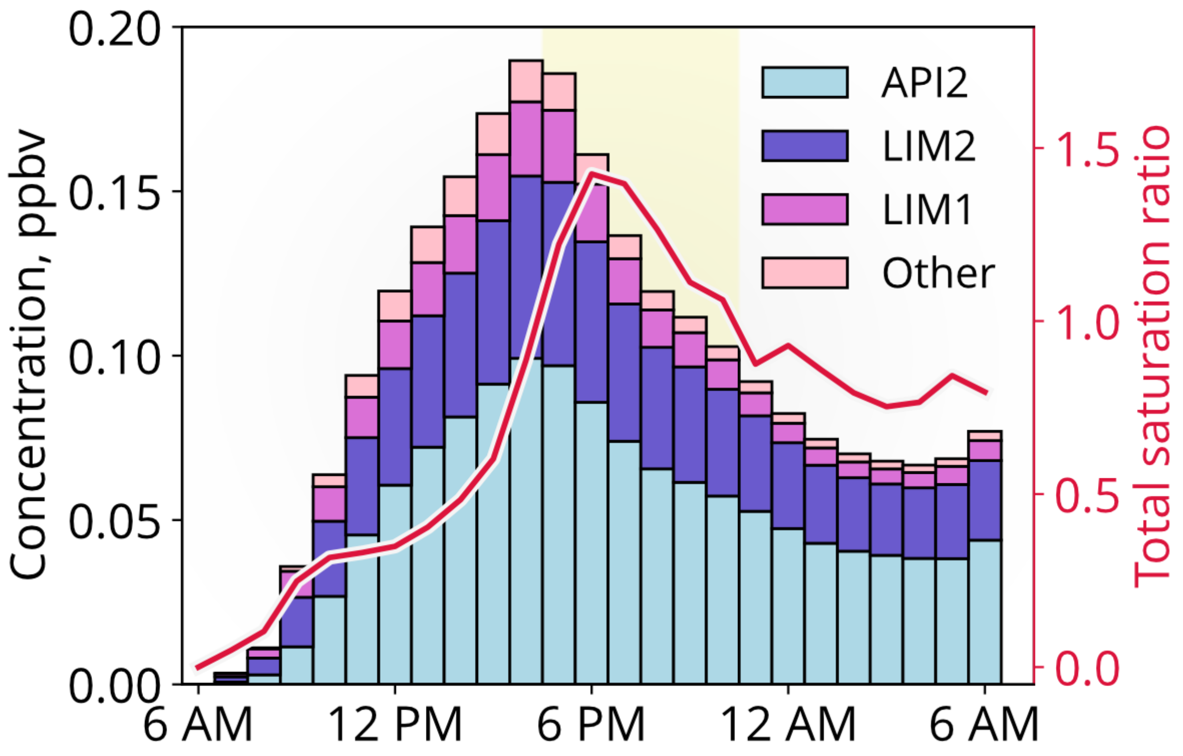
Implications for atmospheric processing

- PartMC-MOSAIC rural scenario
  - Soot emitted in an urban area at night
  - Travels over a forested area
- Aging by oxidation products of biogenic monoterpenes
  - α-pinene
  - limonene



