The Roles of Interfacial Energy and Size-Dependent Morphologies of Atmospheric Aerosols

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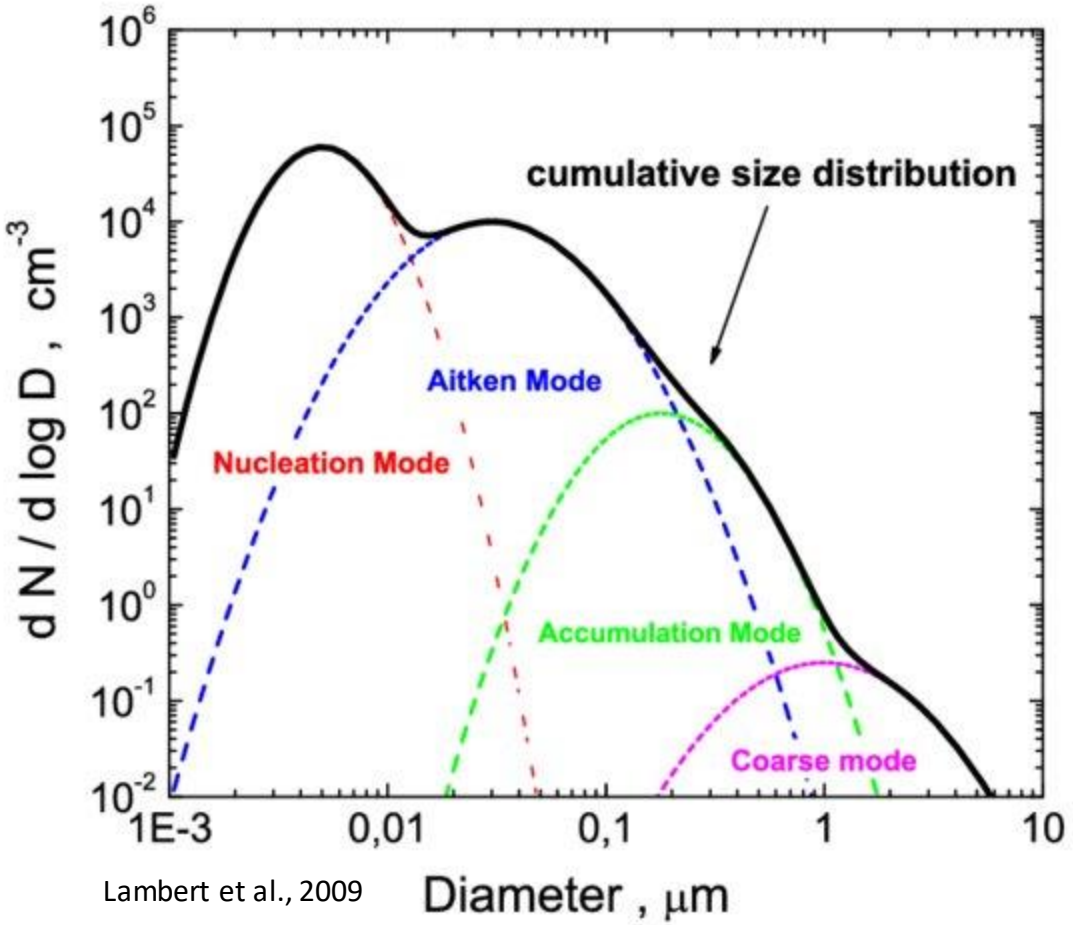
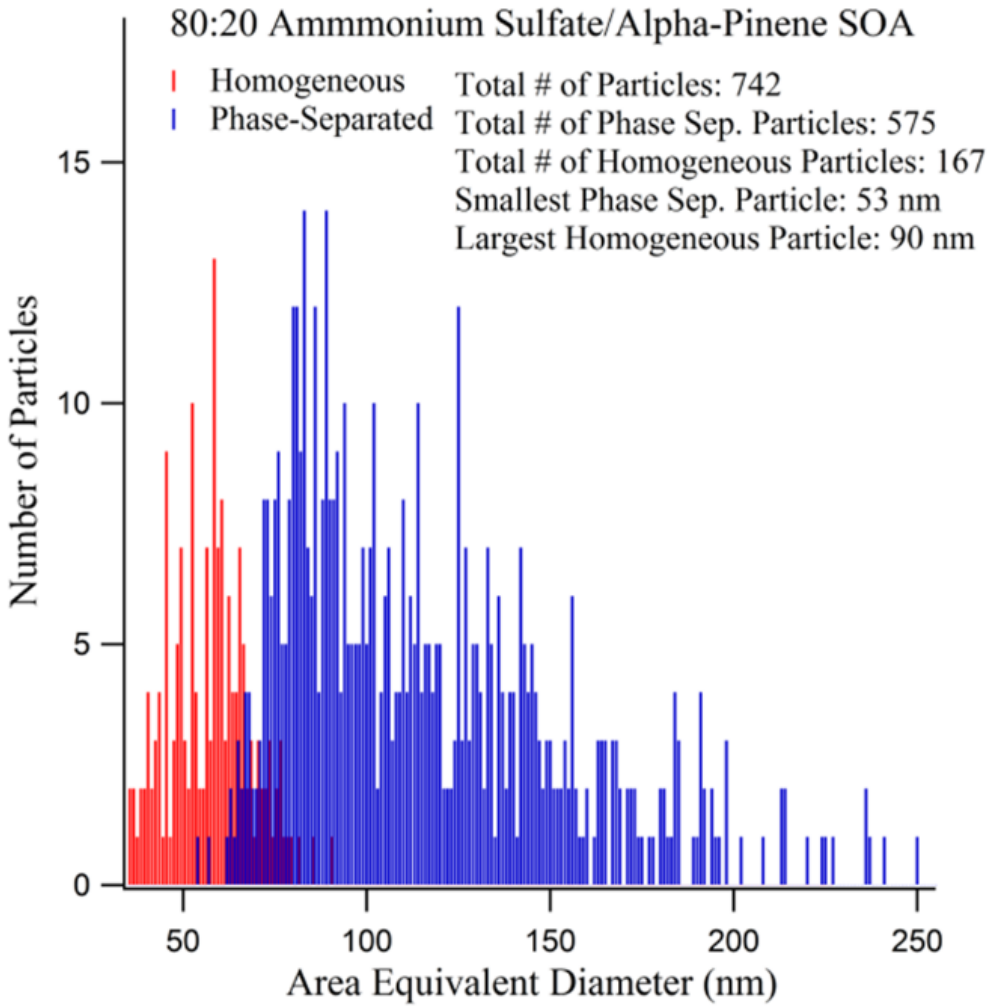
McGill

