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Impact of acidity and surface-modulated acid dissociation on cloud response to organic aerosol

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S1 Dissociation degree and van't Hoff factor

Table S1. The organic aerosol van't Hoff factor (i_{OA}) and dissociation degree (α, in parenthesis) for decanoic acid and malonic acid, respectively, corresponding to bulk acid dissociation ($\mathrm{p}K_a^{\mathrm{bulk}}$) and surface modulated suppressed organic acid dissociation ($\mathrm{p}K_a^{\mathrm{bulk}}+1$ and $\mathrm{p}K_a^{\mathrm{bulk}}+2$) considering initial organic mass fractions $\chi_{\mathrm{OA}} = \{0.2, 0.4, 0.6, 0.8, 1\}$.

Malonic acid				
χ_{OA}	i_{OA} (α), $\mathrm{p}K_a^{\mathrm{bulk}}$	i_{OA} (α), $\mathrm{p}K_a^{\mathrm{bulk}}+1$	i_{OA} (α), $\mathrm{p}K_a^{\mathrm{bulk}}+2$	i_{OA} (α), no diss
0.2	2.12 (0.560)	1.90 (0.450)	1.70 (0.350)	1 (0)
0.4	2.064 (0.532)	1.85 (0.425)	1.66 (0.331)	1 (0)
0.6	2.02 (0.510)	1.81 (0.405)	1.63 (0.315)	1 (0)
0.8	1.996 (0.498)	1.770 (0.385)	1.590 (0.295)	1 (0)
1	1.960 (0.480)	1.720 (0.360)	1.560 (0.280)	1 (0)
Decanoic acid				
χ_{OA}	i_{OA} (α), $\mathrm{p}K_a^{\mathrm{bulk}}$	i_{OA} (α), $\mathrm{p}K_a^{\mathrm{bulk}}+1$	i_{OA} (α), $\mathrm{p}K_a^{\mathrm{bulk}}+2$	i_{OA} (α), no diss
0.2	1.993 (0.993)	1.905 (0.905)	1.720 (0.720)	1 (0)
0.4	1.989 (0.989)	1.890 (0.890)	1.715 (0.715)	1 (0)
0.6	1.985 (0.985)	1.875 (0.875)	1.705 (0.705)	1 (0)
0.8	1.981 (0.981)	1.845 (0.845)	1.680 (0.680)	1 (0)
1	1.978 (0.978)	1.823 (0.823)	1.650 (0.650)	1 (0)

