

Supporting Information

Impact of the particle mixing state on the hygroscopicity of internally mixed sodium chloride-ammonium sulfate single droplets: A theoretical and experimental study

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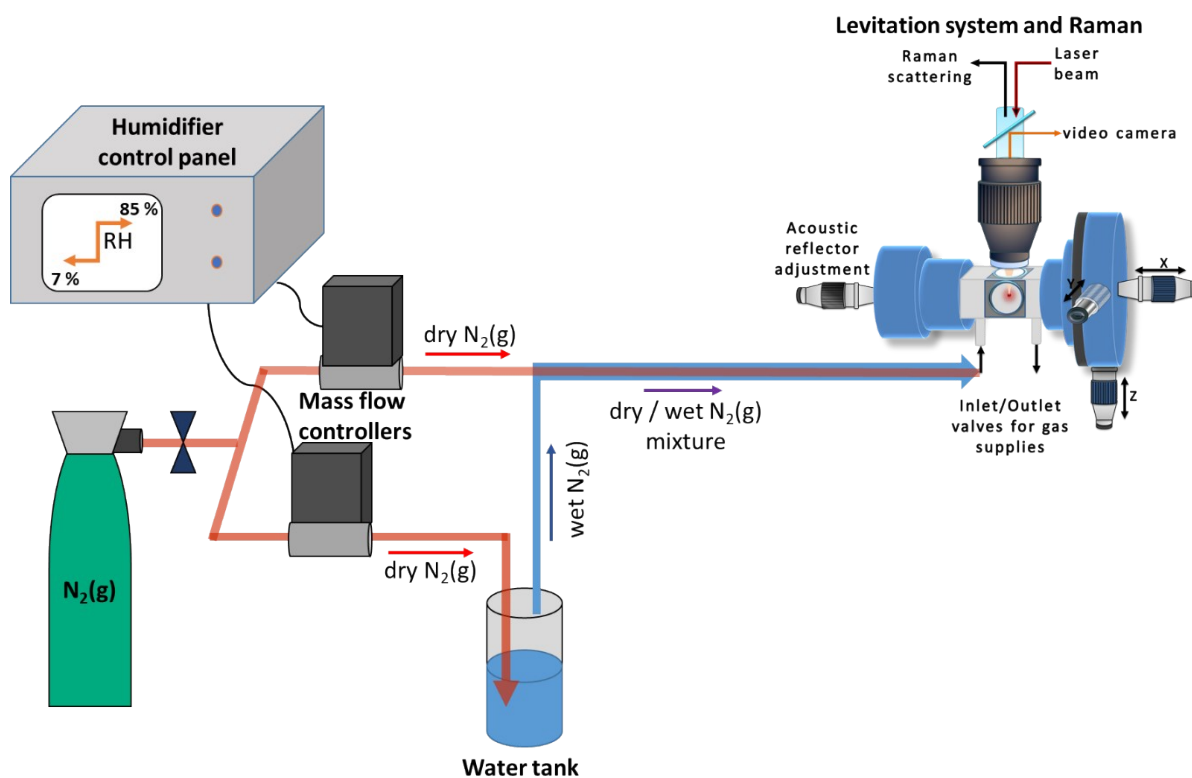
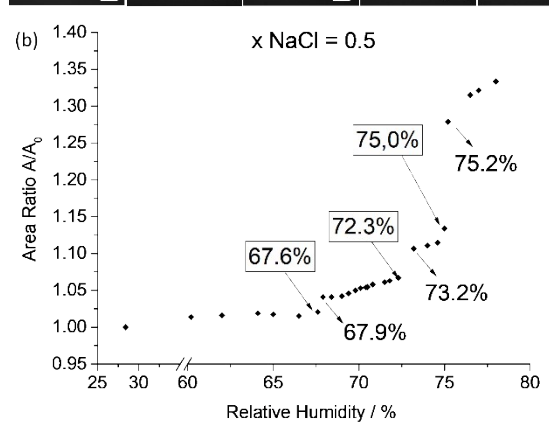
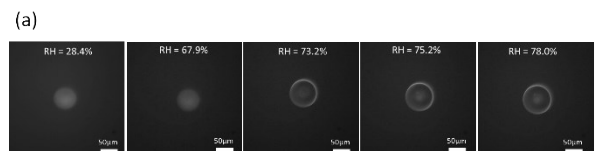
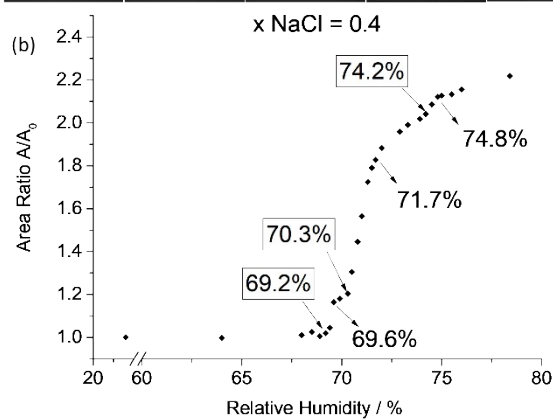
