Supporting Information: A Conceptual Framework for the Influence of Water Activity Parameterization on Hygroscopic Growth and Cloud Droplet Activation in Aerosols Containing Strong Nonionic Surfactants

Alison Bain*

Department of Chemistry, Oregon State University, Corvallis, OR. USA.

E-mail: alison.bain@oregonstate.edu

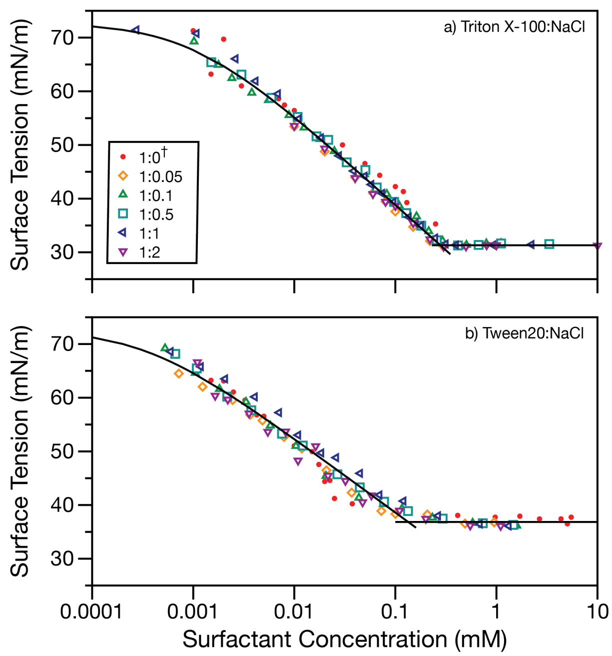

Figure S1: Macroscopic surface tension data for a) Triton X-100 and B) Tween20 in different mole ratio mixtures with NaCl. Langmuir isotherm and plateau region fits for all surfactant-NaCl mole ratios combined are overlaid on each panel. †Filled circles for binary surfactant-water data shown for Triton X-100 from Ref. 1 and binary surfactant-water data for Tween20 from Ref. 2 are shown for comparison.