S2 Hydrogen ion concentration in the aerosol with varying ionic strengths

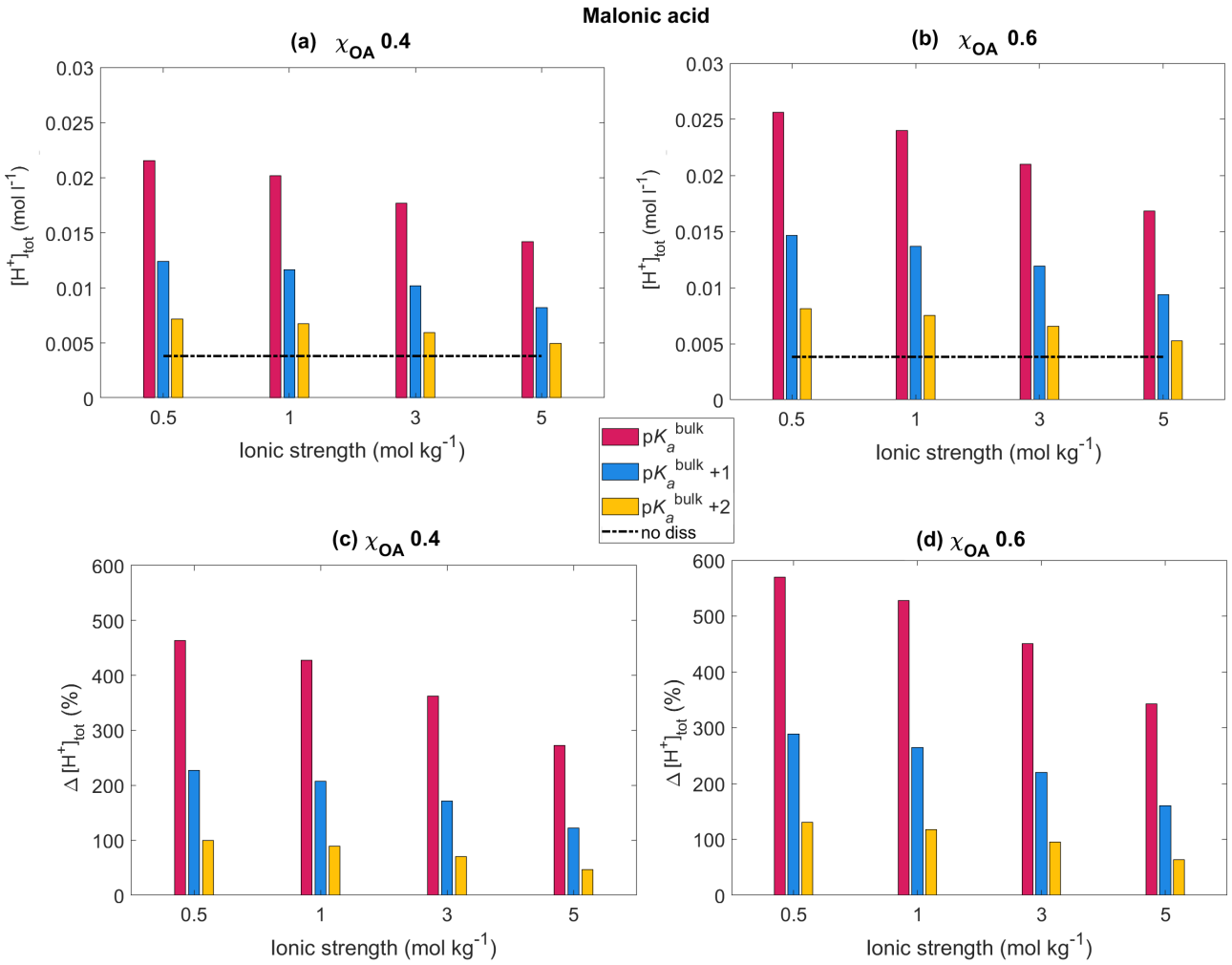

Figure S1. Total hydrogen ion concentration, $[\mathrm{H}^+]_{\mathrm{tot}}$, and the relative change in hydrogen ion concentration with respect to the 'no diss' ($\Delta[\mathrm{H}^+]_{\mathrm{tot}}$) for varying ionic strengths (I), in the aqueous aerosol population with sizes between $D_{\mathrm{wet}} = 0.317 - 40$ µm, after 1 hour of simulation time, considering the entire OA fraction as malonic acid and initial OA mass fractions, $\chi_{\mathrm{OA}} = 0.4$ (a,c) and 0.6 (b,d), with varying pK_a corresponding to representations of bulk (pK_a^{bulk} pink,) and surface modulated (p$K_a^{\mathrm{bulk}} + 1$, blue, and p$K_a^{\mathrm{bulk}} + 2$, yellow) organic acid dissociation.