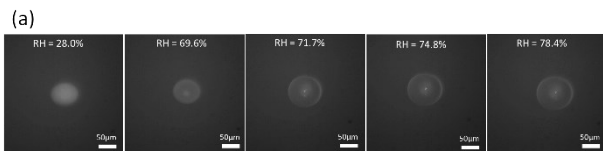
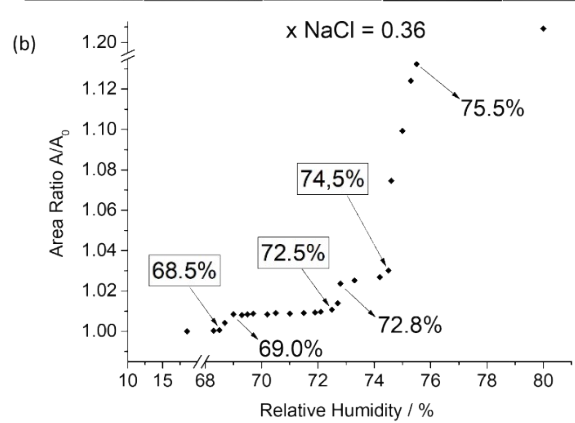
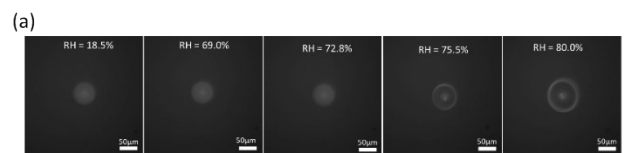
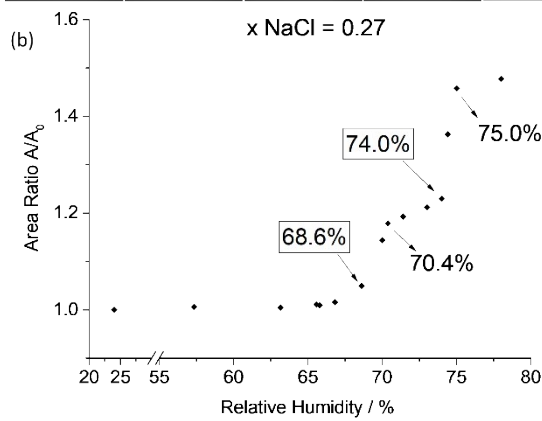
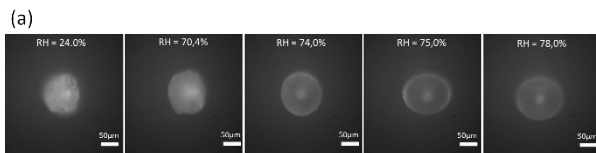
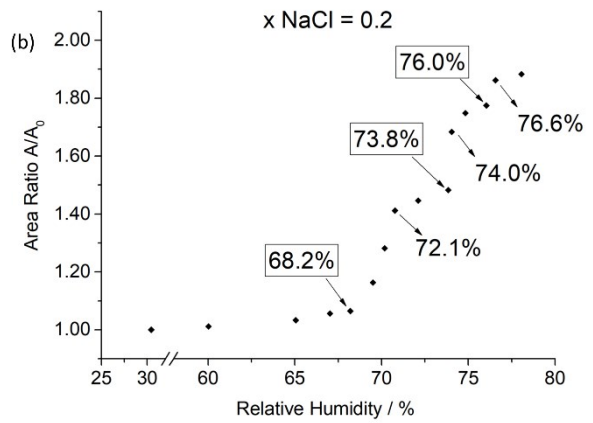
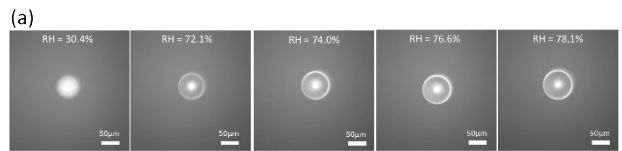
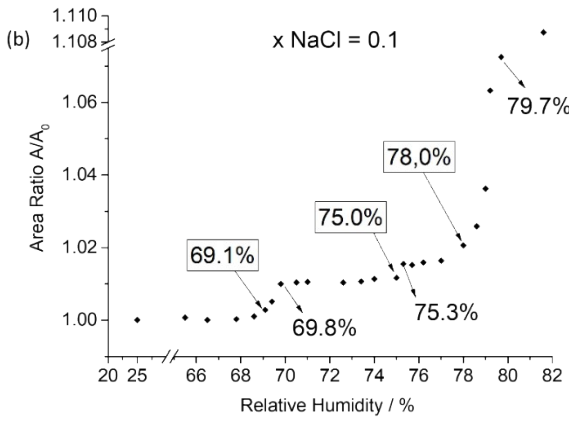
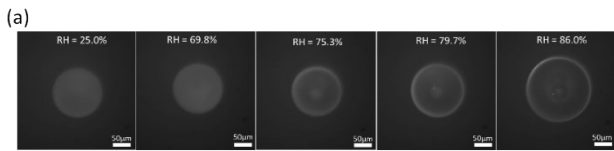