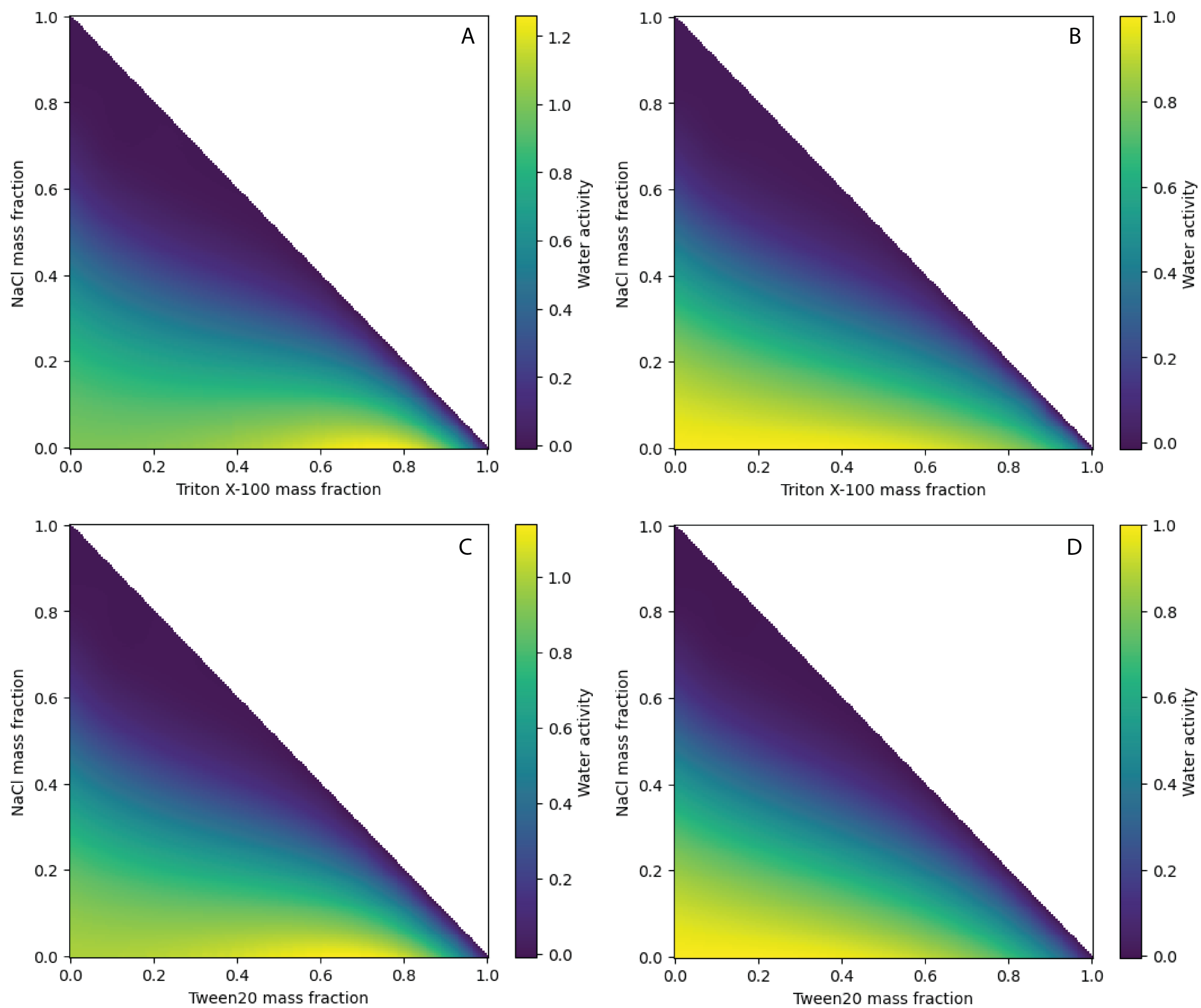

Figure S2: Interpolated water activity from AIOMFAC calculations for Triton X-100 (A/B) and Tween20 (C/D) using the ether-alkyl subgroup description (A/C) and the oxyethylene subgroup description (B/D). Note the corresponding color bar spans the range of calculated water activities for each case. Using the ether-alkyl subgroup descriptions, AIOMFAC outputs water activities greater than one at some solute mass ratios.

