Supporting information

Responses of Ammonium Sulfate Particles Coated With Glutaric Acid to Cyclic Changes in Relative Humidity: Hygroscopicity and Raman Characterization

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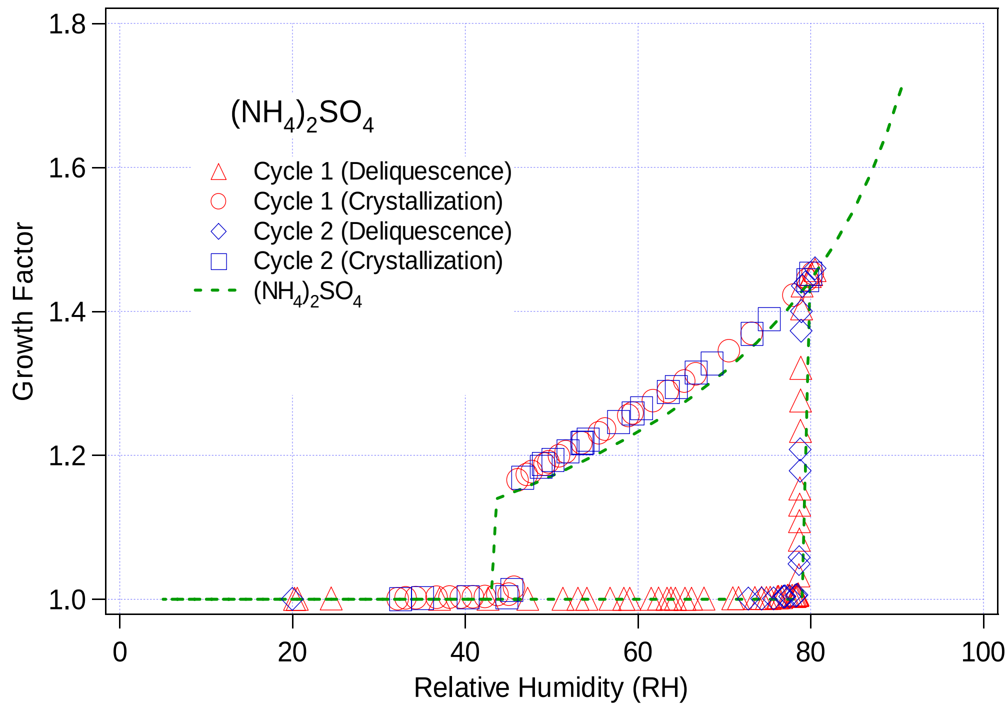
Table S1: The physical properties of ammonium sulfate and glutaric acid.

Chemical	Molecular weight (g/mol)	Density (g/cm^3)	Solubility (g per 100 cm^3 H_2O)	State (25^oC)
Ammonium Sulfate ($(NH_4)_2SO_4$)	132.14	1.769	75.4	Solid
Glutaric Acid ($COOH(CH_3)_3COOH$)	132.11	1.424	116	Solid

Figure S1: The hygroscopicity of ammonium sulfate particles in two deliquescence and crystallization cycles



Experimental Section

Hygroscopic Measurement

Growth Factor Calculation

For comparison with the literature data reported in the form of a growth factor, G_f, our relative mass (mass fraction of solute, *mfs*) measurements are converted to G_f as a function of RH. The G_f is defined as the diameter of the particle, d_p(RH), at a given RH, to the diameter of the particle, $d_p(RH_o)$, at the reference RH_o (RH_o = 20%):

$$G_f(RH) = \frac{d_p(RH)}{d_p(RH_o)} = \left(\frac{mfs(RH_o) \cdot \rho(RH_o)}{mfs(RH) \cdot \rho(RH)} \right)^{1/3} \qquad (1)$$

where ρ is the particle density. The estimated uncertainty in the *mfs* values were about 0.01 and 0.03 for droplets and solid particles, respectively. The densities and volume of solid particles and solution droplets were estimated utilizing the volume additivity rule (1-3). Using the similar approach, this rule for density estimates added about 5-10% uncertainty to the calculation of water absorption (3).

Hygroscopic Growth

In estimating the hygroscopic growth of $(NH_4)_2SO_4$ particles coated with glutaric acid, we assumed that $(NH_4)_2SO_4$ and glutaric acid absorb water independently utilizing the Zdanovskii-Stokes-Robinson (ZSR) equation, which has been found to be useful in estimating the hygroscopic growth of the particles. The G_f of the particles can be estimated as:

$$G_f = (\varepsilon_{organic} G_{f,organic}{}^3 + \varepsilon_{inorganic} G_{f,inorganic}{}^3)^{1/3} \qquad (2)$$

where $\varepsilon_{organic}$ and $\varepsilon_{inorganic}$ are the volume fractions of glutaric acid and $(NH_4)_2SO_4$ of the particles, respectively. G_f, organic and G_f, inorganic are the G_f of glutaric acid and $(NH_4)_2SO_4$ particles at given RH, respectively. The G_f of $(NH_4)_2SO_4$ particles is obtained from Chan et al. (4). The hygroscopicity of glutaric acid particles has been measured by Peng et al. (5) utilizing an EDB and by Prenni et al. (6) utilizing a HTDMA. Prenni et al. (6) reported a G_f of 1.29, which is close to a G_f of 1.30 as reported by Peng et al. (5). The G_f of glutaric acid particles is obtained from Peng et al. (5) because both deliquescence and crystallization data were available and the particle size was similar to those investigated in our study. The estimated G_f of the particles from Eq. 2 is depicted in Figures 1a and 2a (orange dotted line). The estimated G_f are the same for two deliquescence and crystallization cycles because the same aerosol composition was used in the ZSR calculations since the evaporation loss of glutaric acid was not significant.

As shown in the Figures 1a and 2a, the ZSR predictions can generally capture the hygroscopic growth of mixed $(NH_4)_2SO_4$-glutaric acid solution droplets. These results are consistent with the previous studies that the ZSR method can provide a good estimation on the hygroscopic growth of mixed $(NH_4)_2SO_4$ -glutaric acid solution droplets after deliquescence at a single high RH (e.g., 85%RH) (2, 3).

We have also compared our results with other experimental results. The G_f of the particle was 1.42-1.43 at 80%RH in the two cycles, which compares favorably with the G_f of 1.39±0.04 at 80%RH for mixed $(NH_4)_2SO_4$-glutaric acid particles (~9wt% glutaric acid) reported by Prenni et al. (7). $(NH_4)_2SO_4$ particles coated with 49wt% glutaric acid (Figure 2a) had a G_f of 1.34-1.36 at 80%RH, which was comparable with the G_f of 1.30±0.04 for mixed $(NH_4)_2SO_4$-glutaric acid particles (~50wt% glutaric acid) as reported by Prenni et al. (7).

Literature cited

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