Fig. S1 Schematic diagram of experimental setup



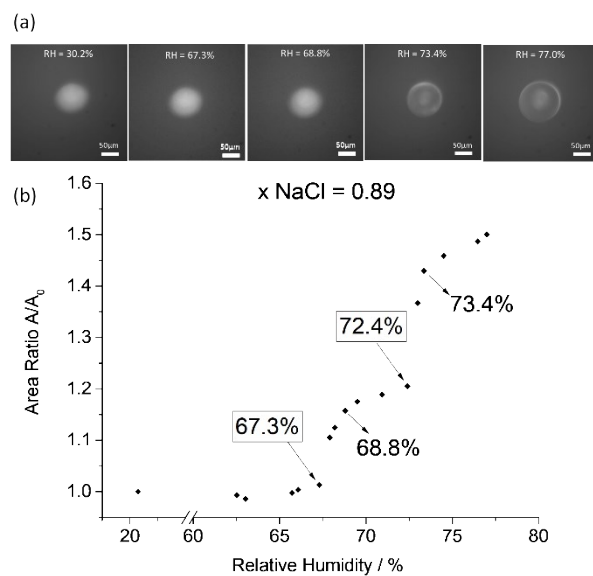
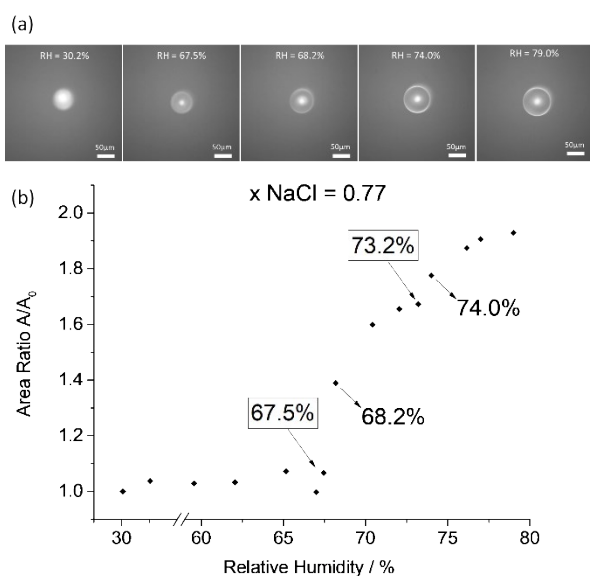
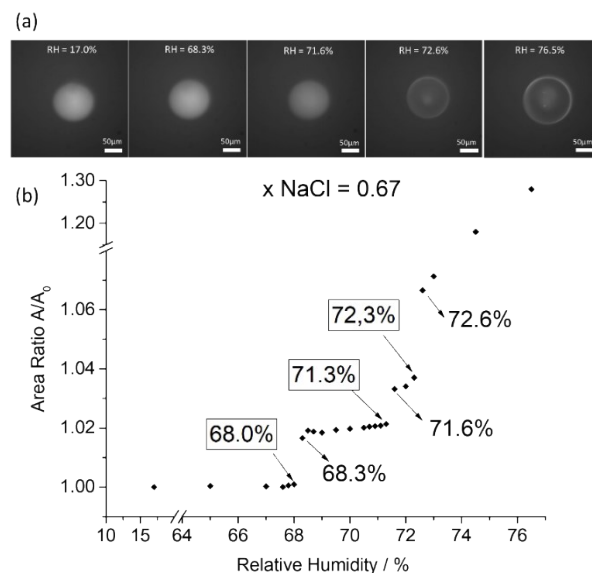
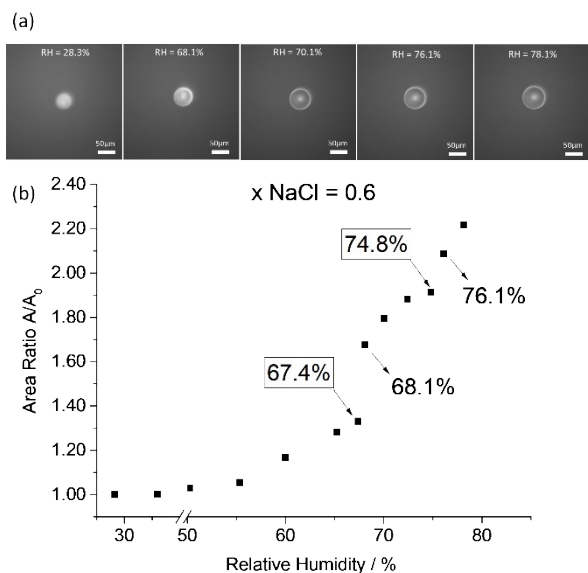


Fig. S2 (a) Optical images and (b) humidograms of mixed single particles of NaCl/ $(\text{NH}_4)_2\text{SO}_4$ containing NaCl mole fraction of 0.1, 0.2, 0.27, 0.36, 0.4, 0.5, 0.6, 0.67, 0.77 and 0.89.