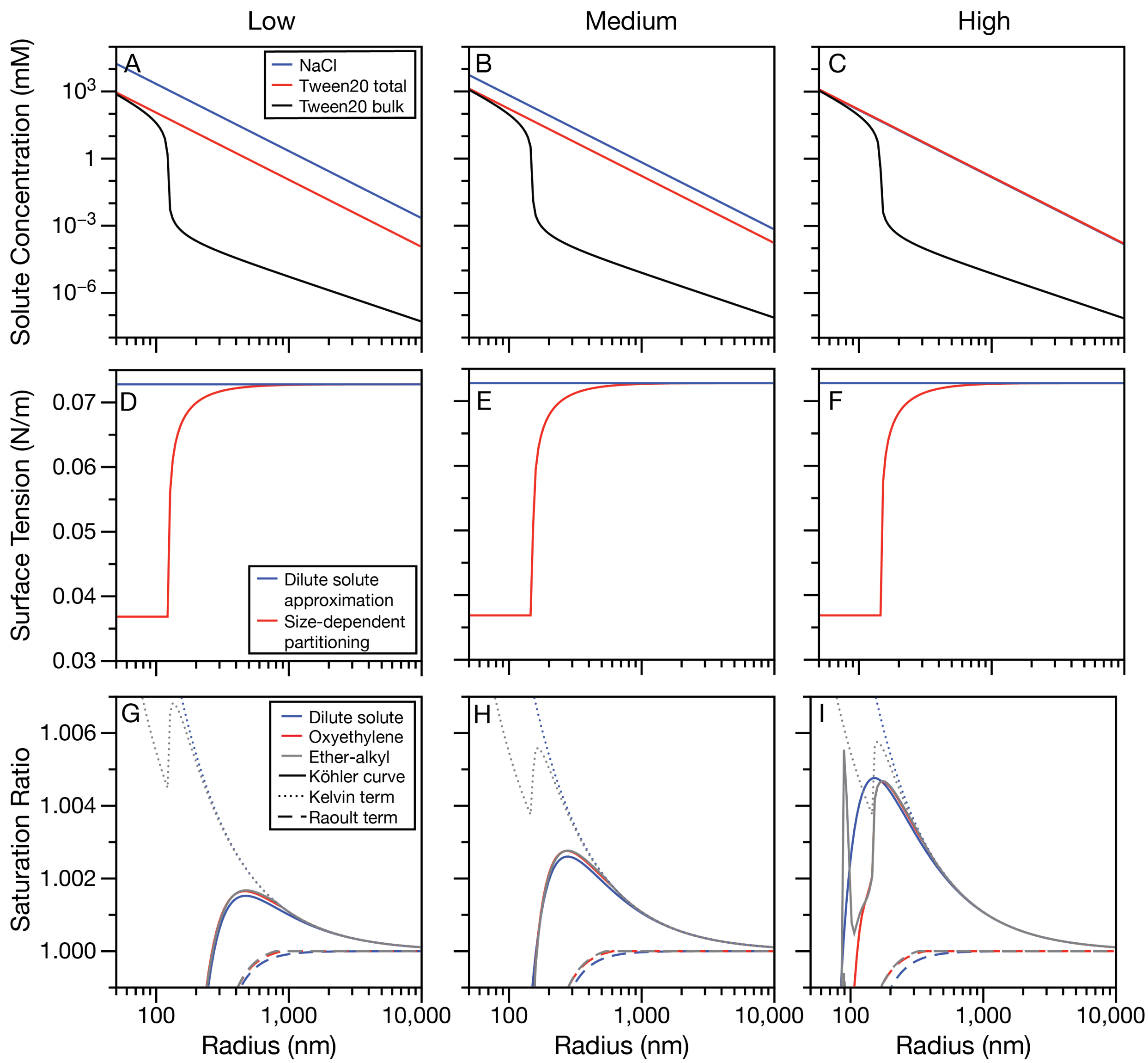

Figure S3: Solute concentration (A-C), surface tension (D-E), and saturation ratio (G-I) for 50 nm radius dry particles containing low (column 1, $w_{org} = 0.522$), medium (columns 2, $w_{org} = 0.838$), and high (column 3, $w_{org} = 0.660$) Tween20 mass fractions. Köhler curves are calculated using the dilute solute approximation as well as using the AIOMFAC activity interpolations using the oxyethylene and ether-alkyl subgroup descriptions.