The Smallest Aerosol Particles Are The Most Numerous



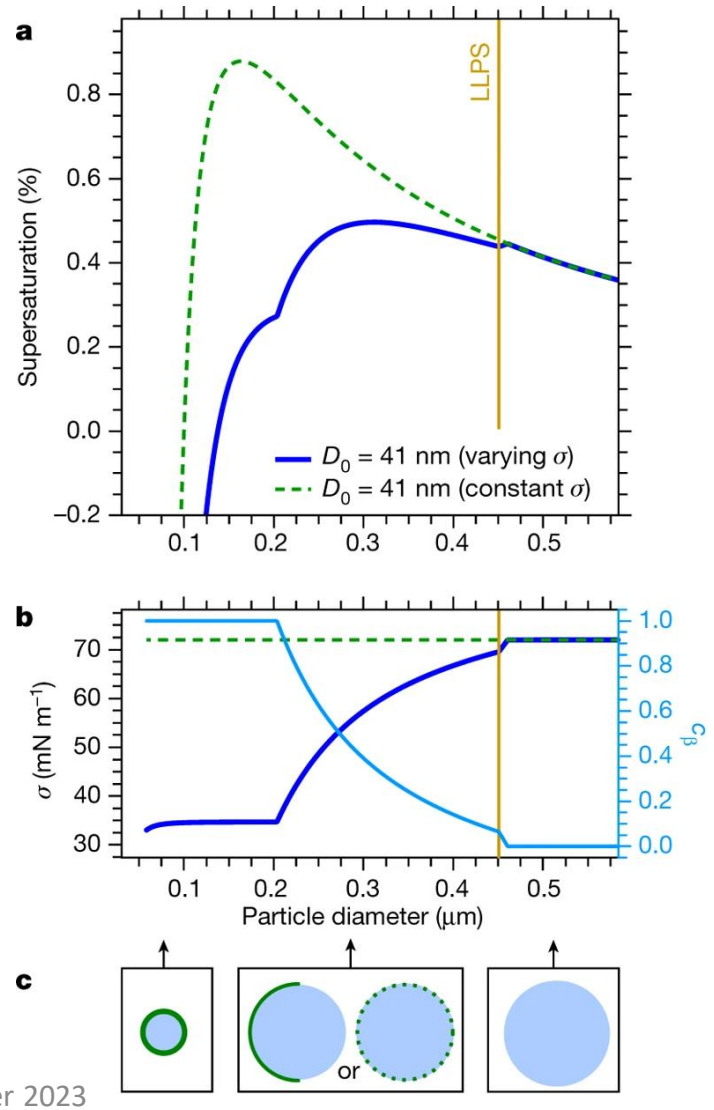
Lambert et al., 2009

Smaller Particles May Behave Differently Than Large ones

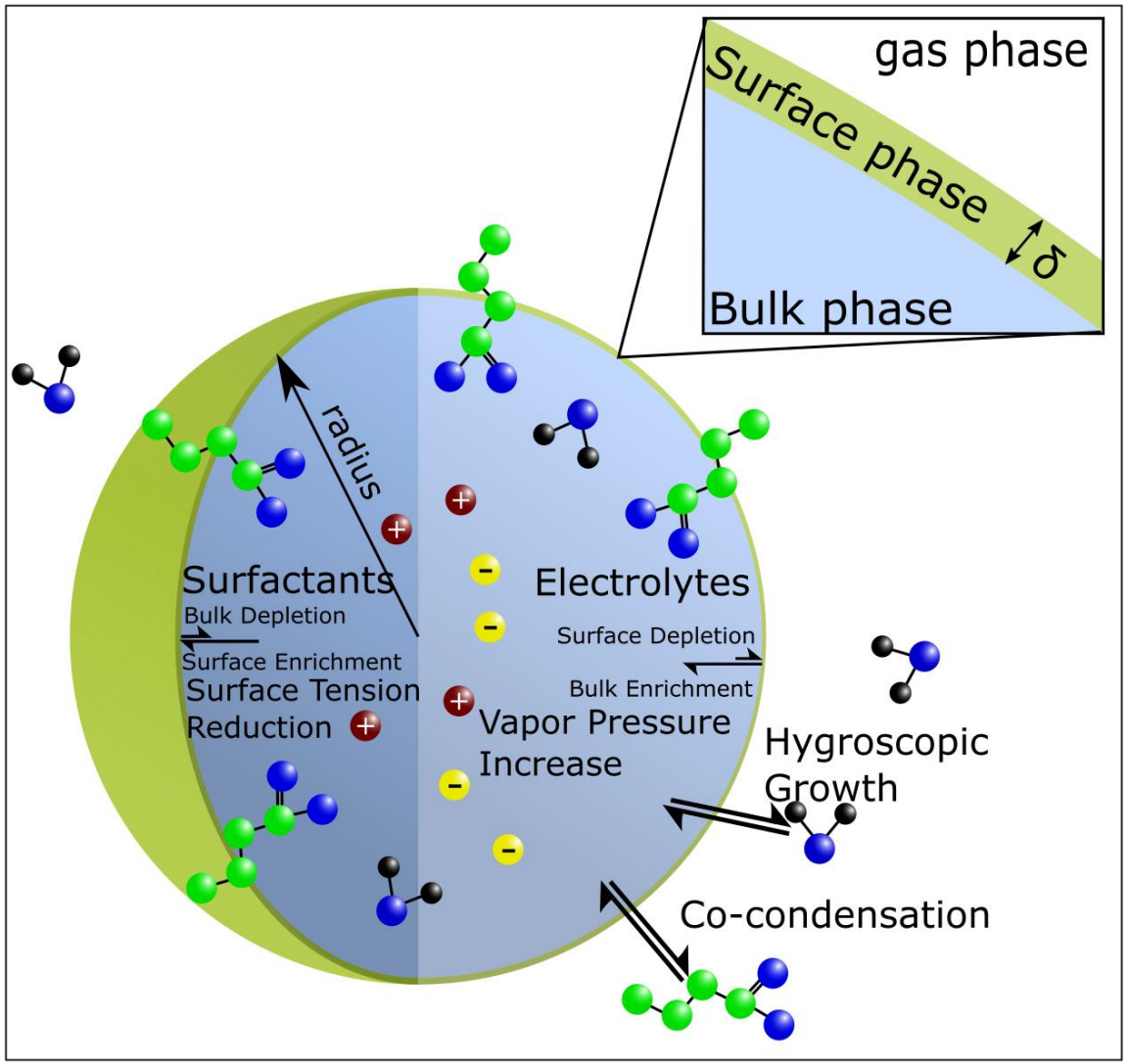


Kucinski et al., 2019

Surface Effects Can Modify Aerosol Properties

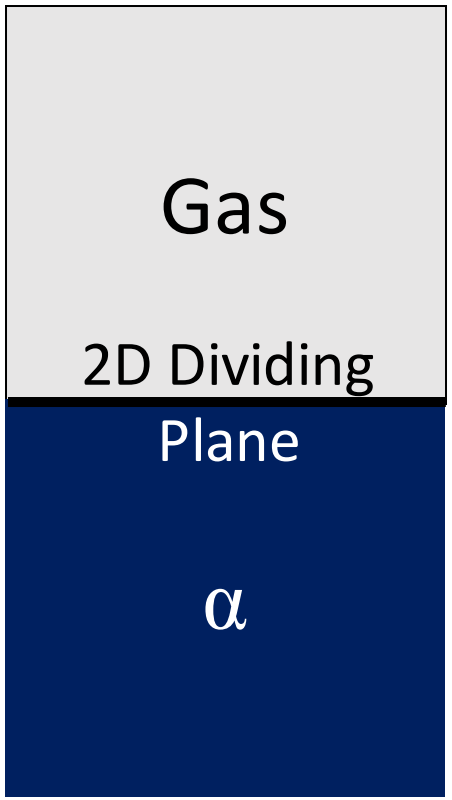


Ovadnevaite et al., 2017

