# 1 Table S1: Deliquescence and efflorescence relative humidity of ammonium sulfate below 100 nm reported by difference studies in

2 temperature ranging from 290-300K

Deliquescence relative	Efflorescence relative	Technique	Reference
humidity (DRH)	humidity (ERH)	(initial particle size)	
80-86%* (8 nm)		HTDMA	Hämeri et al. (2000)
80-85%* (10 nm)		(8.10,15,30,50 nm)	(cf .Figure 2a, 2b, 2c, 2d, and 2e)
80-90%* (15 nm)			
78-80%* (30 nm)			
76-79%* (50 nm)			
76-80%*	65%*	HTDMA	Gysel et al. (2002)
		(100 nm)	(cf. Figure 2)
82% (6 nm)	34% (6 nm)	HTDMA	Biskos et al. (2006b)
81% (8 nm)	33% (8 nm)	(6,8,10,20,40,60 nm)	
80% (10 nm)	35% (10 nm)		
82% (20 nm)	35% (20 nm)		
80% (40 nm)	36% (40 nm)		
80% (60 nm)	33% (6 nm)		
-	27-31%* (43.7 nm)	HTDMA	Gao et al. (2006)
	21-30.7%* (47 nm)	(43.7,47 nm)	(cf. Figure 5)
78-81%*	-	HTDMA	Duplissy et al. (2009)
		(100 nm)	(cf. Figure 4)
77-78%*	-	HTDMA	Duplissy et al. (2009)

		(100 nm)	(cf. Figure 4)	
78-80%*	29-34%*	HTDMA	Mikhailov et al. (2009) (cf. Fig4)	
		(100 nm)		
77-78%	-	HTDMA	Wu et al. (2011)	
		(100 nm)		

3 -: Not reported

### 4 \*: Data retrieved from figures in the references

- 5 80-86%: Non-prompt deliquescence of 8-nm ammonium sulfate from 80% to 86% RH
- 6 27-31%: Non-prompt efflorescence of 43.7-nm ammonium sulfate from 31% to 27% RH
- 7 82%: Prompt deliquescence of 6-nm ammonium sulfate at 82% RH

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Table S2. Residence time (s) for the water equilibrium for particles with diameter ranging from 6
to 100 nm particles at RH=90% at 25°C

	1	0.1	0.01	0.001
100nm	6.26 x10 <sup>-6</sup>	3.55 x10 <sup>-5</sup>	3.12x10 <sup>-4</sup>	0.0031
60nm	6.04 x10 <sup>-6</sup>	3.34 x10 <sup>-5</sup>	3.07x10 <sup>-4</sup>	0.0030
20nm	6.03 x10 <sup>-7</sup>	5.17 x10 <sup>-6</sup>	5.08x10 <sup>-5</sup>	5.07x10 <sup>-4</sup>
10nm	1.88 x10 <sup>-7</sup>	1.74 x10 <sup>-6</sup>	1.73x10 <sup>-5</sup>	1.72x10 <sup>-4</sup>
8nm	3.10x10 <sup>-8</sup>	1.93x10 <sup>-7</sup>	1.82x10 <sup>-6</sup>	1.81x10 <sup>-5</sup>
6nm	1.48x10 <sup>-8</sup>	$1.08 \times 10^{-7}$	1.04x10 <sup>-6</sup>	1.03x10 <sup>-5</sup>
		between nano-DMAs	in the nano-HTDM	A system at RH below
		between nano-DMAs Offset(average) <sup>a</sup>	in the nano-HTDM	A system at RH below
100-nm (NH4)	)2SO4	between nano-DMAs Offset(average) <sup>a</sup> 0.619318	in the nano-HTDM Size agreement and nano-DMA 0.619318%	A system at RH below t between nano-DMA1
100-nm (NH <sub>4</sub> ) 60-nm (NH <sub>4</sub> ) <sub>2</sub>	) <sub>2</sub> SO <sub>4</sub>	between nano-DMAs Offset(average) <sup>a</sup> 0.619318 0.298691	in the nano-HTDM Size agreement and nano-DMA 0.619318% 0.4978%	A system at RH below
100-nm (NH <sub>4</sub> ) 60-nm (NH <sub>4</sub> ) <sub>2</sub> 20-nm (NH <sub>4</sub> ) <sub>2</sub>	)2SO4 SO4	between nano-DMAs Offset(average) <sup>a</sup> 0.619318 0.298691 0.278311	in the nano-HTDM Size agreement and nano-DMA 0.619318% 0.4978% 1.3916%	A system at RH below
100-nm (NH4) 60-nm (NH4)2 20-nm (NH4)2 10-nm (NH4)2	)2SO4 SO4 SO4_ SO4_	between nano-DMAs Offset(average) <sup>a</sup> 0.619318 0.298691 0.278311 0.089647	in the nano-HTDM. Size agreement and nano-DMA 0.619318% 0.4978% 1.3916% 0.8965%	A system at RH below
100-nm (NH <sub>4</sub> ) 60-nm (NH <sub>4</sub> ) <sub>2</sub> 20-nm (NH <sub>4</sub> ) <sub>2</sub> 10-nm (NH <sub>4</sub> ) <sub>2</sub> 8-nm (NH <sub>4</sub> ) <sub>2</sub> S	)2SO4 SO4 SO4 SO4 SO4	between nano-DMAs Offset(average) <sup>a</sup> 0.619318 0.298691 0.278311 0.089647 -0.01598	in the nano-HTDM. Size agreement and nano-DMA 0.619318% 0.4978% 1.3916% 0.8965% -0.19975%	A system at RH below

