



Supplement of

**Identification of micro-dynamics phase transition processes
for ammonium sulfate aerosols by two-dimensional
correlation spectroscopy**

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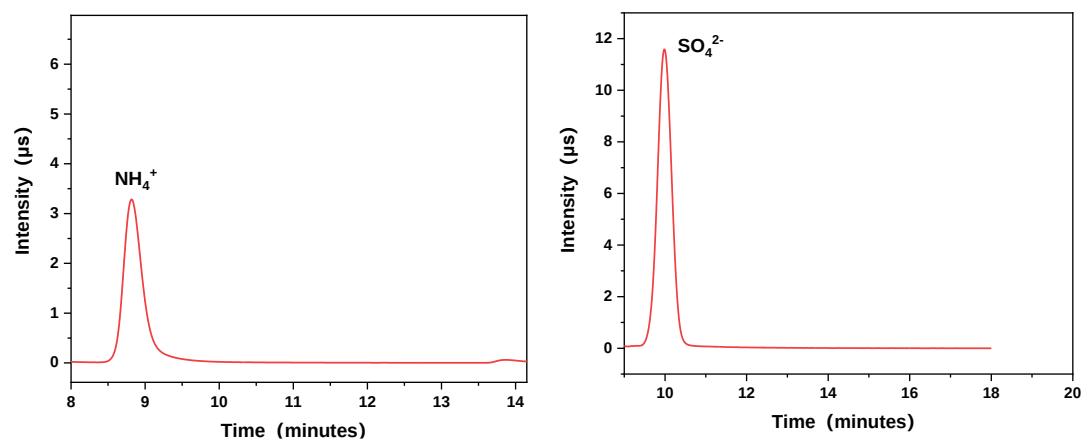


Fig.S1 Ion chromatograms of the deposited AS. Ion chromatograms (ICS-3000, Dionex, United States)

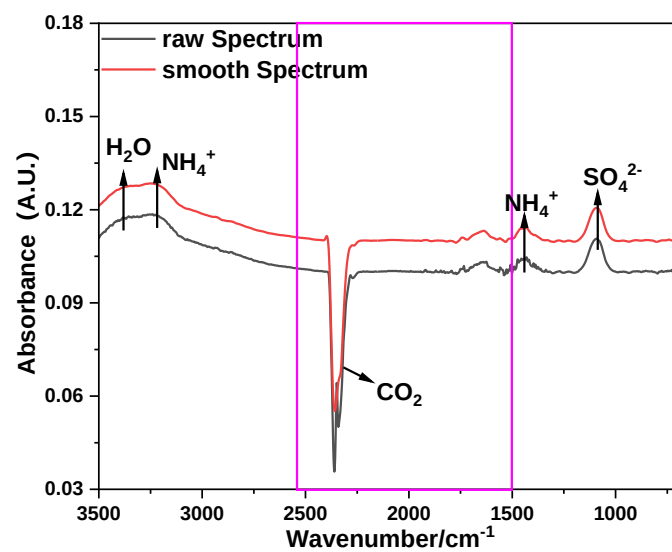


Fig.S2 The complete spectrum for raw and smoothed spectrum at 90% (RH) of Ammonium Sulfate (AS) particles. Magenta rectangle indicates the spectral range that has been omitted from display in Fig. 1.