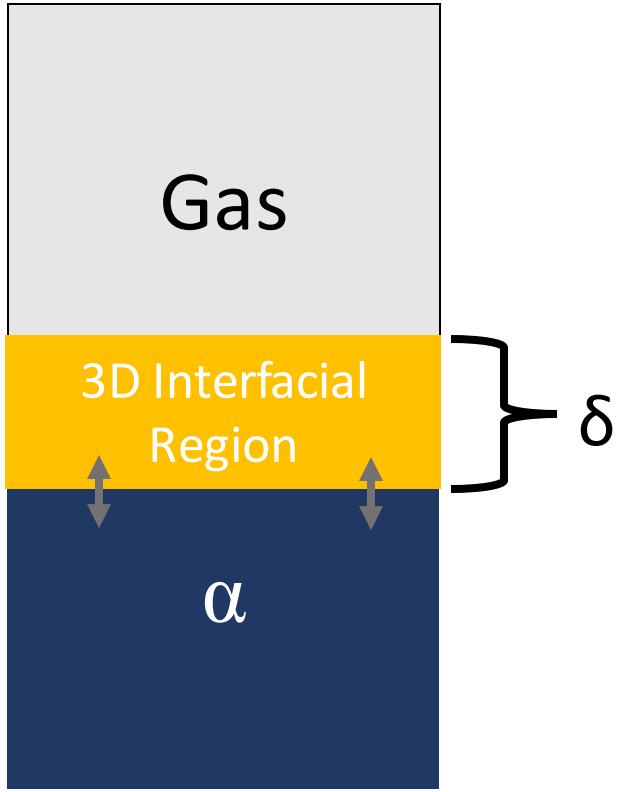


Schmedding & Zuend, 2023

Real Surfaces Have Thin But Finite Depths



More Idealized System



More Realistic System

Surface Tension Can Be Modeled Using AIOMFAC

$$\mu_i^{surf} = \xi_i^{surf} - \sigma \mathcal{A}_i$$

$$\xi_i^{surf} = \xi_i^{surf,o} + RT\ln(a_i^{surf})$$

$$\mu_i^{surf,o} = \xi_i^{surf,o} - \sigma_i^o \mathcal{A}_i^{surf,o}$$