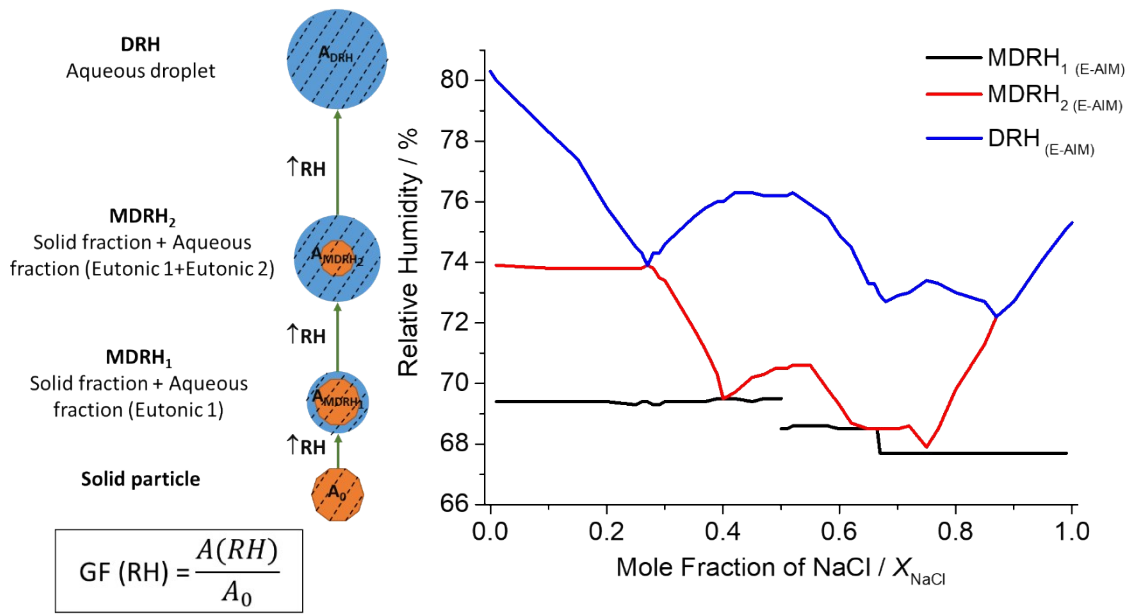


Fig. S3 Illustration of the phase transitions of internally mixed NaCl/(NH₄)₂SO₄ particles.