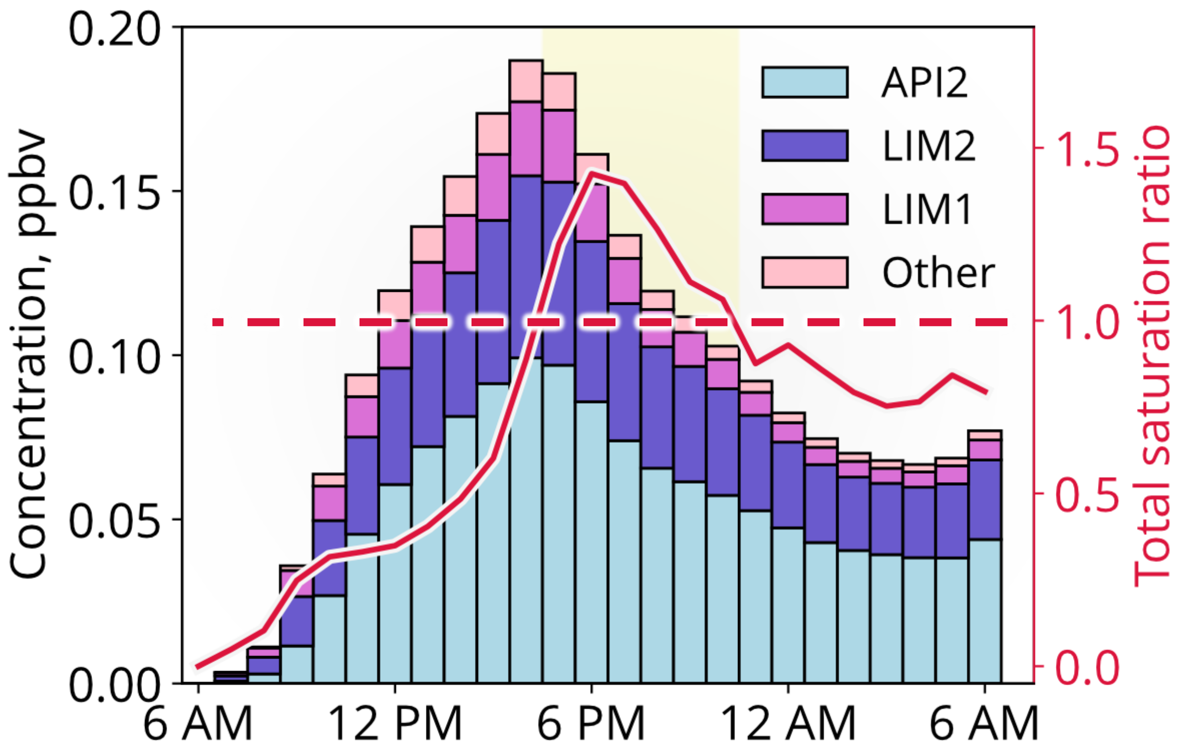
Gas-phase concentrations throughout the day

