

**Supplementary Information:**

**Ozone oxidation of oleic acid surface films decreases aerosol CCN activity**

A. N. Schwier,<sup>1</sup> N. Sareen,<sup>1</sup> T. L. Lathem,<sup>2</sup> A. Nenes,<sup>2,3,\*</sup> and V. F. McNeill<sup>1,\*</sup>

1. Department of Chemical Engineering, Columbia University

2. School of Earth and Atmospheric Sciences, Georgia Institute of Technology

3. School of Chemical and Biomolecular Engineering, Georgia Institute of Technology

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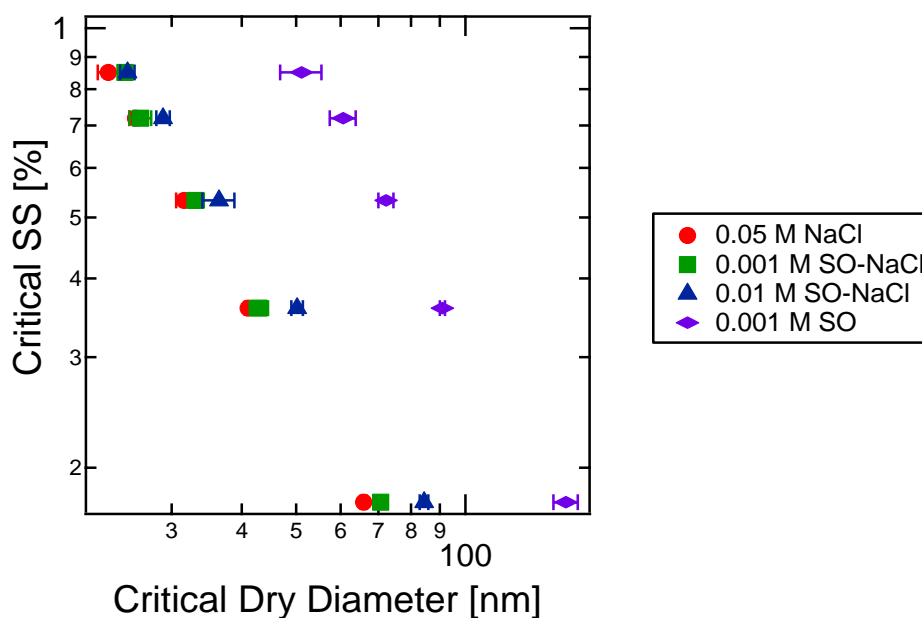


Figure S1. CCN activity of 0.05 M NaCl with increasing concentrations of SO towards pure 0.001 M SO.

**Table 1.** Power Log Fits and  $\kappa$  parameters of SO/NaCl/Na<sub>2</sub>SO<sub>4</sub>/H<sub>2</sub>SO<sub>4</sub> oxidation experiments