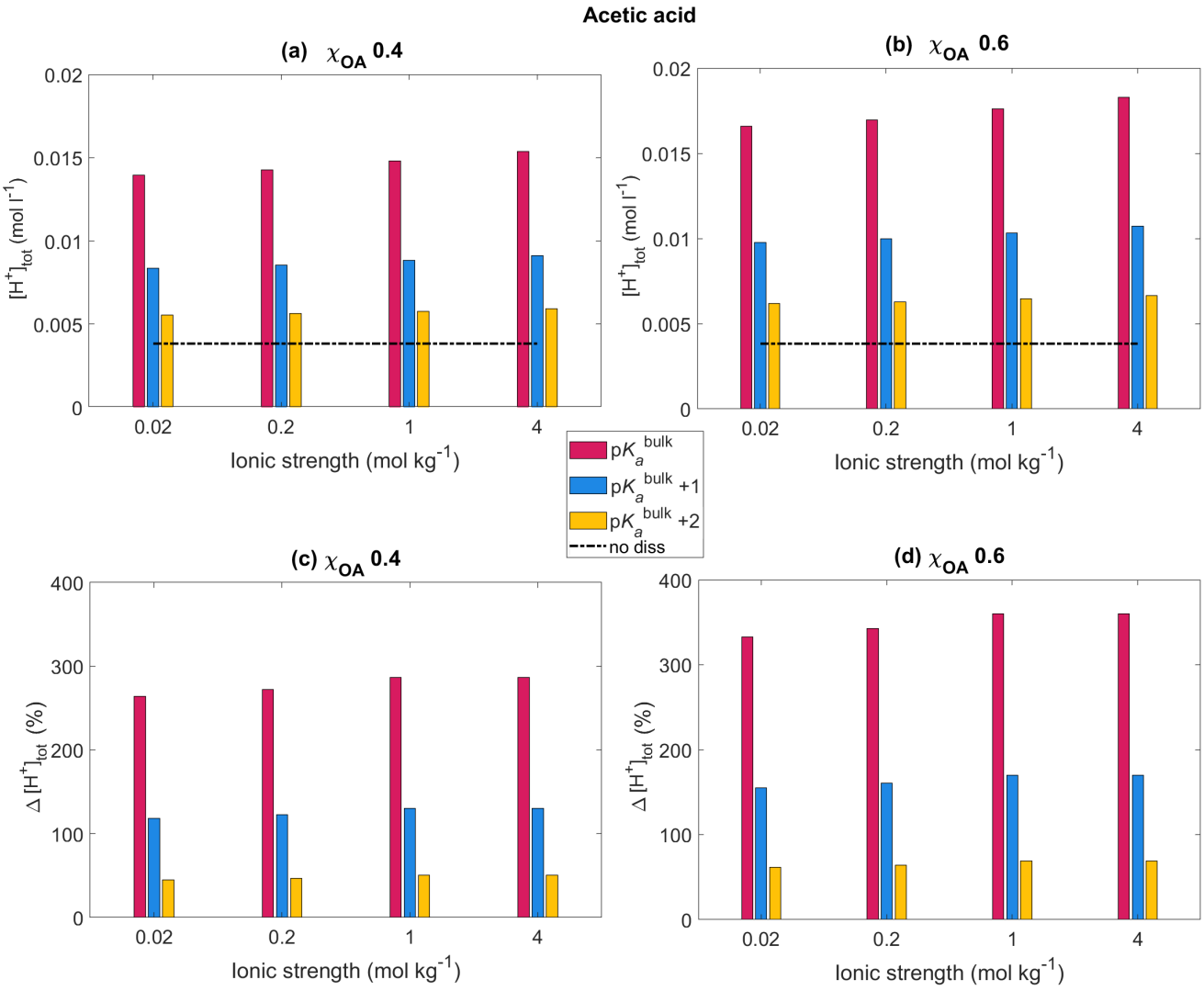

Figure S2. Total hydrogen ion concentration, $[\mathrm{H}^+]_{\mathrm{tot}}$, and the relative change in hydrogen ion concentration with respect to the 'no diss' ($\Delta[\mathrm{H}^+]_{\mathrm{tot}}$) for varying ionic strengths (I), in the aqueous aerosol population with sizes between $D_{\mathrm{wet}} = 0.317 - 40$ µm, after 1 hour of simulation time, assuming the entire OA fraction as acetic acid and initial OA mass fractions, $\chi_{\mathrm{OA}} = 0.4$ (a,c) and 0.6 (b,d), with varying pK_a corresponding to representations of bulk (pK_a^{bulk} pink,) and surface modulated (p$K_a^{\mathrm{bulk}} + 1$, blue, and p$K_a^{\mathrm{bulk}} + 2$, yellow) organic acid dissociation.