- Gas-phase concentrations of condensable compounds increase during daytime

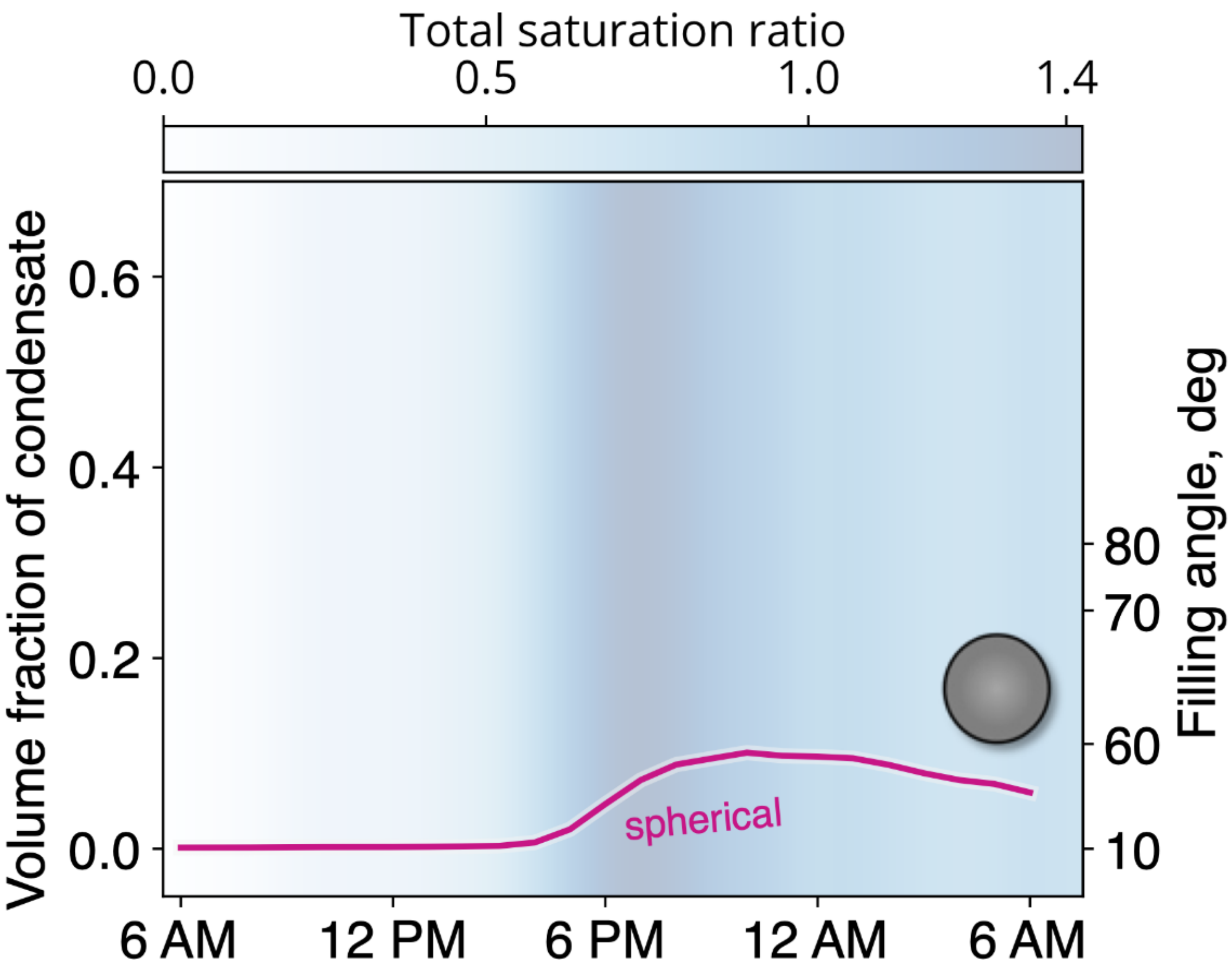


Gas-phase concentrations throughout the day

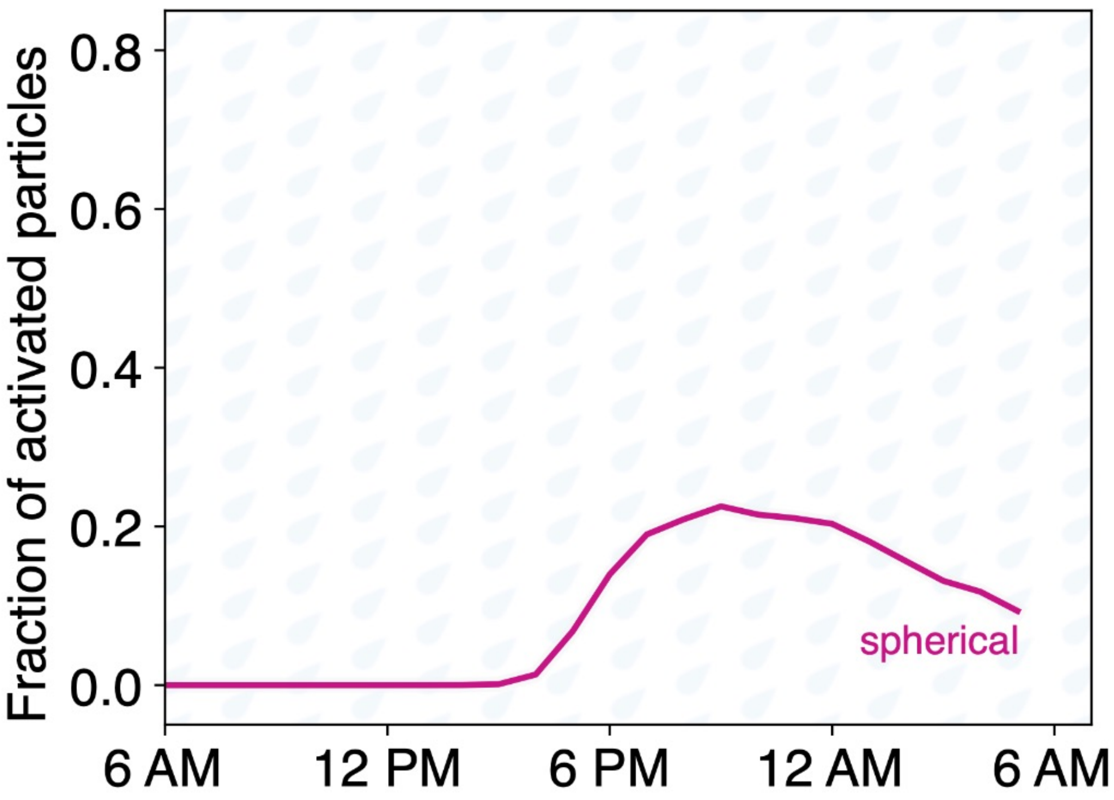
- Gas-phase concentrations of condensable compounds increase during daytime
- Total saturation ratio lags behind because it is controlled by air temperature