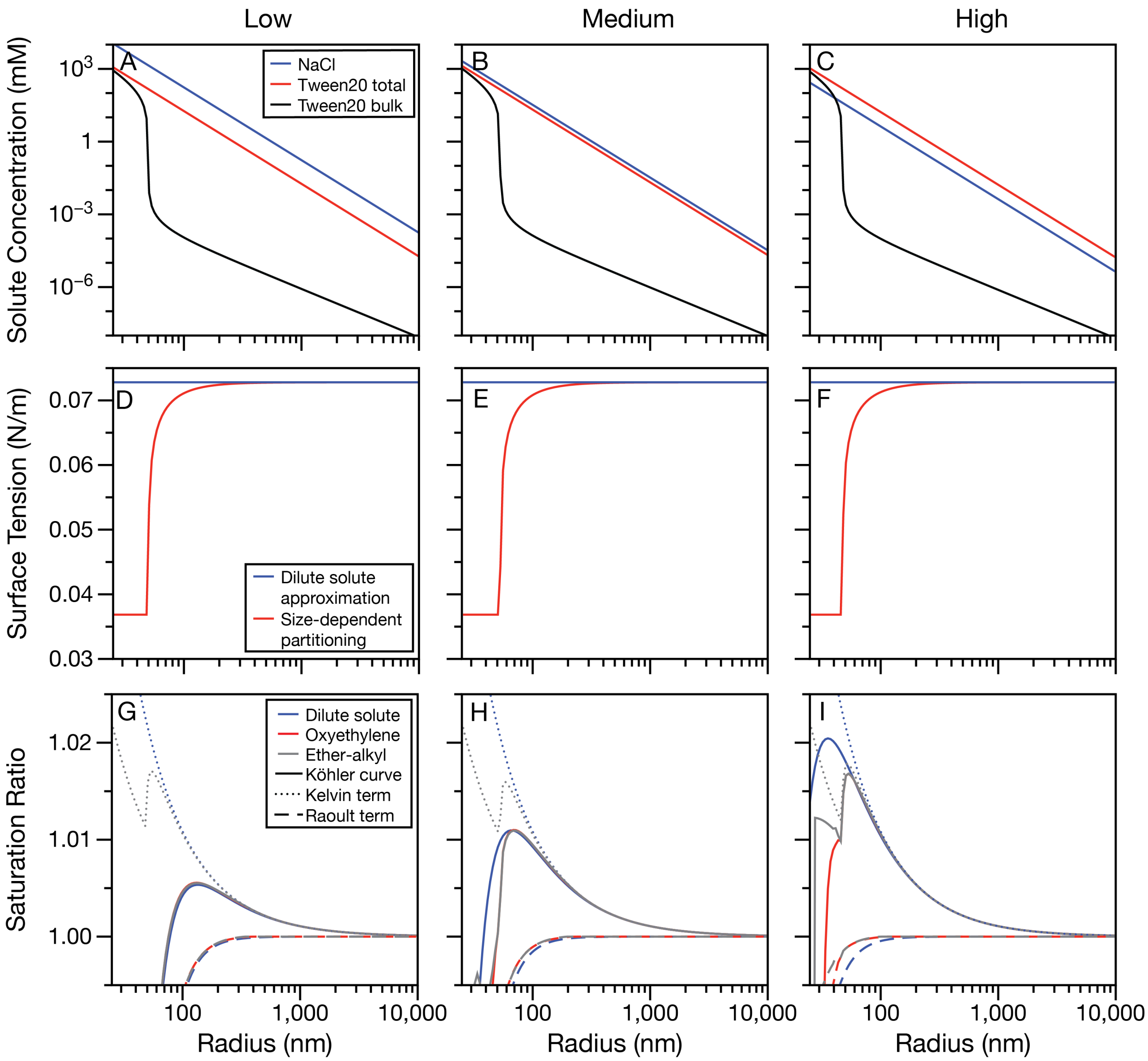

Figure S4: Solute concentration (A-C), surface tension (D-E), and saturation ratio (G-I) for 25 nm radius dry particles containing low (column 1, $w_{org} = 0.689$), medium (columns 2, $w_{org} = 0.930$), and high (column 3, $w_{org} = 0.988$) Tween20 mass fractions. Köhler curves are calculated using the dilute solute approximation as well as using the AIOMFAC activity interpolations using the oxyethylene and ether-alkyl subgroup descriptions.