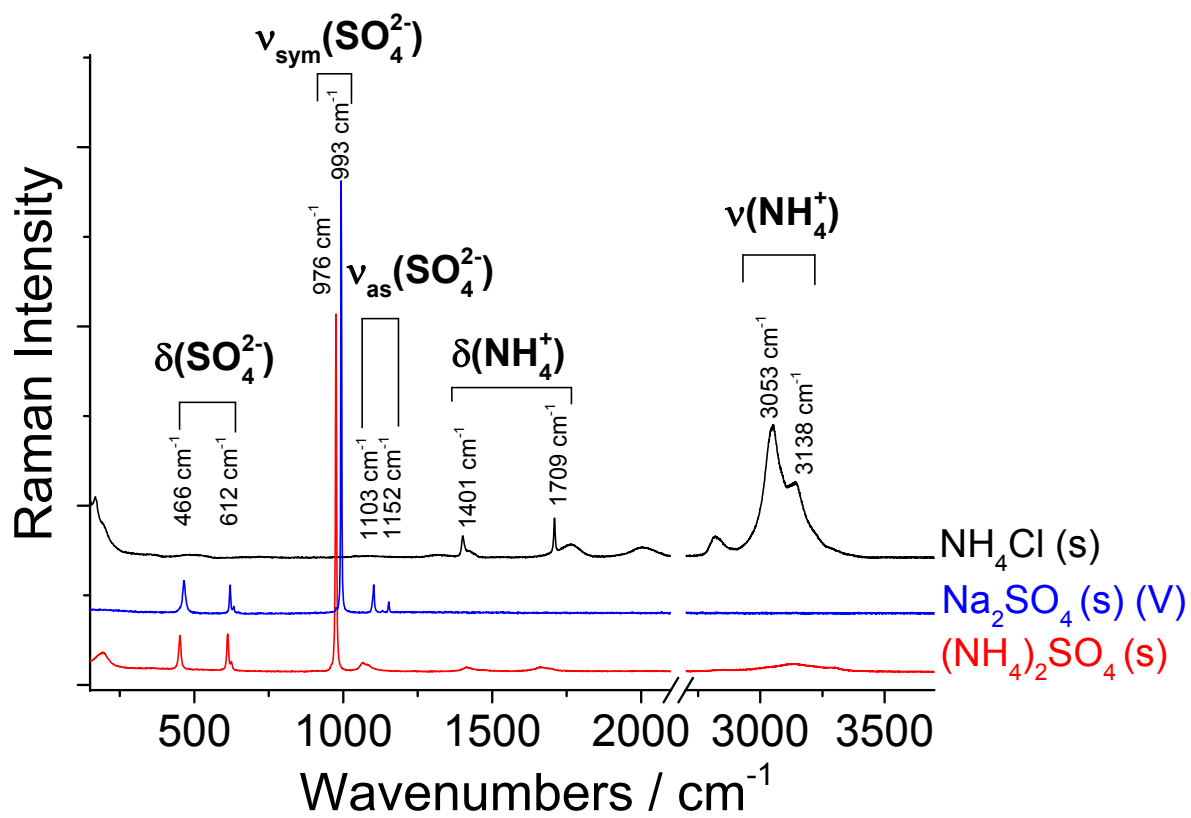


Fig. S4. Raman spectra of pure solid NH_4Cl , Na_2SO_4 and $(\text{NH}_4)_2\text{SO}_4$

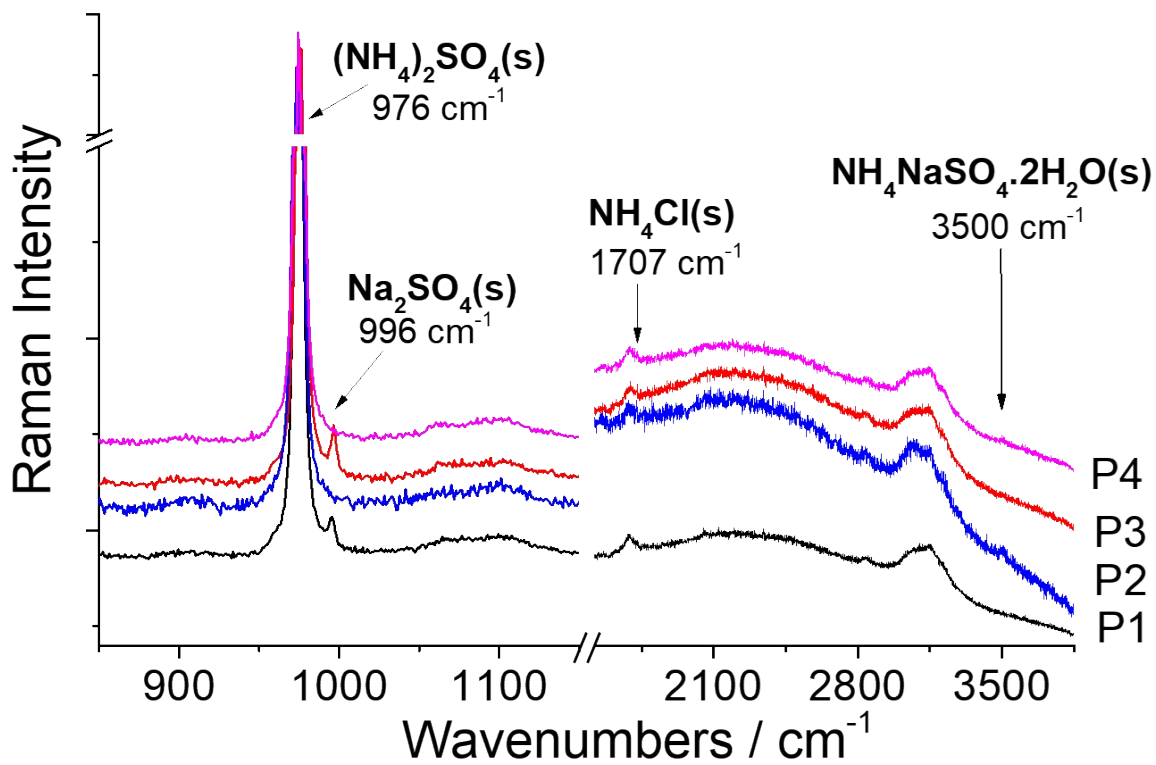


Figure S5. Raman spectra showing variable composition of several levitated dried particles containing mixed $\text{NaCl}/(\text{NH}_4)_2\text{SO}_4$ ($x_{\text{NaCl}} = 0.1$)

Table S1. Summary list of input parameters used in E-AIM model

E-AIM model	Model III				
Type of calculation	Parametric, varying: (a) relative humidity, or total water				
Output	Column				
Water content of the system	Relative humidity				
Start value	0.65				
End value	0.85				
No. of points	100				
Inorganic composition in moles per m³	xNaCl	Na⁺	Cl⁻	NH₄⁺	SO₄⁻
	0	0	0	2	1
	0.1	0.1	0.1	1.8	0.9
	0.2	0.2	0.2	1.6	0.8
	0.27	0.27	0.27	1.46	0.73
	0.36	0.36	0.36	1.28	0.64
	0.4	0.4	0.4	1.2	0.6
	0.5	0.5	0.5	1	0.5
	0.55	0.55	0.55	0.9	0.45
	0.6	0.6	0.6	0.8	0.4
	0.67	0.67	0.67	0.66	0.33
	0.77	0.77	0.77	0.46	0.23
0.89	0.89	0.89	0.22	0.11	
1	1	1	0	0	
Trace gases	Default: partitioning is calculated for each gas				
Solid phases	Default: all solids can form				

Table S2. E-AIM simulation of chemical species and concentrations, at different humidification levels, of a particle initially containing 0.1 mol.m⁻³ of Na⁺ and Cl⁻, 1.8 mol.m⁻³ of NH₄⁺ and 0.9 mol.m⁻³ of SO₄²⁻ (xNaCl = 0.1).