S3 Change in sulfate concentrations, $\Delta[\mathrm{SO}_4^{2-}]''$, with respect to 'no diss'

The relative change in sulfate concentration compared to 'no diss' is given by

$$\Delta \left[\mathrm{SO}_4^{2-}\right]'' = \frac{\left[\mathrm{SO}_4^{2-}\right]'' - \left[\mathrm{SO}_4^{2-}\right]''_0}{\left[\mathrm{SO}_4^{2-}\right]''_0} \times 100, \tag{S1}$$

where $\left[\mathrm{SO}_4^{2-}\right]''$ is the total sulfate concentration considering organic acid dissociation and $\left[\mathrm{SO}_4^{2-}\right]''_0$ is the total sulfate concentration for 'no diss'.

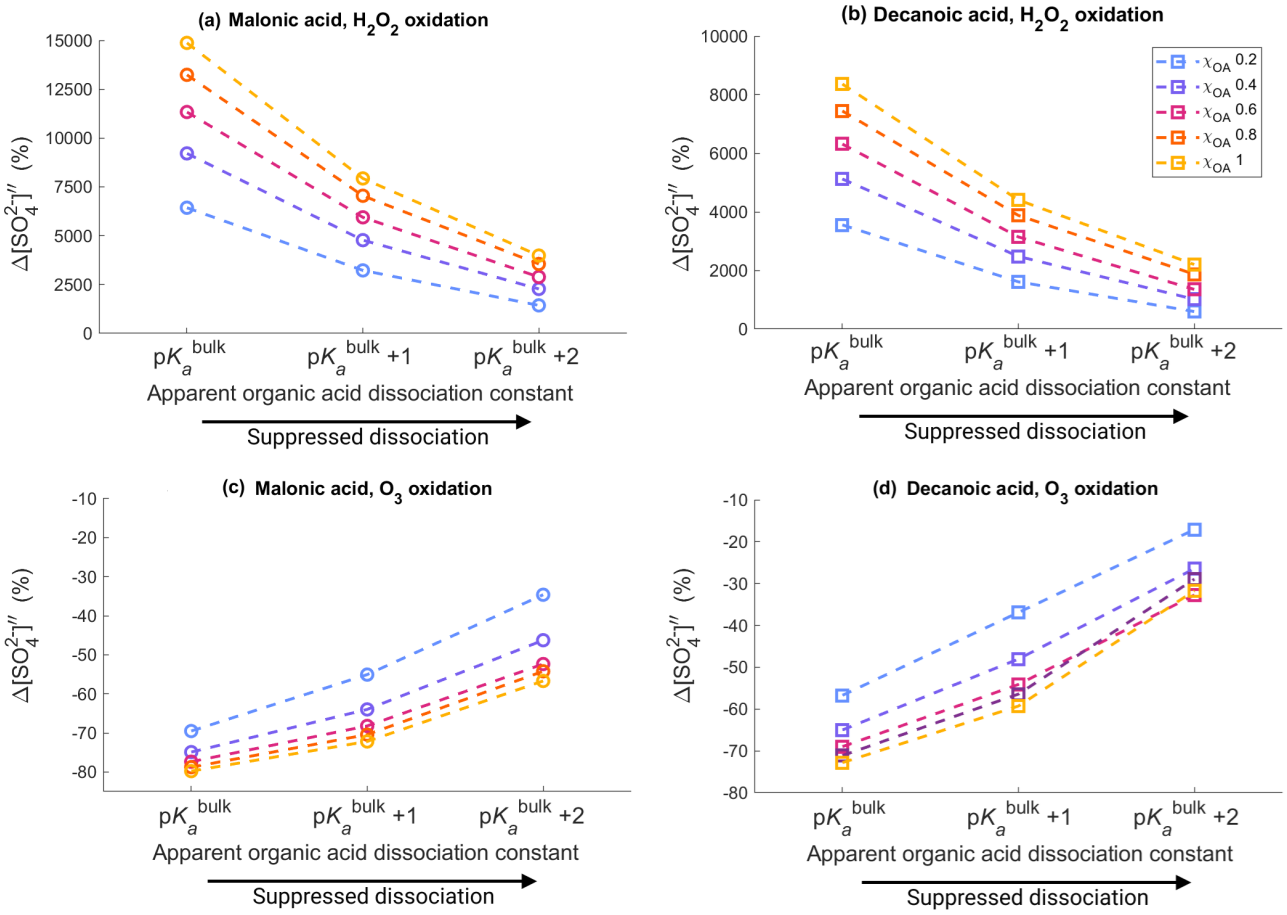

Figure S3. The relative change in secondary sulfate concentrations ($\Delta[SO_4^{2-}]''$) from oxidation of SO_2 by (a, b) H_2O_2, and (c, d) O_3, with respect to the 'no diss', in the aqueous aerosol population with sizes between $D_{wet} = 0.317 - 40$ µm, after 1 hour of simulation time, assuming the entire OA fraction as (a, c) malonic acid, and (b, d) decanoic acid, with varying pK_a corresponding to representations of bulk (pK_a^{bulk}) and surface modulated (p$K_a^{bulk} + 1$ and p$K_a^{bulk} + 2$) organic acid dissociation, for the five different initial organic mass fractions $\chi_{OA} = \{0.2, 0.4, 0.6, 0.8, 1\}$ considered, denoted by blue, purple, pink, orange, and yellow, respectively.