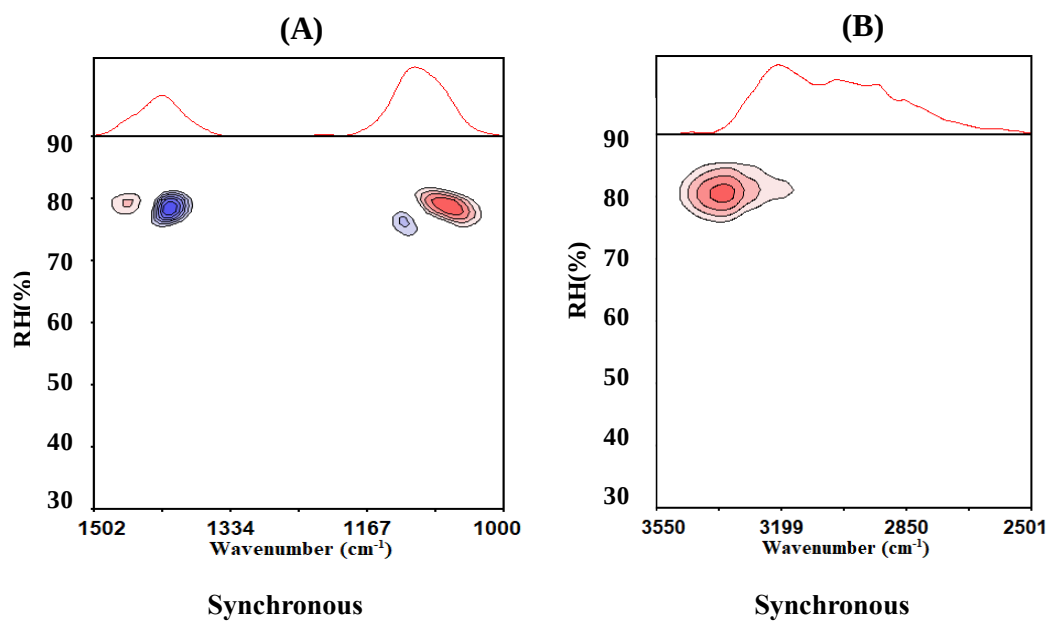


Fig.S3 Synchronous PCMW2D spectra in the (A) 1502 $\sim$ 1000 cm^{-1} and (B) 3550 $\sim$ 2501 cm^{-1} region during the humidification process of 300nm AS particles.

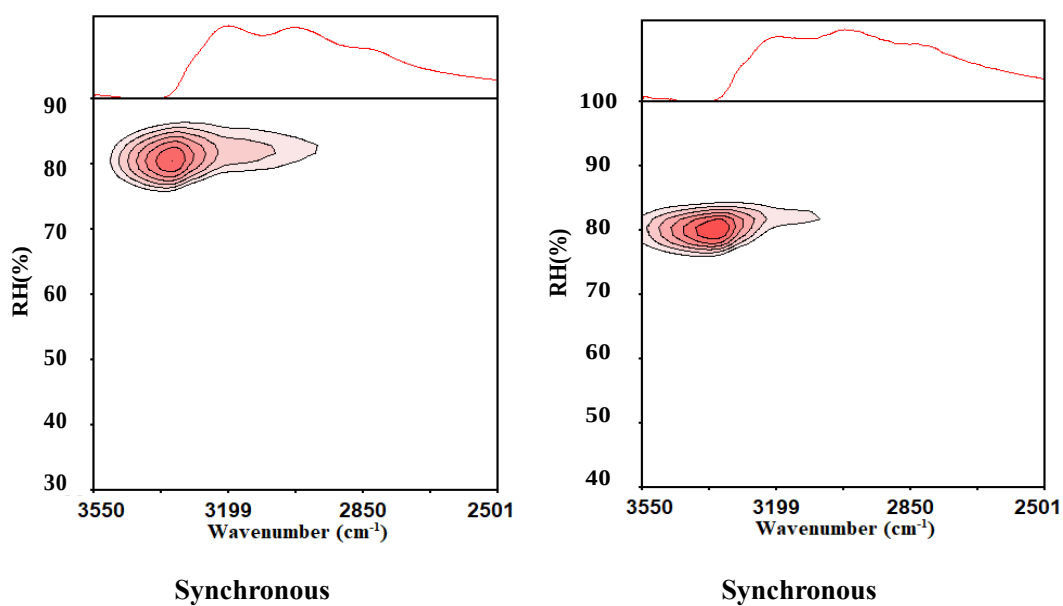


Fig.S4 Synchronous PCMW2D spectra in the 3550 ~ 2501 cm⁻¹ during the humidification process of AS fine particles (A) 400nm AS particles; (B) 600nm AS particles.

Table S1 Summary of deliquescence RH (DRH) and efflorescence RH (ERH) from literature.

Compounds	DRH (%)	ERH (%)	Ref
AS	79.9 ± 0.3	37.9 ± 0.5	Ahn et al. (2010) (Ahn et al., 2010)
	79 ± 1	33 ± 2	Cziczo et al. (1999)(Cziczo & Abbatt, 1999)
	79.7	51.9 ~ 34.9	Cai et al. (2017) (Cai et al., 2017)
	80	35	Schlenker et al. (2004)(Schlenker, Malinowski, Martin, Hung, & Rudich, 2004)
	79 ± 2	/	Li et al. (2017) (Li et al., 2017)
	/	38 ~ 40	Yeung et al. (2009) (Yeung, Y., & Chan, 2009)

Reference:

1. Onasch, T. B.; Siefert, R. L.; Brooks, S. D.; Prenni, A. J.; Murray, B.; Wilson, M. A.; Tolbert, M. A., Infrared spectroscopic study of the deliquescence and efflorescence of ammonium sulfate aerosol as a function of temperature. *Journal of Geophysical Research: Atmospheres* **1999**, *104* (D17), 21317-21326.
2. Nájera, J. J.; Horn, A. B., Infrared spectroscopic study of the effect of oleic acid on the deliquescence behaviour of ammonium sulfate aerosol particles. *Physical Chemistry Chemical Physics* **2009**, *11* (3), 483-494.