State	Aqueous phase			Solid phase		
Solid	Species	mol.m⁻³	X	Species	mol.m⁻³	X
	Na ⁺	0.000	0.000	Na ⁺	0.100	0.035
	Cl ⁻	0.000	0.000	Cl ⁻	0.100	0.035
	NH ₄ ⁺	0.000	0.000	NH ₄ ⁺	1.800	0.620
	SO ₄ ²⁻	0.000	0.000	SO ₄ ²⁻	0.900	0.310
	total	0.000	0.000	total	2.900	1.000
mol.m⁻³	0			NH ₄ Cl=0.100 ; (NH ₄) ₂ SO ₄ =0.800 ; NH ₄ NaSO ₄ ·2H ₂ O=0.100		
MDRH₁ 69.4%	Species	mol.m⁻³	X	Species	mol.m⁻³	X
	Na ⁺	0.040	0.088	Na ⁺	0.060	0.025
	Cl ⁻	0.100	0.220	Cl ⁻	0.000	0.000
	NH ₄ ⁺	0.230	0.505	NH ₄ ⁺	1.570	0.642
	SO ₄ ²⁻	0.085	0.187	SO ₄ ²⁻	0.815	0.333
	total	0.455	1.000	total	2.445	1.000
mol.m⁻³	First solubilized species: NH ₄ Cl=0.100 ; (NH ₄) ₂ SO ₄ =0.045 ; NH ₄ NaSO ₄ ·2H ₂ O=0.040			(NH ₄) ₂ SO ₄ =0.755 ; NH ₄ NaSO ₄ ·2H ₂ O=0.060		
MDRH₂ 73.8%	Species	mol.m⁻³ (accumulate)	X	Species	mol.m⁻³	X
	Na ⁺	0.060 (0.100)	0.108 (0.099)	Na ⁺	0.000	0.000
	Cl ⁻	0.000 (0.100)	0.000 (0.099)	Cl ⁻	0.000	0.000
	NH ₄ ⁺	0.310 (0.540)	0.559 (0.535)	NH ₄ ⁺	1.260	0.667
	SO ₄ ²⁻	0.185 (0.270)	0.333 (0.267)	SO ₄ ²⁻	0.630	0.333
	total	0.555 (1.010)	1.000 (1.000)	total	1.890	1.000
mol.m⁻³	Second solubilized species: (NH ₄) ₂ SO ₄ =0.125 ; NH ₄ NaSO ₄ ·2H ₂ O=0.060			(NH ₄) ₂ SO ₄ =0.630		
DRH 78.4%	Species	mol.m⁻³ (accumulate)	X	Species	mol.m⁻³	X
	Na ⁺	0.000 (0.100)	0.000 (0.034)	Na ⁺	0.000	0.000
	Cl ⁻	0.000 (0.100)	0.000 (0.034)	Cl ⁻	0.000	0.000
	NH ₄ ⁺	1.260 (1.800)	0.667 (0.621)	NH ₄ ⁺	0.000	0.000
	SO ₄ ²⁻	0.630 (0.900)	0.333 (0.310)	SO ₄ ²⁻	0.000	0.000
	total	1.890 (2.900)	1.000 (1.000)	total	0.000	0.000
mol.m⁻³	Last solubilized species: (NH ₄) ₂ SO ₄ =0.630			0		

Table S3. E-AIM simulation of chemical species and concentrations, at different humidification levels, of a particle initially containing 0.36 mol.m⁻³ of Na⁺ and Cl⁻, 1.28 mol.m⁻³ of NH₄⁺ and 0.64 mol.m⁻³ of SO₄²⁻ (xNaCl = 0.36).