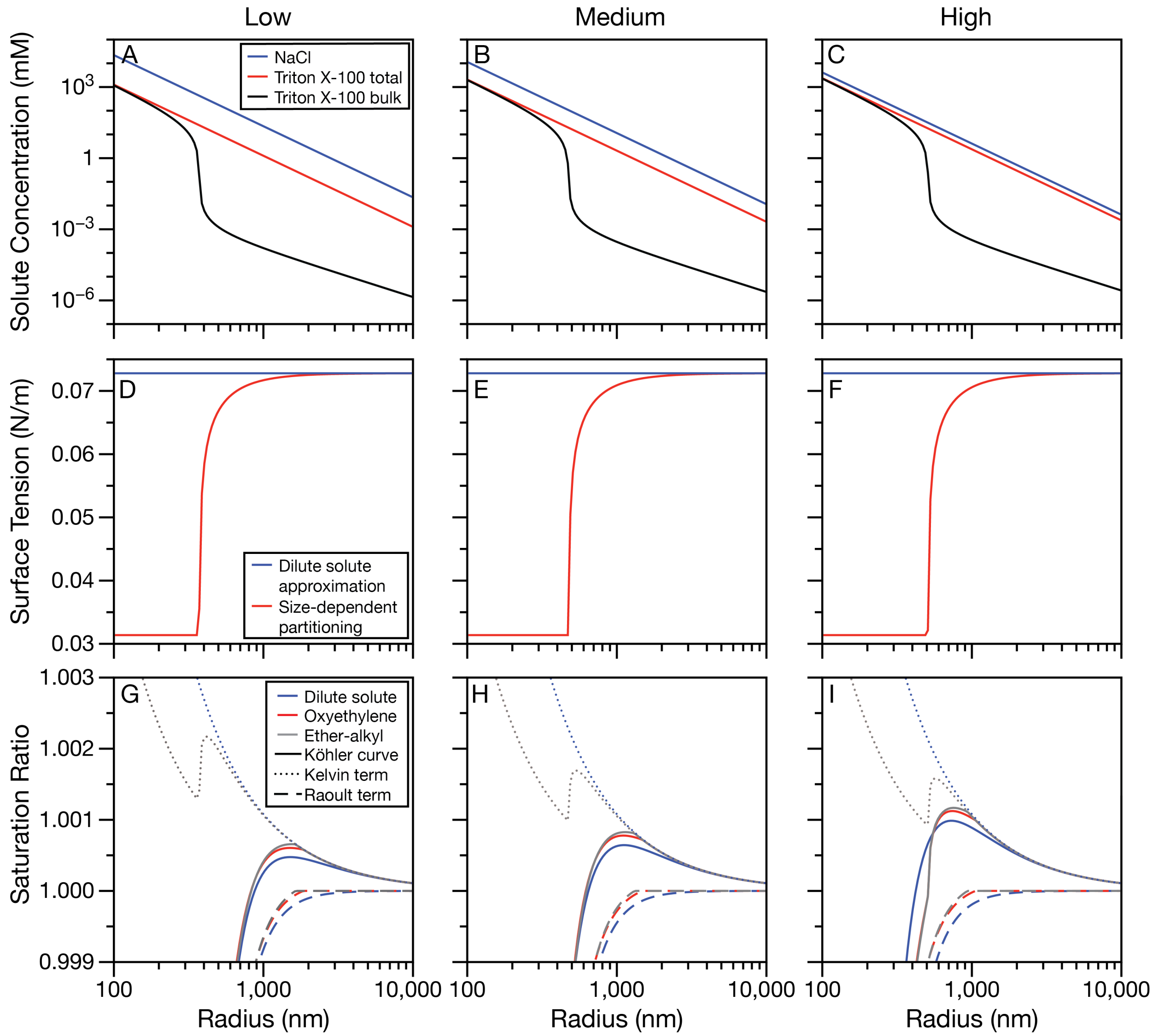

Figure S5: Solute concentration (A-C), surface tension (D-E), and saturation ratio (G-I) for 100 nm radius dry particles containing low (column 1, $w_{org} = 0.386$), medium (columns 2, $w_{org} = 0.667$), and high (column 3, $w_{org} = 0.864$) Triton X-100 mass fractions. Köhler curves are calculated using the dilute solute approximation as well as using the AIOMFAC activity interpolations using the oxyethylene and ether-alkyl subgroup descriptions.