S4 Particle size distribution with and without sulfur chemistry module

S4.1 Malonic acid

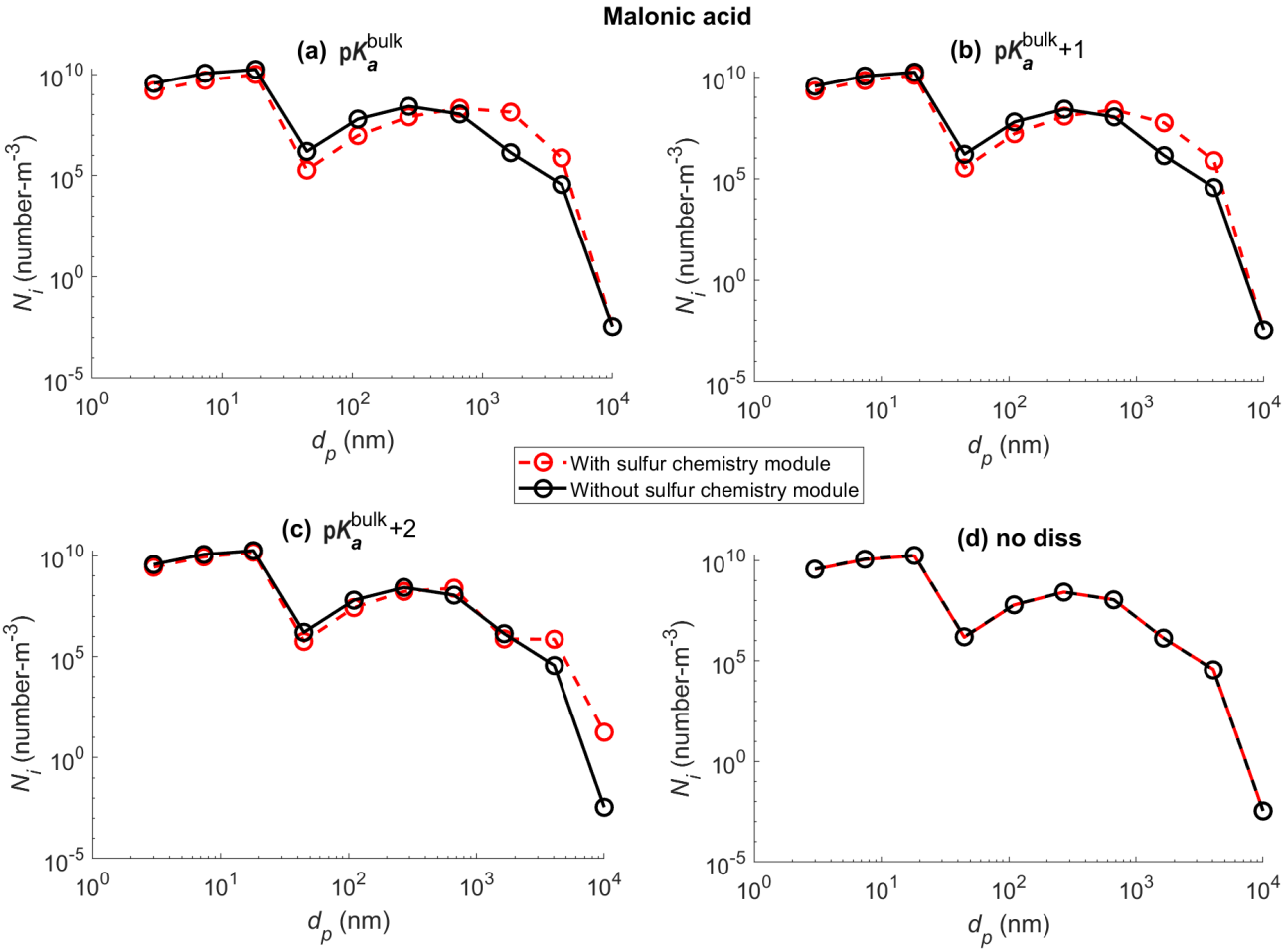

Figure S4. Aerosol size distribution represented by the number concentration (N_i) as a function of particle diameter (d_p) after 1 hour simulation time with (red) and without (black) including the sulfur chemistry module considering the entire OA fraction as malonic acid with $\chi_{\mathrm{OA}} = 0.8$, assuming organic acid dissociation according to (a) $\mathrm{p}K_a^{\mathrm{bulk}}$, (b) $\mathrm{p}K_a^{\mathrm{bulk}} + 1$, (c) $\mathrm{p}K_a^{\mathrm{bulk}} + 2$, and (d) no diss.