State	Aqueous phase			Solid phase		
Solid	Species	mol.m⁻³	X	Species	mol.m⁻³	X
	Na ⁺	0.000	0.000	Na ⁺	0.360	0.136
	Cl ⁻	0.000	0.000	Cl ⁻	0.360	0.136
	NH ₄ ⁺	0.000	0.000	NH ₄ ⁺	1.280	0.485
	SO ₄ ²⁻	0.000	0.000	SO ₄ ²⁻	0.640	0.242
	total	0.000	0.000	total	2.640	1.000
mol.m⁻³	0			(NH ₄) ₂ SO ₄ =0.280 ; NH ₄ NaSO ₄ .2H ₂ O=0.360; NH ₄ Cl=0.360		
MDRH₁ 69.4%	Species	mol.m⁻³	X	Species	mol.m⁻³	X
	Na ⁺	0.144	0.088	Na ⁺	0.216	0.216
	Cl ⁻	0.360	0.219	Cl ⁻	0.000	0.000
	NH ₄ ⁺	0.830	0.506	NH ₄ ⁺	0.450	0.450
	SO ₄ ²⁻	0.307	0.187	SO ₄ ²⁻	0.333	0.333
	total	1.641	1.000	total	0.999	1.000
mol.m⁻³	First solubilized species: (NH ₄) ₂ SO ₄ =0.163 ; NH ₄ NaSO ₄ .2H ₂ O=0.144; NH ₄ Cl=0.360			(NH ₄) ₂ SO ₄ =0.117 ; NH ₄ NaSO ₄ .2H ₂ O= 0.216		
MDRH₂ 71.5%	Species	mol.m⁻³ (accumulate)	X	Species	mol.m⁻³	X
	Na ⁺	0.057 (0.201)	0.110 (0.093)	Na ⁺	0.159	0.333
	Cl ⁻	0.000 (0.360)	0.000 (0.166)	Cl ⁻	0.000	0.000
	NH ₄ ⁺	0.291 (1.121)	0.557 (0.518)	NH ₄ ⁺	0.159	0.333
	SO ₄ ²⁻	0.174 (0.481)	0.333 (0.222)	SO ₄ ²⁻	0.159	0.333
	total	0.523 (2.163)	1.000 (1.000)	total	0.476	1.000
mol.m⁻³	Second solubilized species: (NH ₄) ₂ SO ₄ =0.117 ; NH ₄ NaSO ₄ .2H ₂ O= 0.057			NH ₄ NaSO ₄ .2H ₂ O = 0.159		
DRH 75.6%	Species	mol.m⁻³ (accumulate)	X	Species	mol.m⁻³	X
	Na ⁺	0.159 (0.360)	0.333 (0.136)	Na ⁺	0.00	0.00
	Cl ⁻	0.000 (0.360)	0.000 (0.136)	Cl ⁻	0.00	0.00
	NH ₄ ⁺	0.159 (1.280)	0.333 (0.485)	NH ₄ ⁺	0.00	0.00
	SO ₄ ²⁻	0.159 (0.640)	0.333 (0.242)	SO ₄ ²⁻	0.00	0.00
	total	0.476 (2.640)	1.000 (1.000)	total	0.00	0.00
mol.m⁻³	Last solubilized species: (NH ₄) ₂ SO ₄ = 0.159			0		

Table S4. E-AIM simulation of chemical species and concentrations, at different humidification levels, of a particle initially containing 0.55 mol.m⁻³ of Na⁺ and Cl⁻, 0.90 mol.m⁻³ of NH₄⁺ and 0.45 mol.m⁻³ of SO₄²⁻ (xNaCl = 0.55).

State	Aqueous phase			Solid phase		
Solid	Species	mol.m⁻³	X	Species	mol.m⁻³	X
	Na ⁺	0.000	0.000	Na ⁺	0.550	0.224
	Cl ⁻	0.000	0.000	Cl ⁻	0.550	0.224
	NH ₄ ⁺	0.000	0.000	NH ₄ ⁺	0.900	0.368
	SO ₄ ²⁻	0.000	0.000	SO ₄ ²⁻	0.450	0.184
	total	0.000	0.000	total	2.450	1.000
mol.m⁻³	0			NaCl=0.100 ; NH ₄ Cl=0.450 ; NH ₄ NaSO ₄ .2H ₂ O=0.450		
MDRH₁ 68.4%	Species	mol.m⁻³	X	Species	mol.m⁻³	X
	Na ⁺	0.165	0.244	Na ⁺	0.385	0.217
	Cl ⁻	0.241	0.356	Cl ⁻	0.309	0.174
	NH ₄ ⁺	0.206	0.304	NH ₄ ⁺	0.694	0.391
	SO ₄ ²⁻	0.065	0.096	SO ₄ ²⁻	0.385	0.217
	total	0.677	1.000	total	1.773	1.000
mol.m⁻³	First solubilized species: NaCl=0.100 ; NH ₄ Cl=0.141 ; NH ₄ NaSO ₄ .2H ₂ O=0.065			NH ₄ Cl=0.309 ; NH ₄ NaSO ₄ .2H ₂ O=0.385		
MDRH₂ 70.2%	Species	mol.m⁻³ (accumulate)	X	Species	mol.m⁻³	X
	Na ⁺	0.139 (0.304)	0.134 (0.178)	Na ⁺	0.246	0.333
	Cl ⁻	0.309 (0.550)	0.298 (0.321)	Cl ⁻	0.000	0.000
	NH ₄ ⁺	0.448 (0.654)	0.433 (0.382)	NH ₄ ⁺	0.246	0.333
	SO ₄ ²⁻	0.139 (0.204)	0.134 (0.119)	SO ₄ ²⁻	0.246	0.333
	total	1.036 (1.712)	1.000 (1.000)	total	0.738	1.000
mol.m⁻³	Second solubilized species: NH ₄ Cl=0.309 ; NH ₄ NaSO ₄ .2H ₂ O=0.139			NH ₄ NaSO ₄ .2H ₂ O= 0.246		
DRH 75.8%	Species	mol.m⁻³ (accumulate)	X	Species	mol.m⁻³	X
	Na ⁺	0.246 (0.550)	0.333 (0.224)	Na ⁺	0.00	0.00
	Cl ⁻	0.000 (0.550)	0.000 (0.224)	Cl ⁻	0.00	0.00
	NH ₄ ⁺	0.246 (0.900)	0.333 (0.368)	NH ₄ ⁺	0.00	0.00
	SO ₄ ²⁻	0.246 (0.450)	0.333 (0.184)	SO ₄ ²⁻	0.00	0.00
	total	0.738 (2.450)	1.000 (1.000)	total	0.00	0.00
mol.m⁻³	Last solubilized species: NH ₄ NaSO ₄ .2H ₂ O= 0.246			0		