Trace chemical condensation – representation comparison



Critical water supersaturation < 0.2%



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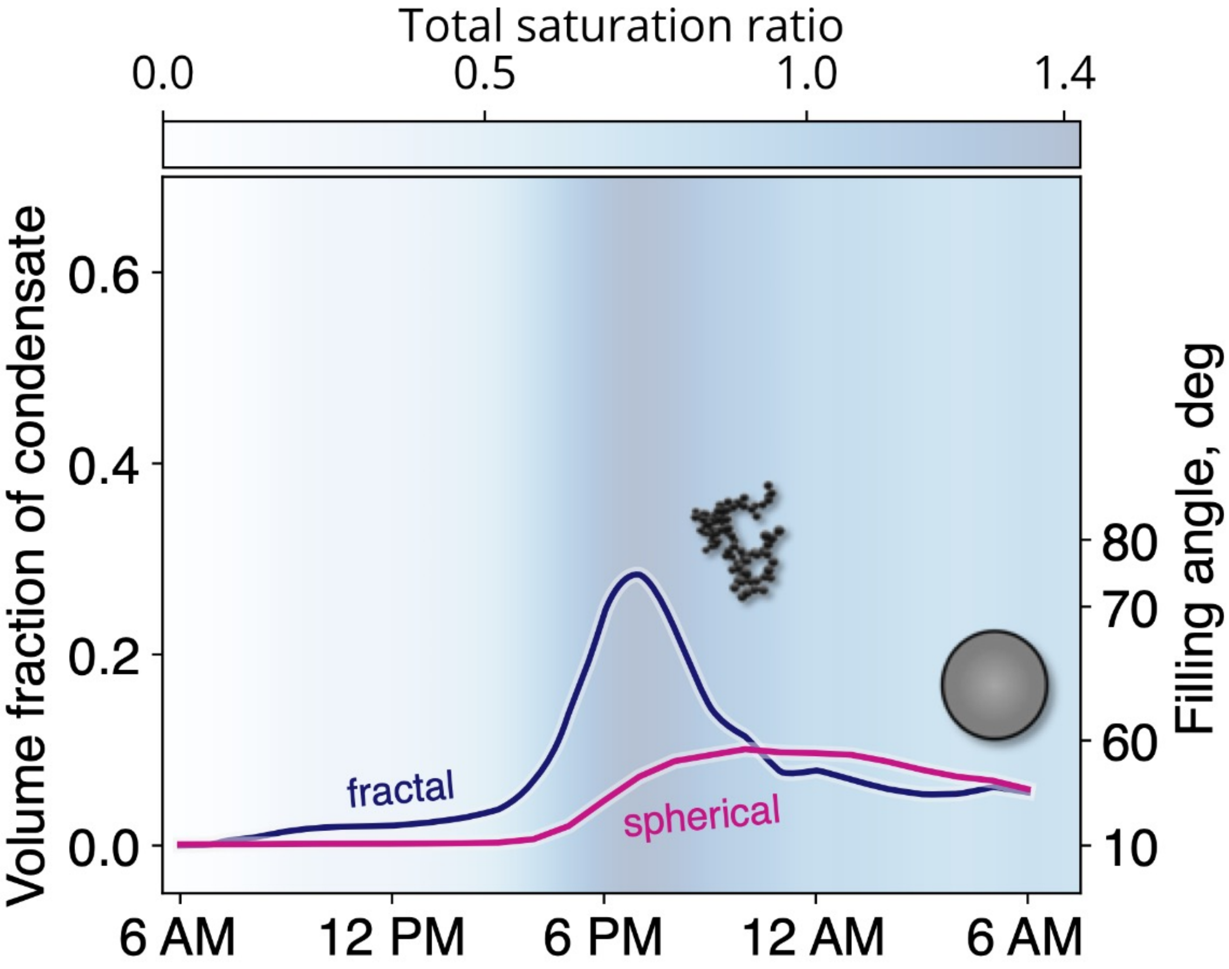