$$\boldsymbol{a_i^{surf} = x_i^{surf}\left(\gamma_i^{AIOMFAC}\right)^t}$$

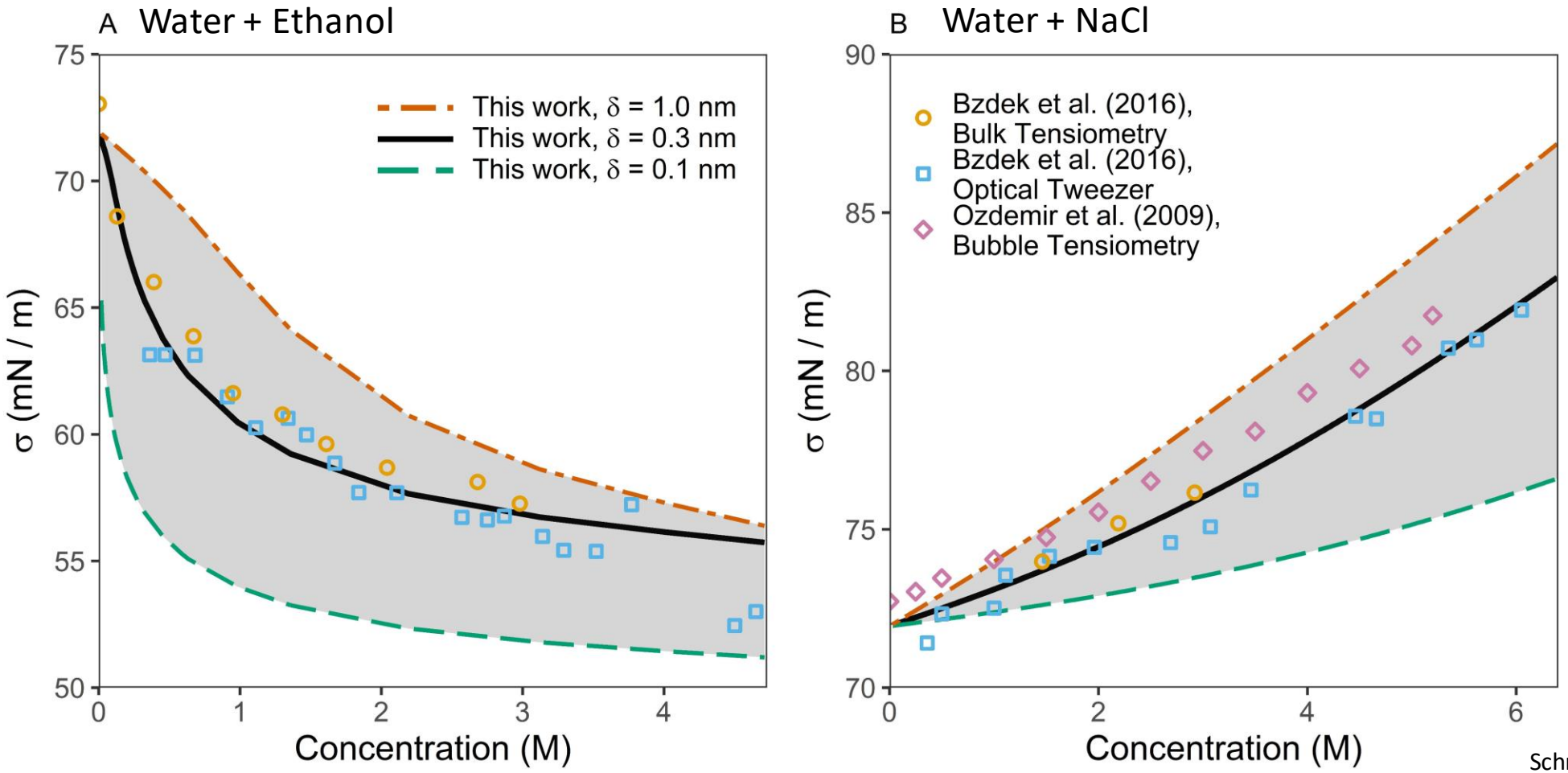
$$\mu_i^{surf} = \xi_i^{surf,o} + RT\ln\left(a_i^{surf}\right) + \mathcal{A}_i^o \sigma_i^o - \mathcal{A}_i \sigma$$

$$\mathcal{A}_i^o \cong \mathcal{A}_i$$

$$\boldsymbol{\frac{RT}{\mathcal{A}_i}\ln\left(\frac{a_i^{surf}}{a_i^{bulk}}\right) + \sigma_i^o = \sigma}$$