Table S5. E-AIM simulation of chemical species and concentrations, at different humidification levels, of a particle initially containing 0.67 mol.m⁻³ of Na⁺ and Cl⁻, 0.66 mol.m⁻³ of NH₄⁺ and 0.33 mol.m⁻³ of SO₄²⁻ (xNaCl = 0.67).

State	Aqueous phase			Solid phase		
Solid	Species	mol.m⁻³	X	Species	mol.m⁻³	X
	Na ⁺	0.000	0.000	Na ⁺	0.670	0.288
	Cl ⁻	0.000	0.000	Cl ⁻	0.670	0.288
	NH ₄ ⁺	0.000	0.000	NH ₄ ⁺	0.660	0.283
	SO ₄ ²⁻	0.000	0.000	SO ₄ ²⁻	0.330	0.141
	total	0.000	0.000	total	2.330	1.000
mol.m⁻³	0			NaCl= 0.110 ; NH ₄ Cl= 0.560 ; Na ₂ SO ₄ = 0.235 ; NH ₄ NaSO ₄ .2H ₂ O=0.095		
MDRH₁ 67.7%	Species	mol.m⁻³	X	Species	mol.m⁻³	X
	Na ⁺	0.020	0.269	Na ⁺	0.650	0.288
	Cl ⁻	0.030	0.397	Cl ⁻	0.640	0.284
	NH ₄ ⁺	0.020	0.265	NH ₄ ⁺	0.640	0.284
	SO ₄ ²⁻	0.005	0.069	SO ₄ ²⁻	0.325	0.144
	total	0.076	1.000	total	2.254	1.000
mol.m⁻³	First solubilized species: NaCl= 0.020 ; NH ₄ Cl= 0.010 ; NH ₄ NaSO ₄ .2H ₂ O=0.005			NaCl= 0.090 ; NH ₄ Cl= 0.550 ; Na ₂ SO ₄ = 0.235 ; NH ₄ NaSO ₄ .2H ₂ O=0.090		
MDRH₂ 68.4%	Species	mol.m⁻³ (accumulate)	X	Species	mol.m⁻³	X
	Na ⁺	0.444 (0.463)	0.245 (0.246)	Na ⁺	0.207	0.465
	Cl ⁻	0.640 (0.670)	0.354 (0.356)	Cl ⁻	0.000	0.000
	NH ₄ ⁺	0.550 (0.570)	0.304 (0.302)	NH ₄ ⁺	0.090	0.202
	SO ₄ ²⁻	0.177 (0.181)	0.098 (0.096)	SO ₄ ²⁻	0.148	0.333
	total	1.809 (1.884)	1.000 (1.000)	total	0.445	1.000
mol.m⁻³	Second solubilized species: NaCl= 0.090 ; NH ₄ Cl= 0.550 ; Na ₂ SO ₄ = 0.177			NH ₄ NaSO ₄ .2H ₂ O=0.090 ; Na ₂ SO ₄ = 0.058		
MDRH₃ 73.0%	Species	mol.m⁻³ (accumulate)	X	Species	mol.m⁻³	X
	Na ⁺	0.207 (0.670)	0.465 (0.288)	Na ⁺	0.000	0.000
	Cl ⁻	0.000 (0.670)	0.000 (0.288)	Cl ⁻	0.000	0.000
	NH ₄ ⁺	0.090 (0.660)	0.202 (0.283)	NH ₄ ⁺	0.000	0.000
	SO ₄ ²⁻	0.148 (0.330)	0.333 (0.141)	SO ₄ ²⁻	0.000	0.000
	total	0.445 (2.330)	1.000 (1.000)	total	0.000	0.000
mol.m⁻³	Last solubilized species: Na ₂ SO ₄ = 0.058 ; NH ₄ NaSO ₄ .2H ₂ O=0.090			0		