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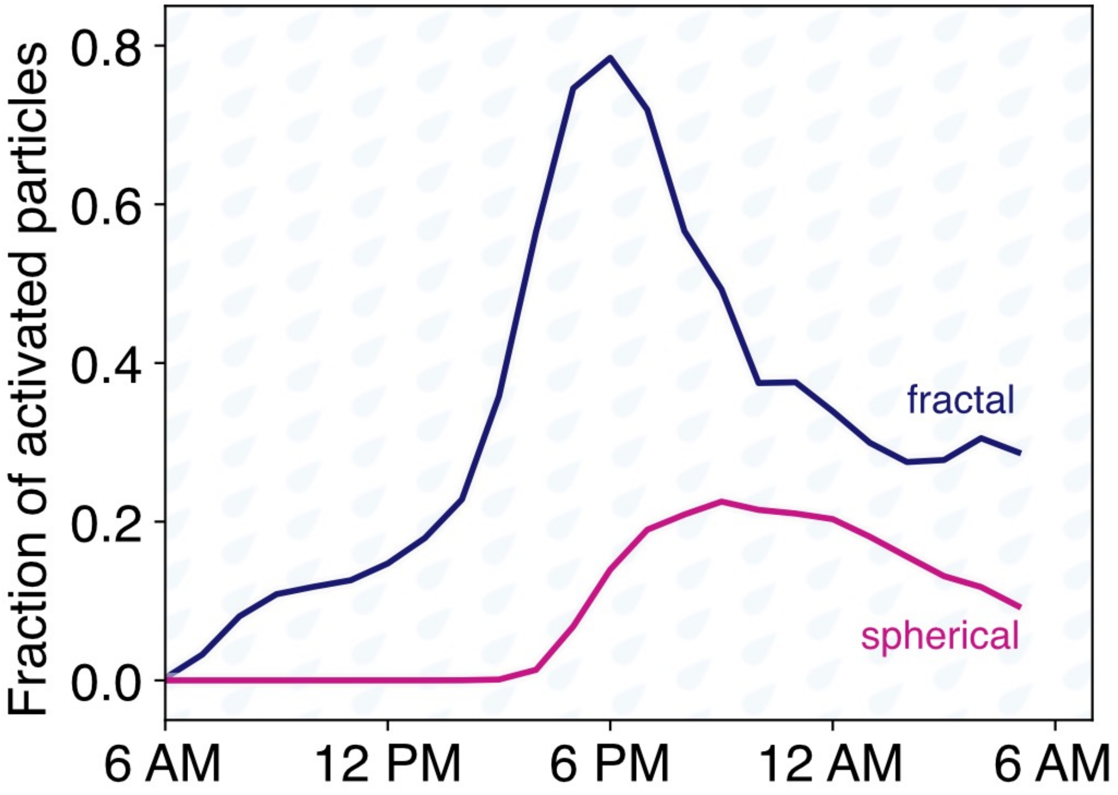