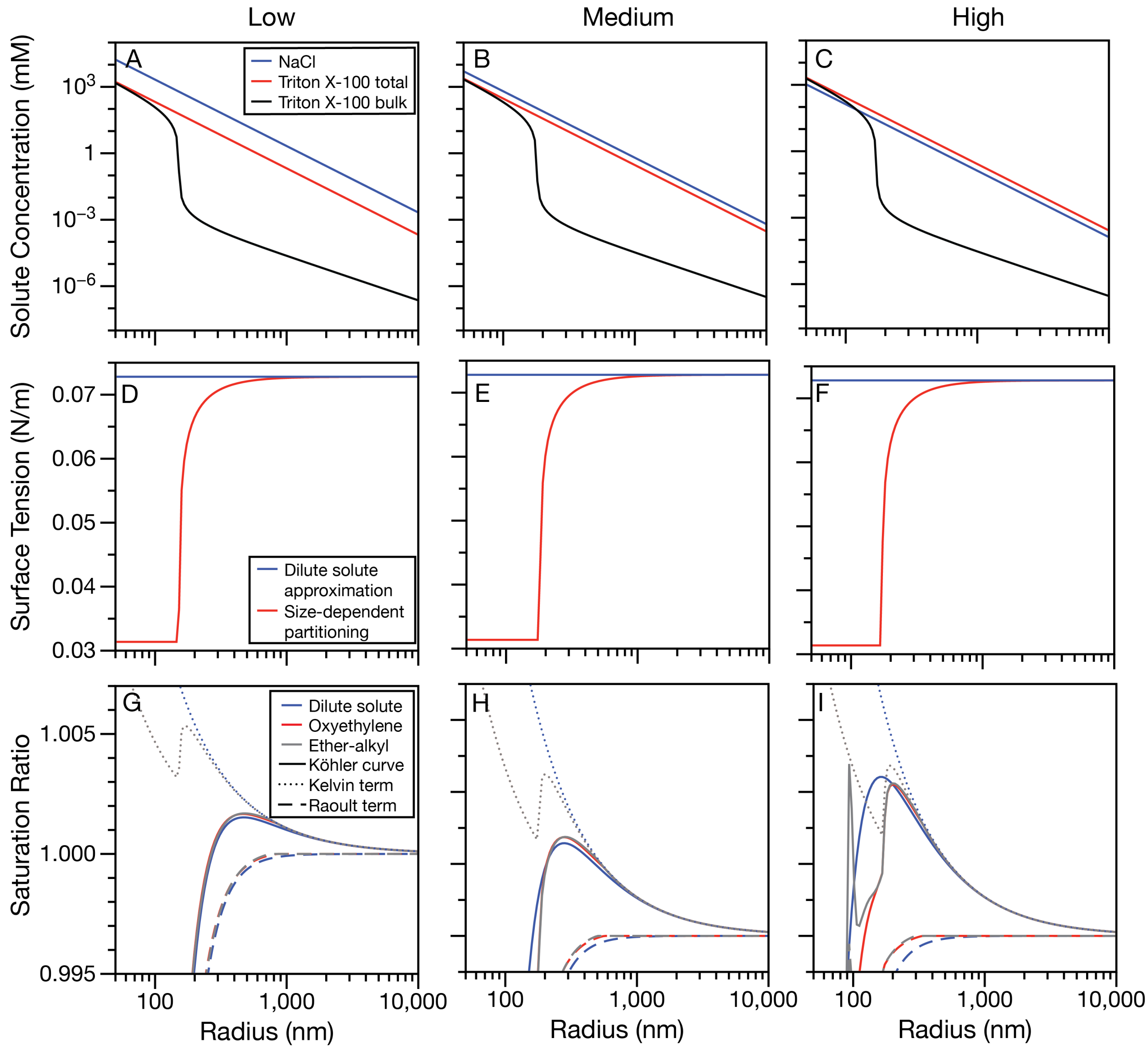

Figure S6: Solute concentration (A-C), surface tension (D-E), and saturation ratio (G-I) for 50 nm radius dry particles containing low (column 1, $w_{org} = 0.522$), medium (columns 2, $w_{org} = 0.838$), and high (column 3, $w_{org} = 0.660$) Triton X-100 mass fractions. Köhler curves are calculated using the dilute solute approximation as well as using the AIOMFAC activity interpolations using the oxyethylene and ether-alkyl subgroup descriptions.