S4.2 Decanoic acid

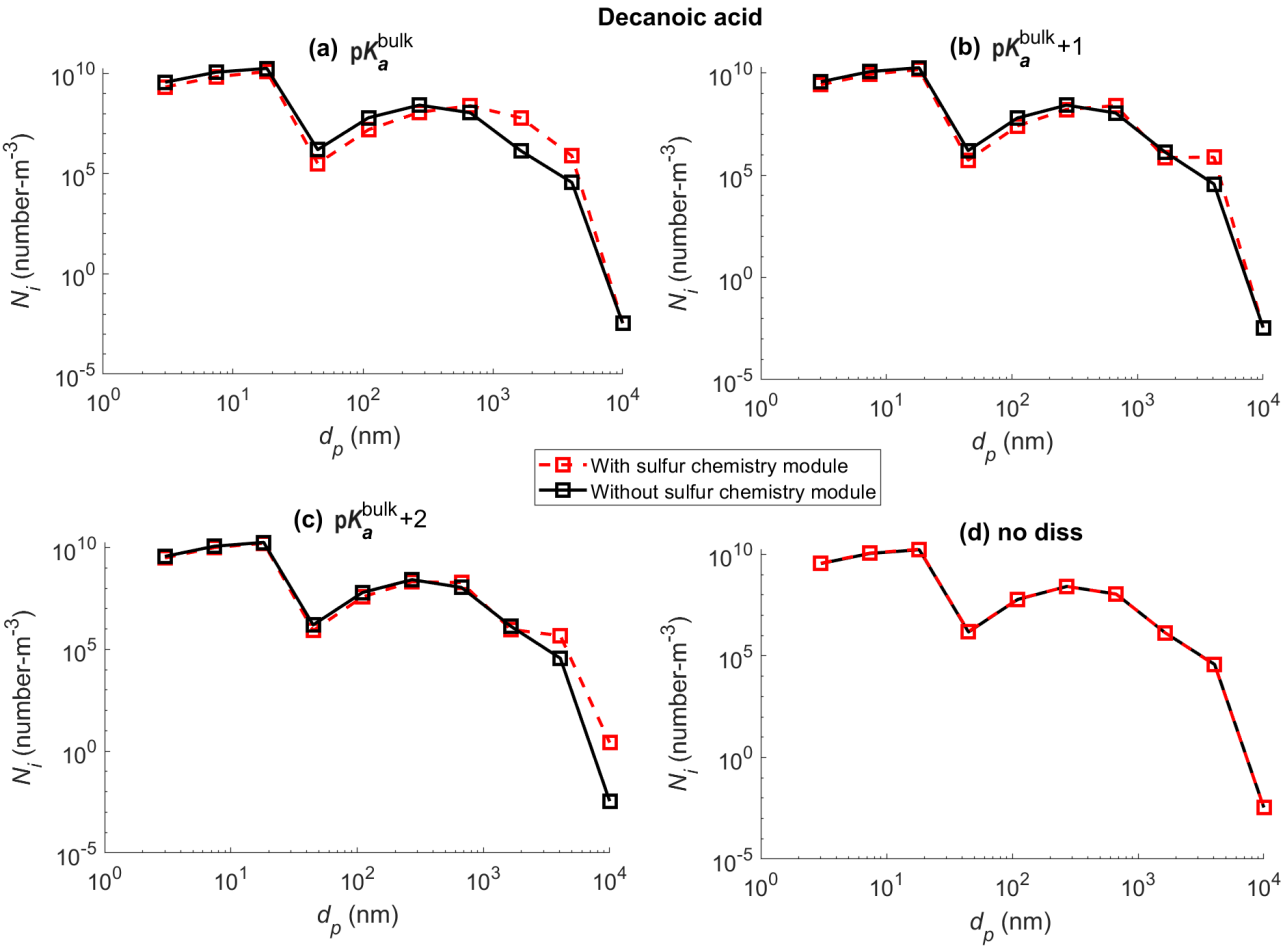

Figure S5. Particle size distribution represented by the number concentration (N_i) as a function of particle diameter (d_p) after 1 hour simulation time with (red) and without (black) including the sulfur chemistry module considering the entire OA fraction as decanoic acid with $\chi_{\mathrm{OA}} = 0.8$, assuming organic acid dissociation according to (a) pK_a^{bulk}, (b) p$K_a^{\mathrm{bulk}} + 1$, (c) p$K_a^{\mathrm{bulk}} + 2$, and (d) no diss.