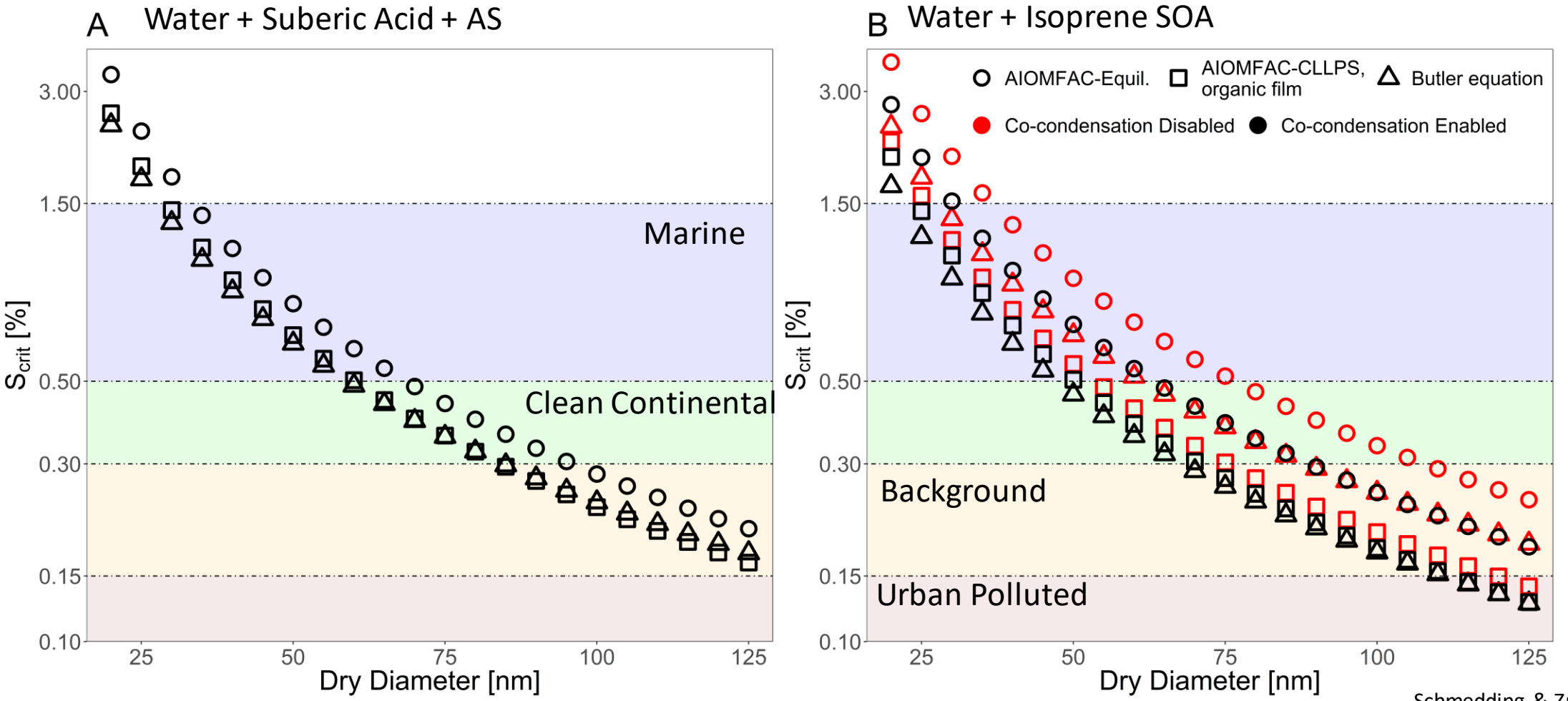
Aston & Herrington, 1994
Lane, 1983

Surface Tension Predictions Match Experimental Data



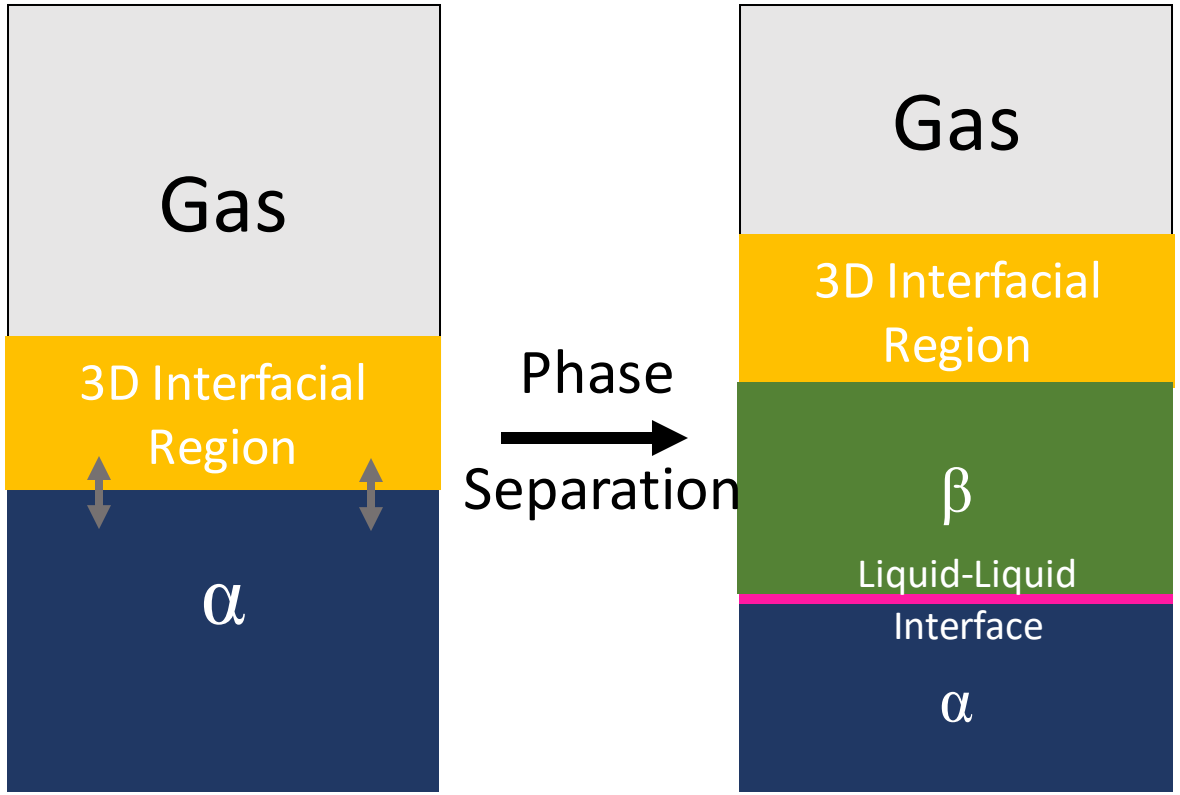
Schmedding & Zuend, 2023

The Inclusion of Bulk-Surface Partitioning Modifies Cloud Activation Conditions