	Power Log Fit	$\kappa$ (avg)
0.001 M SO, Water, (fit to NaCl SS%)	-1.491	0.118 ± 0.004
0.001 M SO, Water, (fit to Na <sub>2</sub> SO <sub>4</sub> SS%)	-1.436	0.119 ± 0.003
NaCl	-1.503	1.378 ± 0.025
NaCl, 1 ppm O <sub>3</sub>	-1.534	1.094 ± 0.045
0.001 M SO, NaCl	-1.474	1.187 ± 0.034
0.001 M SO, NaCl, 0.2 ppm O <sub>3</sub>	-1.505	1.113 ± 0.026
0.001 M SO, NaCl, 1 ppm O <sub>3</sub>	-1.464	1.170 ± 0.027
0.01 M SO, NaCl	-1.296	0.869 ± 0.062
0.01 M SO, NaCl, 0.2 ppm O <sub>3</sub>	-1.368	0.753 ± 0.054
0.01 M SO, NaCl, 1 ppm O <sub>3</sub>	-1.360	0.906 ± 0.041
0.001 M SO, NaCl, H <sub>2</sub> SO <sub>4</sub>	-1.538	1.138 ± 0.051
0.001 M SO, NaCl, H <sub>2</sub> SO <sub>4</sub> , 1 ppm O <sub>3</sub>	-1.473	1.065 ± 0.054
0.01 M SO, NaCl, H <sub>2</sub> SO <sub>4</sub>	-1.271	0.971 ± 0.079
0.01 M SO, NaCl, H <sub>2</sub> SO <sub>4</sub> , 1 ppm O <sub>3</sub>	-1.449	0.786 ± 0.024
Na <sub>2</sub> SO <sub>4</sub>	-1.503	0.872 ± 0.016
Na <sub>2</sub> SO <sub>4</sub> , 1 ppm O <sub>3</sub>	-1.348	0.909 ± 0.044
0.001 M SO, Na <sub>2</sub> SO <sub>4</sub>	-1.475	0.708 ± 0.037
0.001 M SO, Na <sub>2</sub> SO <sub>4</sub> , 1 ppm O <sub>3</sub>	-1.460	0.708 ± 0.024
0.01 M SO, Na <sub>2</sub> SO <sub>4</sub>	-1.486	0.682 ± 0.025
0.01 M SO, Na <sub>2</sub> SO <sub>4</sub> , 1 ppm O <sub>3</sub>	-1.360	0.670 ± 0.032
0.001 M SO, Na <sub>2</sub> SO <sub>4</sub> , H <sub>2</sub> SO <sub>4</sub>	-1.459	0.748 ± 0.011
0.001 M SO, Na <sub>2</sub> SO <sub>4</sub> , H <sub>2</sub> SO <sub>4</sub> , 1 ppm O <sub>3</sub>	-1.468	0.709 ± 0.026
0.01 M SO, Na <sub>2</sub> SO <sub>4</sub> , H <sub>2</sub> SO <sub>4</sub>	-1.370	0.615 ± 0.029
0.01 M SO, Na <sub>2</sub> SO <sub>4</sub> , H <sub>2</sub> SO <sub>4</sub> , 1 ppm O <sub>3</sub>	-1.374	0.548 ± 0.024

### **Calculation for acidification of aerosols with sulfuric acid**

The pH of aerosols flowed over a reservoir of sulfuric acid was calculated using the following method:

The amount of H<sub>2</sub>SO<sub>4</sub> in the gaseous phase was determined by solving  $[H_2SO_4]_{f,gas} = [H_2SO_4]_{i,gas} e^{-k^I t}$

where  $k^I = \frac{\gamma \omega S_a}{4}$  and  $\gamma$  is the H<sub>2</sub>SO<sub>4</sub> reaction probability,  $\omega$  is the molecular velocity of H<sub>2</sub>SO<sub>4</sub>, and  $S_a$  is

the aerosol surface area per volume. Using the vapor pressure of H<sub>2</sub>SO<sub>4</sub> at 25°C and both 93 and 98% H<sub>2</sub>SO<sub>4</sub> (based on the purity of the laboratory H<sub>2</sub>SO<sub>4</sub> used), the residence time that the aerosols spent in the H<sub>2</sub>SO<sub>4</sub> reservoir (0.8 sec), and an arbitrary  $\gamma=0.5$ ,  $[H_2SO_4]_{f,gas}$  was determined.  $[H_2SO_4]_{f,gas}$  was converted

from bar to mol cm<sup>-3</sup> using the ideal gas law, or  $c = \frac{P}{RT}$ . The amount of H<sub>2</sub>SO<sub>4</sub> within the particle was found by  $\frac{c}{N_p V_p}$  where  $N_p$  and  $V_p$  are number density and volume of the particles (using the volume weighted geometric mean diameter) determined by the SMPS. Assuming an initial pH = 8, and determining the number of moles of [H<sup>+</sup>], the final pH of the aerosols was determined.

### **Calculation for natural acidification of aerosols by formation of organic acids**

The natural pH change of the aerosols that could occur through the formation of nonanoic acid, 9-oxononanoic acid and azelaic acid was calculated in the following method:

The  $pK_a$  value for each acid was either known or assumed. The  $K_a$  for each acid could be written (shown here for nonanoic acid) as  $K_a = \frac{[C_9H_{17}O_2^-][H_3O^+]}{[C_9H_{18}O_2]}$ , and it was assumed that the concentrations of the dissociated ions were equivalent. Combining the  $K_a$  of all three organic acids,  $[H_3O^+]VA = \sqrt{K_a[C_9H_{18}O_2]V^2A^2} + \sqrt{K_a[C_9H_{16}O_3]V^2A^2} + \sqrt[3]{K_a[C_9H_{16}O_4]V^3A^3}$  where each  $K_a$  describes a unique acid,  $V$  is volume and  $A$  is Avogadro's number. The final pH was calculated from this point.