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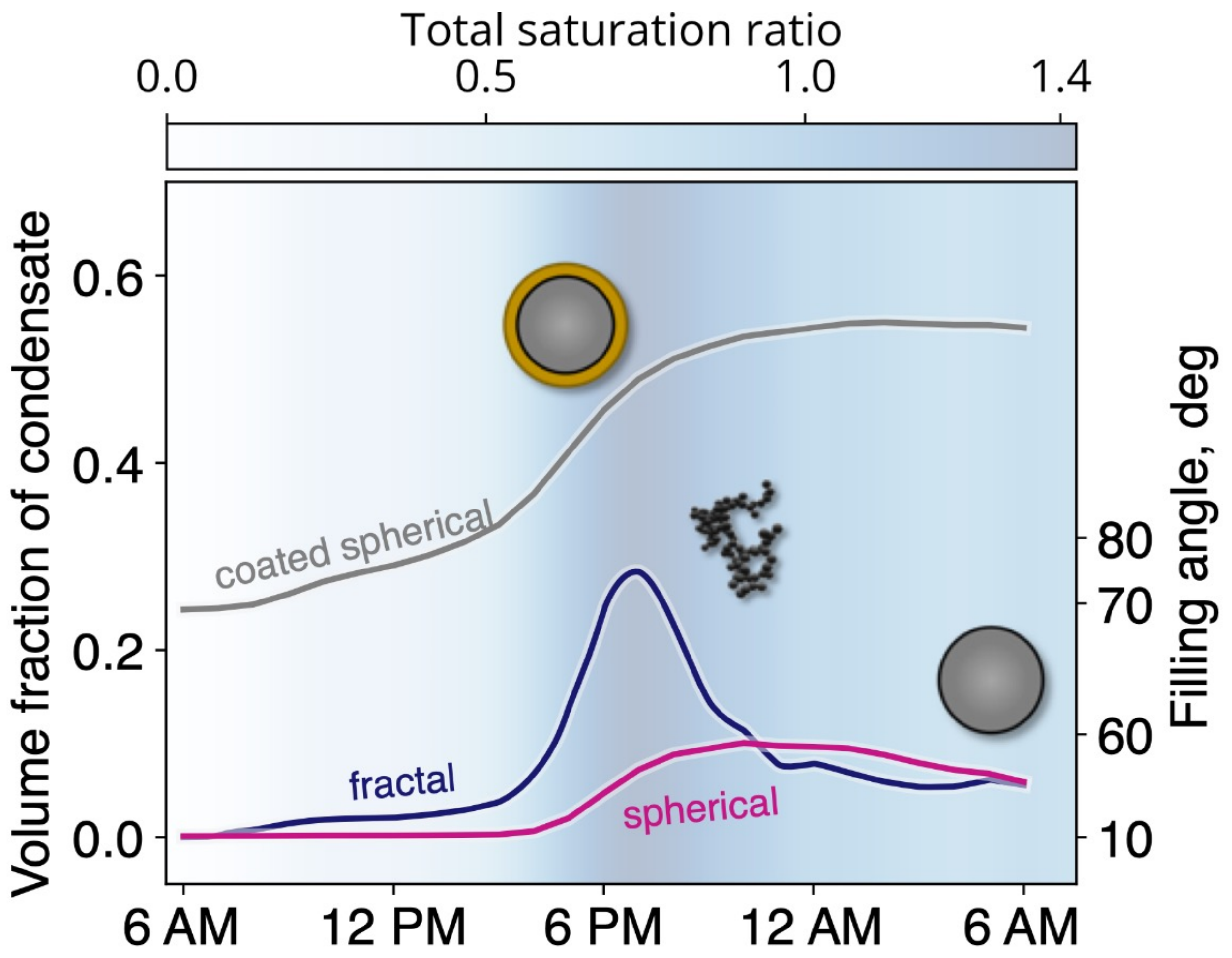