Schmedding & Zuend, 2023

Interfacial Tension Modeling Is Similar To Surface Tension



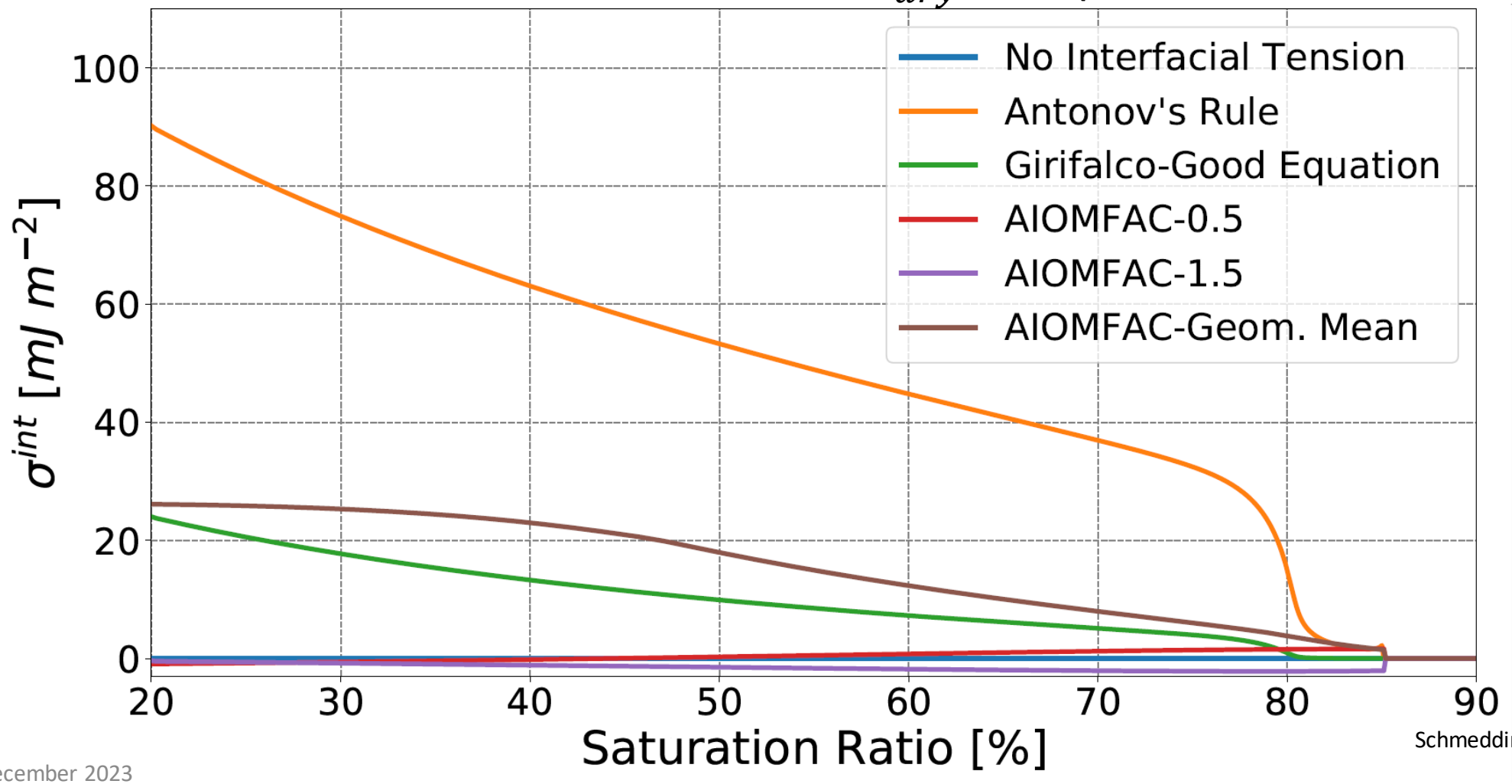
$$\frac{RT}{\mathcal{A}_i^{int}} \ln\left(\frac{a_i^{int}}{a_i^{bulk}}\right) + \sigma_i^{o} = \sigma^{int}$$