S5 Effect on cloud properties from changes in water activity only

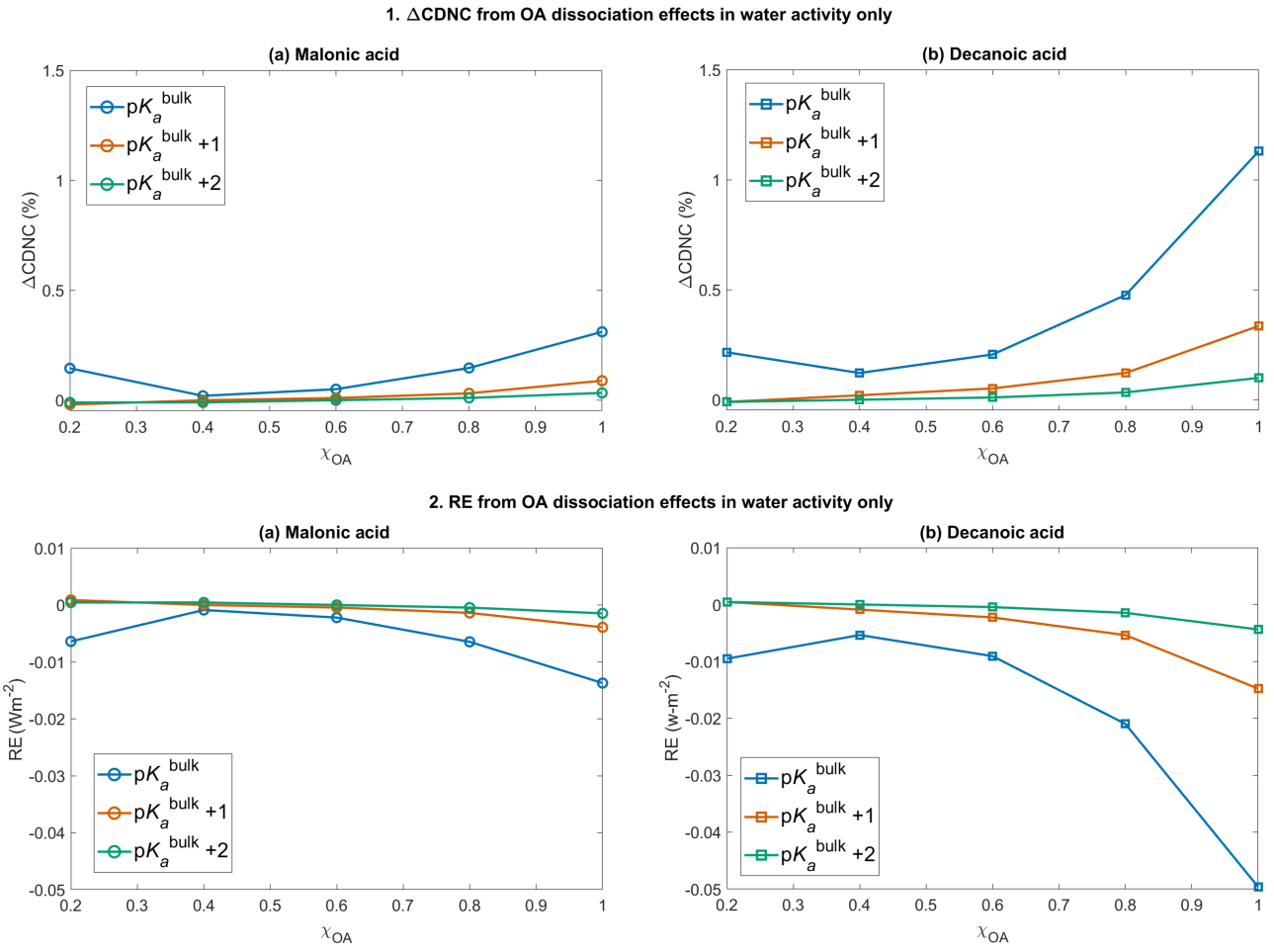

Figure S6. Organic acid dissociation effects in 1) cloud droplet number concentration (ΔCDNC) and 2) the resulting short-wave radiative effect (RE) with respect to 'no diss', for varying initial organic mass fractions (χ_{OA}) assuming bulk organic acidity ($\mathrm{p}K_a^{\mathrm{bulk}}$, blue) and surface modulated organic acid dissociation ($\mathrm{p}K_a^{\mathrm{bulk}}+1$, orange and $\mathrm{p}K_a^{\mathrm{bulk}}+2$, green), from changes in van't Hoff factor and water activity only. The sulfur chemistry module has not been activated in these simulations.