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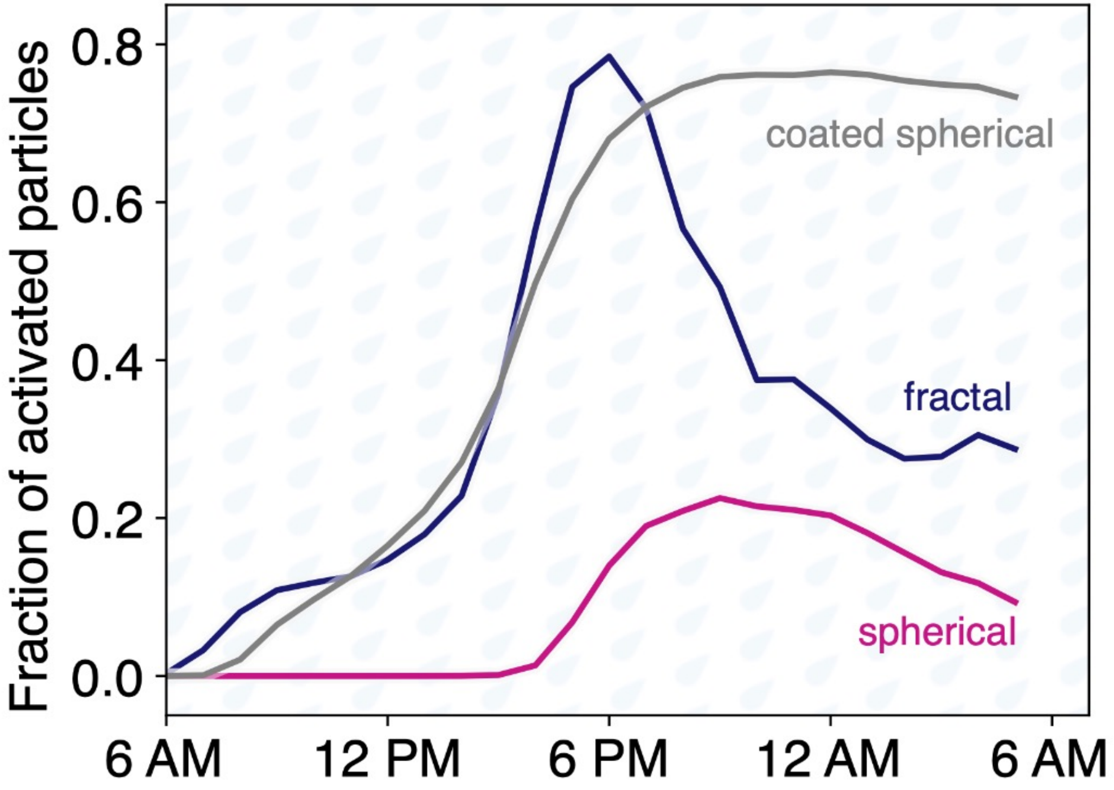

Conclusions

- Capillary condensation can explain experimental observations of compact bare soot
- More particles get activated in a shorter time when fractal morphology is accounted for
- Capillary condensation needs to be incorporated into aerosol models explicitly

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Acknowledgements

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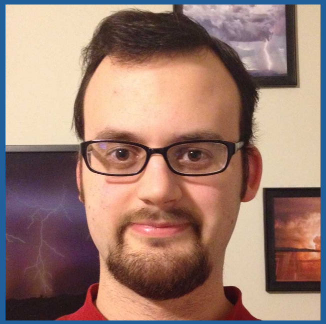

ILLINOIS

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PartMC

NSF

Appendix A: capillary condensation model

$$\frac{dV}{dt} = \frac{1}{4} A \alpha \nu_{\mathsf{T}} \frac{V_{\mathsf{m}} p_{\mathsf{sat,b}}}{N_{\mathsf{A}} R_{\mathsf{g}} T} \left(\nu - \beta \exp \left[\frac{1}{2} \ell_{\mathsf{K}} \kappa \right] \right)$$