Interfacial Tension Treatments Can Be Simplified

Name	Treatment of Interfacial Tension
No Interfacial Tension	$\sigma^{IF} = 0$
Antonov's Rule	$\sigma^{IF} = \sigma^{\alpha} - \sigma^{\beta}$
Girifalco-Good Equation	$\sigma^{IF} = \sigma^{\alpha} + \sigma^{\beta} - 2\varphi \left(\sigma^{\alpha}\sigma^{\beta}\right)^{\frac{1}{2}}$
AIOMFAC-0.5	$\sigma_i^{IF} = \frac{RT}{\mathscr{A}_i} \ln\left(\frac{a_i^{IF}}{a_i^{b}}\right);\ \gamma_i^{IF} = \gamma_i^{AIOMFAC\,\frac{1}{2}}$
AIOMFAC-1.5	$\sigma_i^{IF} = \frac{RT}{\mathscr{A}_i} \ln\left(\frac{a_i^{IF}}{a_i^{b}}\right);\ \gamma_i^{IF} = \gamma_i^{AIOMFAC\,\frac{3}{2}}$
AIOMFAC-Geom-Mean	$\sigma_i^{IF} = \frac{RT}{\mathscr{A}_i} \ln\left(\frac{a_i^{IF}}{a_i^{b}}\right);\ \gamma_i^{IF} = \left(\gamma_i^{\alpha}\gamma_i^{\beta}\right)^{\frac{1}{2}}$

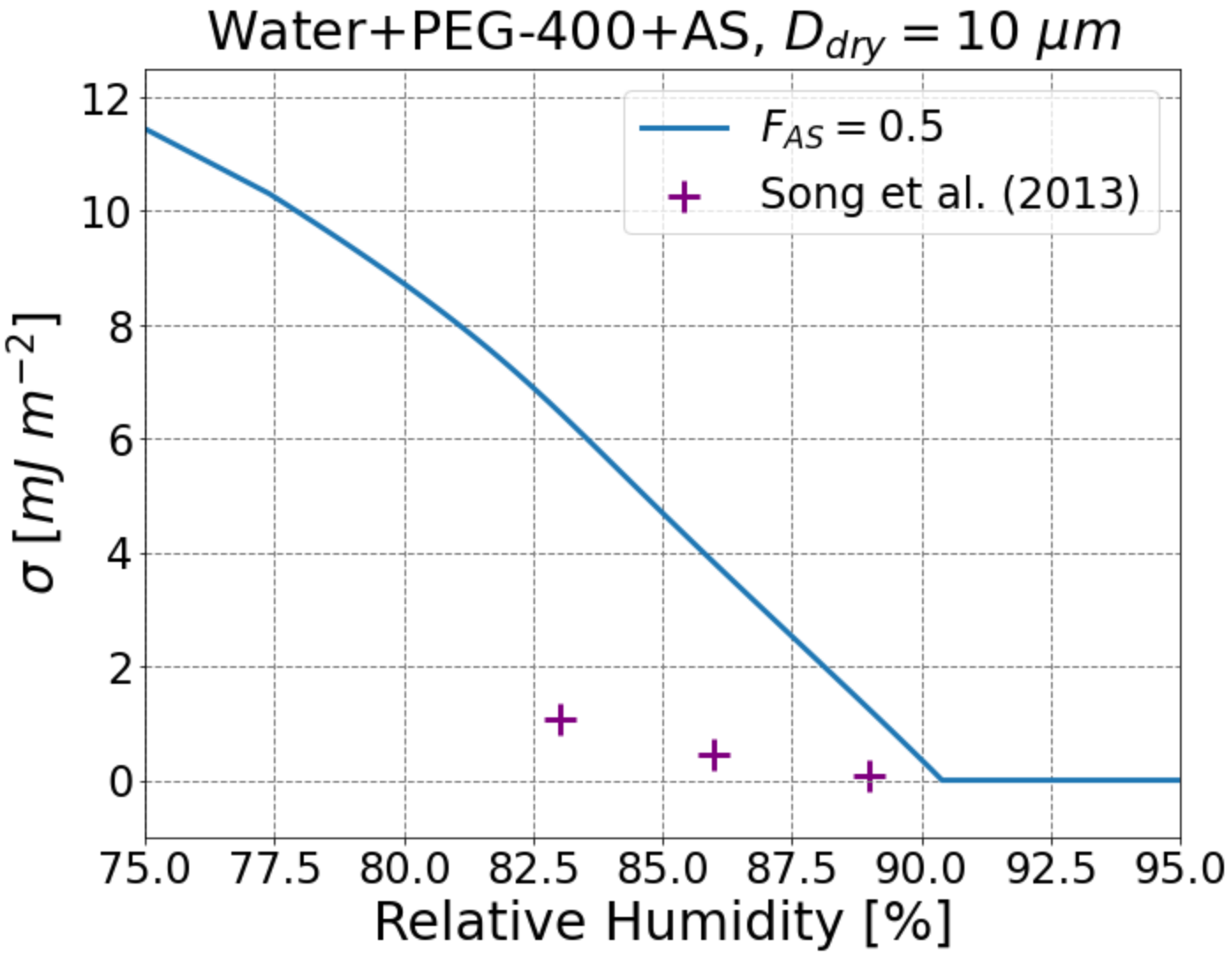
Some Interfacial Tension Treatments Are More Realistic Than Others