19 <sup>a</sup> Calculation from ( $\overline{D}_{measured by nano-DMA2} - D_{selected by nano-DMA1}$ )

20 <sup>b</sup>Calculation from  $[(\overline{D}_{measured by nano-DMA2} - D_{selected by nano-DMA1})/D_{selected by nano-DMA1}] \times 100\%$ 





**23** Figure S1. Methods for measuring hygroscopicity of atmospheric aerosol particles in different size  $(D_p)$ .



Figure S2. (a) Number concentration scanned for water nanoparticles by the nano-DMA2 at RH below 5 % at 298 K.
(b) Normalized number size distribution scanned for 22-nm PSL nanoparticles by nano-DMA2 after calibration.



Figure S3. Number size distribution of ammonium sulfate (AS) nanoparticles (black solid square) generated by the
electrospray. (a) 20mM, (b) 5mM, and (c) 1mM AS solution. The dotted line marks peak diameter from the Gaussian
fits for the scan (red curve). The black solid lines mark the diameters of the monodispersed nanoparticles selected by
the nano-DMA1.





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Figure S4. Deliquescence-mode (a) and efflorescence-mode (b) of 100-nm ammonium sulfate (AS) aerosol nanoparticles. The measured (black square) and fitted (solid lines) normalized size distribution are shown for increasing RH (5% $\rightarrow$ X%, where X is the RH value given in each panel) and decreasing RH (5% $\rightarrow$ 97% $\rightarrow$ X%, where X is the RH value given in each panel), respectively.



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**Figure S5.** Deliquescence-mode (**a**) and efflorescence-mode (**b**) of 60-nm ammonium sulfate (AS) aerosol nanoparticles. The measured (black square) and fitted (solid lines) normalized size distribution are shown for increasing RH (5% $\rightarrow$ X%, where X is the RH value given in each panel) and decreasing RH (5% $\rightarrow$ 97% $\rightarrow$ X%, where X is the RH value given in each panel), respectively.





Figure S6. Deliquescence-mode (a) and efflorescence-mode (b) of 8-nm ammonium sulfate (AS) aerosol nanoparticles.
The measured (black square) and fitted (solid lines, single-mode log-normal fit) normalized size distribution are shown
for increasing RH (5%→X%, where X is the RH value given in each panel) and decreasing RH (5%→97%→X%,
where X is the RH value given in each panel), respectively.





Figure S7. Mobility-diameter hygroscopic growth factors (*g<sub>f</sub>*, black squares), deliquescence and efflorescence relative
humidity (DRH&ERH, black dashed lines) of ammonium sulfate (AS) nanoparticles with dry diameter from 6 to 100
nm, respectively. Red squares and dashed lines show the respective results from Biskos et al. (2006b), respectively.





Figure S8. (a) Comparison of mobility-diameter hygroscopic growth factors  $(g_f)$  of 100-nm (black square) with 6-nm (red square) ammonium sulfate (AS) nanoparticles. (b) Dependence of deliquescence and efflorescence relative humidity (DRH&ERH) of ammonium sulfate (AS) on dry volume equivalent diameter  $(D_{ve})$ . The measured DRH and ERH of ammonium sulfate within RH uncertainty (black line + black square) compared with data from Biskos et al. (2006b) (red square) in the volume equivalent diameter with shape factor ( $\chi$ =1.02) range from 5 to 100 nm.