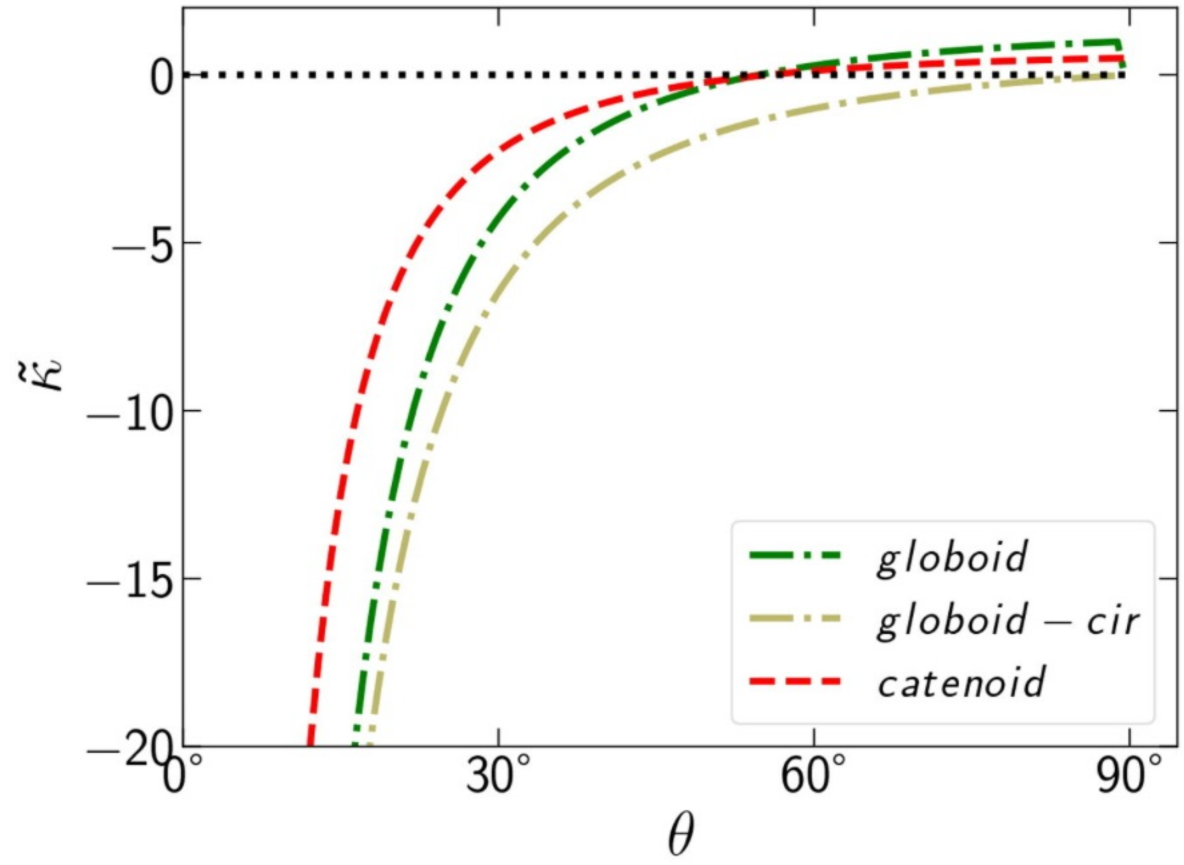
$$\nu_{\mathsf{T}} = \sqrt{\frac{8 R_{\mathsf{g}} T}{\pi M}}$$

$$\beta = \frac{p_{\mathsf{sat,p}}}{p_{\mathsf{sat,b}}}$$

$$\ell_{\mathsf{K}} = \frac{2 \gamma_{\mathsf{LG}} V_{\mathsf{m}}}{R_{\mathsf{g}} T}$$

$$\nu = \frac{p_{\mathsf{gas}}}{p_{\mathsf{sat,b}}}$$

Appendix B: coating mean curvature



Appendix C: kappa hygroscopicity theory

$$\kappa = \frac{4A^3}{27 D_\mathsf{d}^3 \ln^2 S_\mathsf{c}}$$

$$A = \frac{4\sigma_{\mathsf{s/a}} M_\mathsf{w}}{RT\rho_\mathsf{w}}$$

Appendix D: size distribution of BC