Water + PEG-300 + AS, $D_{dry} = 10\mu m$



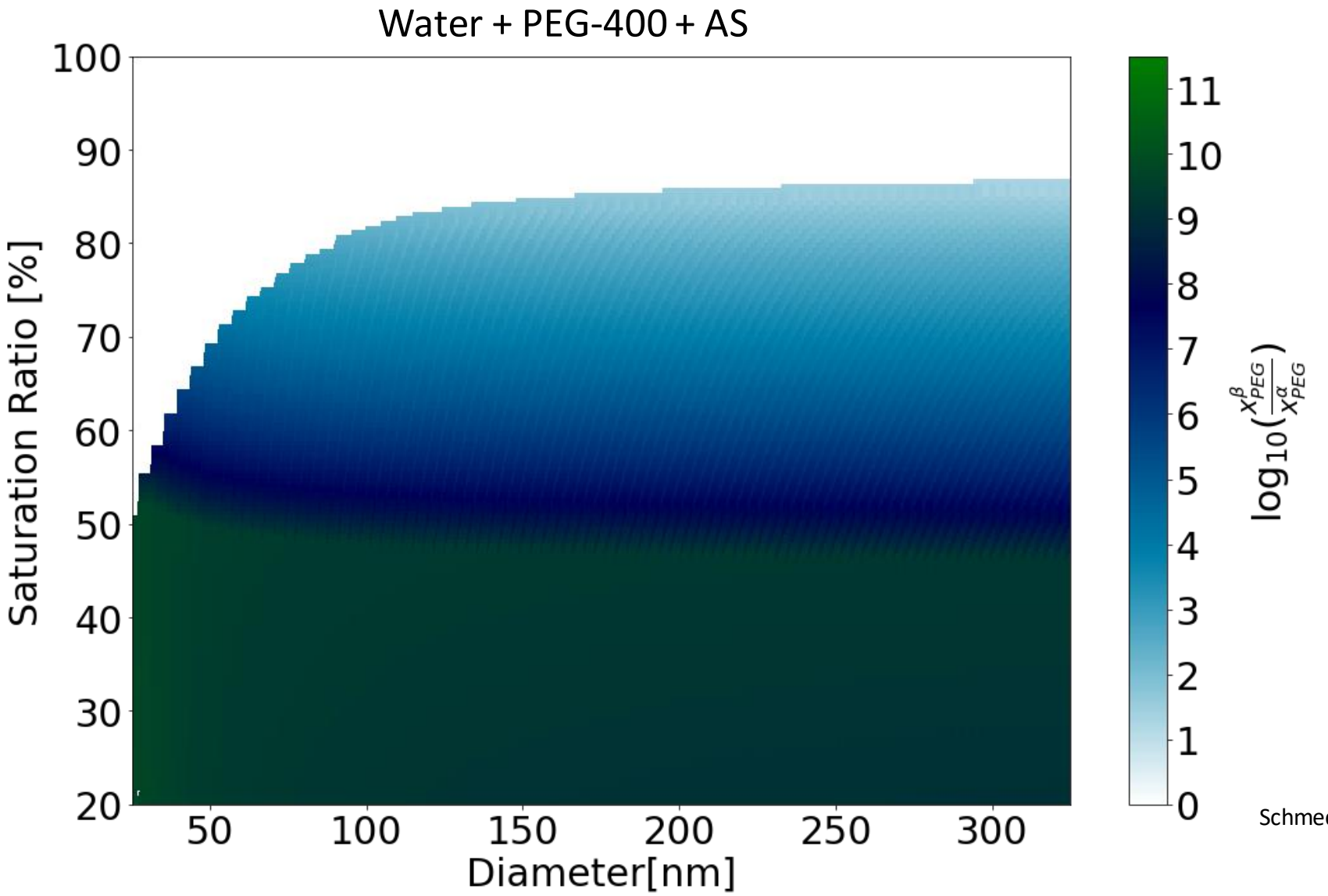
Schmedding & Zuend, in prep

There Are Few Measurements of Aerosol Interfacial Tension



Schmedding & Zuend, in prep

Smaller Particles Still Phase Separate, But At Lower Relative Humidities



Schmedding & Zuend, in prep

Conclusions

- AIOMFAC estimated surface tensions agree with bulk measurements
- Including surface tension estimates reduces critical superstaturations necessary for cloud droplet activation in Koehler curves
- Using the Girifalco-Good Equation or geometric mean of activity coefficients gives reasonable values for interfacial tension in aerosol particles
- AIOMFAC predicted relative humidity of phase separation decreases with decreasing particle size

Questions?

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