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**Figure S9.** Deliquescence-mode (a) and efflorescence-mode (b) of 20-nm sodium sulfate aerosol nanoparticles. The measured (black square) and fitted (solid lines) normalized size distribution are shown for increasing RH (5% $\rightarrow$ X%, where X is the RH value given in each panel) and decreasing RH (5% $\rightarrow$ 97% $\rightarrow$ X%, where X is the RH value given in each panel), respectively. Red/blue solid line is fitted by a single-mode log-normal fit. Red, blue, and black lines are fitted by a double-mode log-normal fit. The voltage applied to the nano-DMAs (0-12500 V) is kept within ±1% around the set value shown in the voltage meter.



**Figure S10.** Deliquescence-mode (**a**) and efflorescence-mode (**b**) of 6-nm sodium sulfate aerosol nanoparticles. The measured (black square) and fitted (solid lines) normalized size distribution are shown for increasing RH (5% $\rightarrow$ X%, where X is the RH value given in each panel) and decreasing RH (5% $\rightarrow$ 97% $\rightarrow$ X%, where X is the RH value given in each panel), respectively. Red/blue solid line is fitted by a single-mode log-normal fit. Red, blue, and black lines are fitted by a double-mode log-normal fit. The voltage applied to the nano-DMAs (0-350 V) is kept within ±1% around the set value shown in the voltage meter.



Figure S11. (a) Comparison of mobility-diameter hygroscopic growth factors  $(g_f)$  of 20-nm (a) and 60-nm (b) ammonium sulfate (AS) nanoparticles with Biskos et al. (2006b) and Hu et al. (2010). (black squares: in this study; red square: Biskos et al. (2006b); blue square: Hu et al. (2010)). (c) Comparison of mobility-diameter hygroscopic growth factors of 20-nm Na<sub>2</sub>SO<sub>4</sub> nanoparticles with Hu et al. (2010). (black squares: in this study; red square: Hu et al. (2010)). (d) Mobility-diameter hygroscopic growth factors of Na<sub>2</sub>SO<sub>4</sub> nanoparticles with diameter from 6 nm to 14~16 um at 84% RH (black solid squares: in this study; black open square: Hu et al. (2010); black open cycle: Tang et al. (2007)). A fitting equation ( $g_f = \frac{1.804}{1 + (0.5267 * D)^{-0.8194}}$ ) based on this study at 6-nm, 20-nm Na<sub>2</sub>SO<sub>4</sub>, and 14~16 um data from Tang et al. (2007).

#### 125 S1. Calculation of sizing offset of 10-nm AS

126 The mobility growth factor  $(g_f)$  is given by:

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$$g_f = \frac{D_m(RH)}{D_m(<10\ \%\ RH)}$$
(S1)

128  $g_f$  was from the data of Biskos et al. (2006b) in the different RHs (see the SI. Fig.5).  $D_m$  was

retrieved the data of Biskos et al. (2006b) in the different RHs (see the SI. Fig.2) as follows:



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Figure S12. Measured (black square) and fitted (red solid line) normalized number size distributions are show for
ammonium sulfate aerosol particles at 25% RH. The black square symbols show the data of Biskos et al. (2006b) (see
the S1. Fig. 2).

Therefore, the initial dry mobility diameter ( $D_m$  (< 5% RH)) was obtained using Eq. (S1) based on values of  $g_i$  and  $D_m$  in the different RHs (see SI. Table S4). We further calculated the average sizing offset of 10-nm ammonium sulfate of Biskos et al. (2006b) system based on the values of  $D_m$  (< 5% RH). The average sizing offset of 10-nm was ~3.1%.

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Relative humidity	$D_m$	$g_{f}$	$D_m$ (<5 % RH)
25%	10.3982439	0.992914120	10.47245043
76%	10.38867117	1.017488426	10.21011237
78%	10.54314064	1.027692308	10.25904404
80%	13.31036607	1.293796610	10.28783502
44%	11.56059002	1.120463542	10.31768513
35%	11.24527292	1.084064417	10.37325157
34%	10.59107394	1.007786565	10.50924304
32%	10.24542551	1.003831854	10.20631639
31%	10.20845456	1.001920937	10.18888236
30%	10.38101934	1.001441750	10.36607405
29%	10.27755951	1.003183756	10.2779752
24%	10.26077112	0.997295121	10.28860053

**Table S4.** The values of  $D_m$ ,  $g_f$ , and  $D_m$  (< 5% RH) of 10-nm ammonium sulfate of Biskos et al. 145 (2006b) system in the different RHs.