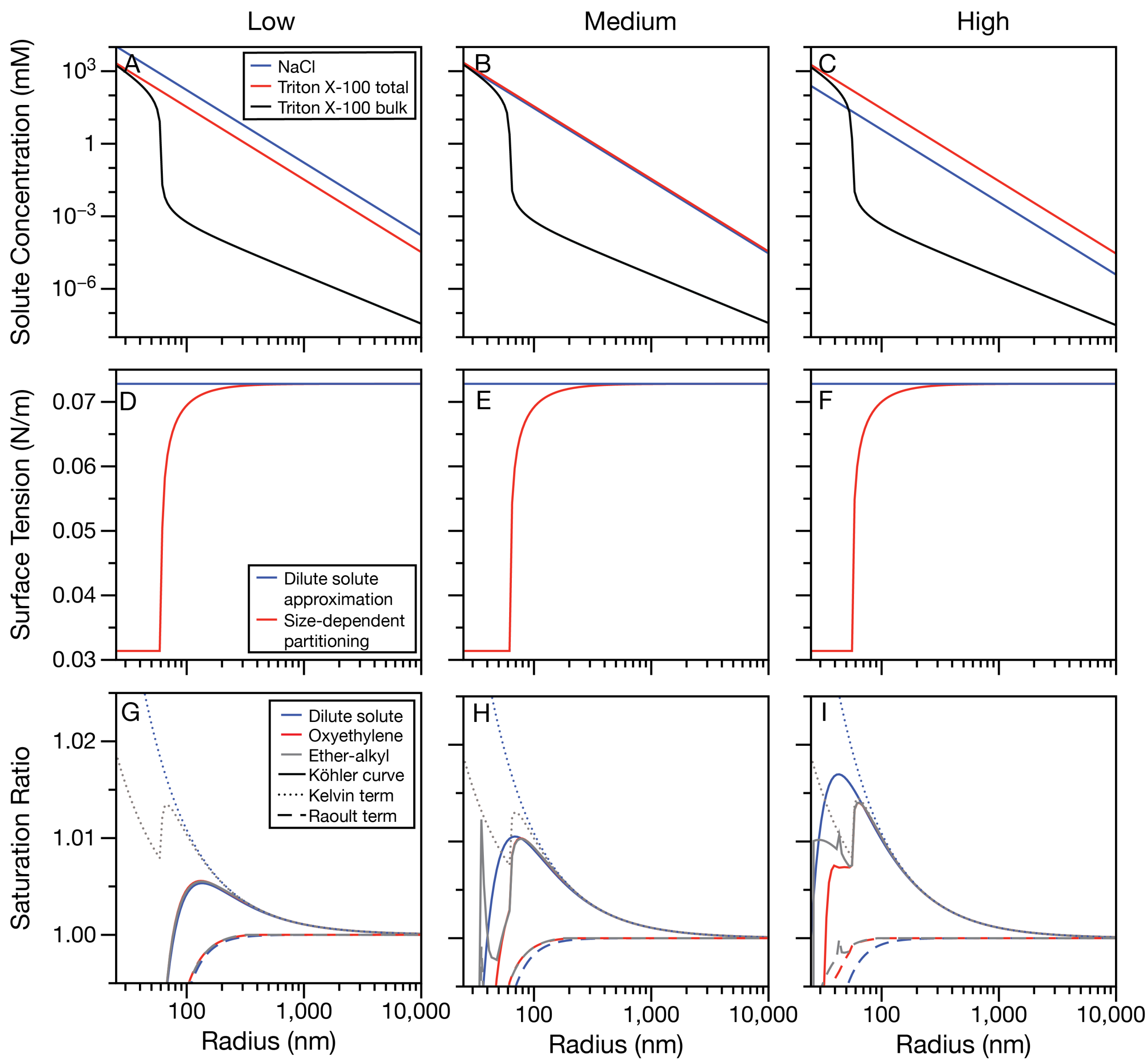

Figure S7: Solute concentration (A-C), surface tension (D-E), and saturation ratio (G-I) for 25 nm radius dry particles containing low (column 1, $w_{org} = 0.689$), medium (columns 2, $w_{org} = 0.930$), and high (column 3, $w_{org} = 0.988$) Triton X-100 mass fractions. Köhler curves are calculated using the dilute solute approximation as well as using the AIOMFAC activity interpolations using the oxyethylene and ether-alkyl subgroup descriptions.

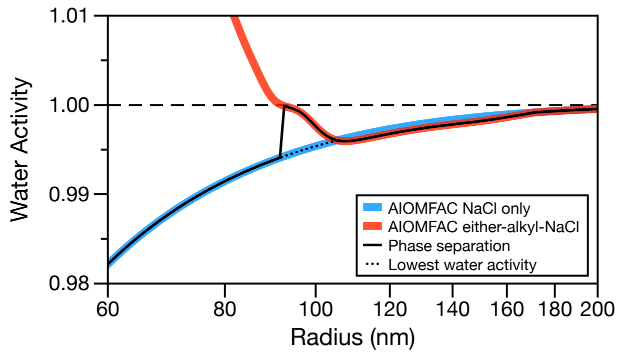

Figure S8: Water activity description during hygroscopic growth using the ether-alkyl subgroup description in AIOMFAC in the region of a discontinuity due to a_w predictions greater than unity. Phase separation: If the water activity for the surfactant-NaCl mixture (red line) is predicted to be greater than unity (dashed black line), then the water activity from the NaCl concentration only (blue line) is used. The solid black line shows the water activity using this approach. Lowest water activity: The lower of either the either-alkyl-NaCl or NaCl-only water activity interpolations is used. The dotted black line shows the water activity using this approach.

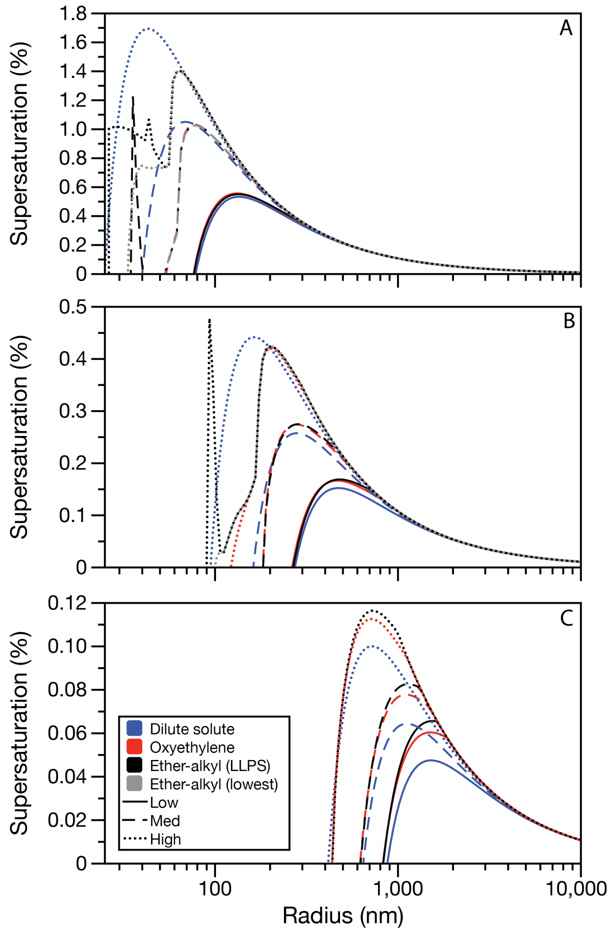

Figure S9: Köhler curves for particles with dry radius A) 25 nm, B) 50 nm, and C) 100 nm and low, medium, and high Triton X-100 mass fractions (Table 1). The water activity for the ether-alkyl subgroup description is shown using both the LLPS assumption and the lowest water activity assumption, where the calculated water activity goes above unity at any point during activation (25 nm medium and high, as well as 50 nm high).

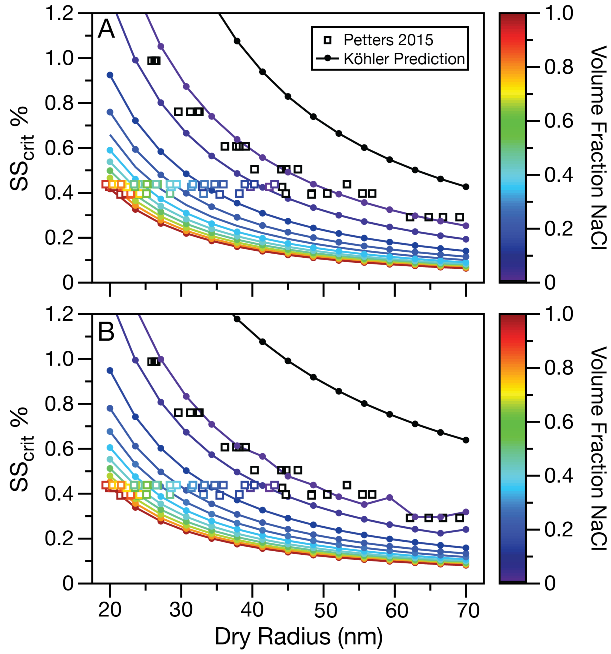

Figure S10: Critical supersaturation (SS_{crit}) predicted using Köhler theory with size- and composition-dependent surfactant partitioning using A) without surfactant partitioning and B) the ether subgroup (liquid-liquid phase separation) to determine droplet water activity. CCN measurements from Petters 2015 are overlaid.[3]

Table S1: Critical supersaturation (SS^*) and critical radii (r^*) for 25, 50 and 100 nm dry radius particles containing low, meduium, and high surfactant mass fractions.

	low		medium		high	
Tween20						
	SS_{crit} (%)	r_{crit} (nm)	SS_{crit} (%)	r_{crit} (nm)	SS_{crit} (%)	r_{crit} (nm)
25 nm						
dilute	0.53416	131.68	1.09575	65.0721	2.04448	35.5633
oxyethylene	0.55602	131.68	1.09988	68.4323	1.67979	53.2023
ether	0.55183	131.68	1.09329	68.4323	1.68071	53.2023
50 nm						
dilute	0.15234	463.222	0.26014	271.489	0.47637	152.188
oxyethylene	0.1651	463.222	0.27591	271.489	0.46702	173.936
ether (LLPS)	0.16746	484.313	0.27645	283.85	0.55420	89.1955
ether (lowest)					0.46825	173.937
100 nm						
dilute	0.04754	1501.31	0.06448	1101.58	0.1001	719.686
oxyethylene	0.0588	1501.31	0.07617	1101.58	0.11263	719.686
ether	0.06351	1560.55	0.08052	1145.05	0.11644	719.686
Triton X-100						
25 nm						
dilute	0.53321	138.48	1.05109	68.4323	1.69429	43.4977
oxyethylene	0.5573	131.68	1.03168	75.6821	1.39723	65.0721
ether (LLPS)	0.55091	131.68	1.22762	35.5633	1.40069	65.0721
ether (lowest)			1.02560	79.5901	1.40069	65.0721
50 nm						
dilute	0.15229	463.222	0.25758	283.85	0.44174	166.361
oxyethylene	0.1666	463.222	0.27417	283.85	0.42087	198.792
ether (LLPS)	0.16893	484.313	0.27502	283.85	0.47775	93.2566
ether (lowest)					0.42087	198.792
100 nm						
dilute	0.04754	1501.31	0.06437	1101.58	0.09865	719.686
oxyethylene	0.06042	1501.31	0.0778	1101.58	0.11231	748.083
ether	0.06568	1560.55	0.08265	1145.05	0.11679	748.083

References

(1) Werner, E. K.; Hammond, M.; Bain, A. Surface tension predictions during hygroscopic growth and cloud droplet activation using a simple kinetic surfactant partitioning model. *Aerosol Sci. Technol.* **2025**, *59*, 781–793.

(2) Bain, A.; Ghosh, K.; Prisle, N. L.; Bzdek, B. R. Surface-Area-to-Volume Ratio Determines Surface Tensions in Microscopic, Surfactant-Containing Droplets. *ACS Cent. Sci.* **2023**, *9*, 2076–2083.

(3) Petters, S. S. On the Physicochemical Processes Controlling Organic Aerosol Hygroscopicity. 2015; PhD thesis.