



Supplement of

Chemical composition-dependent hygroscopic behavior of individual ambient aerosol particles collected at a coastal site

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Particle group	Mole fractions	No.	Size (µm)	С	Ν	0	Na	Mg	Al	Si	S	Cl	Κ	Ca	Cr	Mn	Fe
	$X_{(Na, Mg)Cl}$																
SSAs Cl-rich	0.75	5	4.93	29.7	4.3	17.5	22.1	2.8			1.4	20.9	0.6	0.7			
	0.76	3	3.81	36.4	4.4	19.0	19.5	1.6			0.9	17.3	0.5	0.4			
	0.76	24	4.97	24.7	7.3	18.6	24.0	2.0			1.1	21.2	0.5	0.6			
	0.71	19	4.43	35.5	5.9	17.0	20.3	2.2			0.9	17.6	0.4	0.3			
	0.68	21	4.26	38.3	6.5	19.4	17.6	1.9			1.0	14.5	0.4	0.4			
	0.65	13	4.76	34.6	8.3	18.3	19.5	2.0			1.1	15.2	0.4	0.5			
	0.61	9	3.73	46.1	6.1	21.4	13.3	1.6			0.7	10.1	0.3	0.3			
	0.55	26	3.80	49.3	4.6	23.3	11.9	1.4			0.9	8.1	0.2	0.3			
	0.52	23	4.56	47.4	6.0	23.0	12.3	1.6			0.9	8.0	0.4	0.3			
	0.41	35	3.84	48.0	8.2	25.7	10.4	1.3			0.7	5.3	0.2	0.2			
SSAs Cl-depleted	0.34	1	4.64	44.8	11.0	24.9	11.6	1.5			0.8	4.9	0.2	0.2			
	0.29	31	3.36	49.3	7.5	25.0	11.2	1.5			1.0	4.1	0.2	0.3			
	0.28	33	5.27	46.3	8.0	27.9	11.3	1.4			0.7	4.0	0.2	0.2			
	0.29	22	4.82	50.0	8.1	26.2	9.8	1.2			0.5	3.5	0.3	0.3			
	0.27	39	4.88	45.7	10.2	28.1	10.1	1.2			0.7	3.4	0.3	0.3			
	0.25	30	4.33	47.8	8.4	30.0	8.9	1.1			0.6	2.8	0.2	0.2			
	0.25	10	3.65	50.2	8.6	27.0	9.4	1.0			0.5	2.8	0.2	0.3			
	0.23	11	2.94	50.6	8.3	26.3	9.8	1.2			0.5	2.8	0.3	0.3			
	0.22	12	2.70	43.6	6.7	33.8	10.6	1.2			0.9	2.9	0.2	0.2			
	0.15	16	3.79	38.4	12.8	32.7	11.1	1.2			1.0	2.1	0.3	0.2			
	0.15	0	4.12	43.4	9.8	29.0	11.9	1.0			0.8	2.3	0.2	0.3			
	0.15	18	3.17	40.9	12.5	20.0	12.0	1.5			0.1	1.9	0.2	0.2			
overall average			43.0	7.9	24.0	13.0	1.5			0.8	8.0	0.5	0.3				
	standard deviati	on	2.01	7.2	2.3	4.9	4.6	0.4	1.7	5.1	0.3	6.6	0.1	0.1			
		17	3.01	23.2	8.0	50.7		2.6	1.7	5.1		0.6	0.3	7.8			
с . · ·		20	3.87	20.7	7.4	49.7		0.4	0.2	0.2	0.1	0.7		20.6			
Ca-containing		25	5.84	33.5	4.3	48.5		4.8	0.2	0.3	0.1	0.2		8.2			0.0
		34	2.47	19.8	5.6	50.4		0.4	2.1	4.0	27	0.1		10.8			0.9
		38	3.04	20.9	8./	50.4		0.5	0.4	0.3	3.7	1.1	1.1	14.0		1.6	2.2
	1	4	1.87	23.4	7.0	49.5		1.1	/.4	12.6	0.2	0.2	1.1	0.6		1.6	2.2
	aged	8	3.35	22.8	7.0	48.7		1.1	4.1	8.5		0.3	0.5	5.5			1.4
Alumino-	aged	14	2.98	13.4	3.6	48.6	4.8	5.0	5.0	12.0	0.7	0.7	1.0	3.4			1.9
silicates	aged	28	2.89	14.1	3.8	50.0	0.9	5.2	6.1	12.8	0.4	0.2	1.8	3.2			1.4
		29	2.35	1.5		57.7	6.2		8.7	25.9							
		32	2.38	17.6		44.0		1.4	9.4	19.5			1.1	0.6		2.8	3.7
SiO ₂	aged	37	2.05	9.2	2.9	59.5		0.8	3.1	22.4			0.8	0.9			0.5
Organic and ammonium sulfate		36	2.21	75.1	4.1	10.2					10.7						
Fe-rich		2	1.93	24.7		39.7				0.5		1.1			3.0		31.0
		7	2.50	22.1	0.0	18.7			0.6	11.6					7.2		39.9

Table S1. Elemental concentrations (in at.%) of 37 ambient particles determined by low-*Z* particle EPMA (SSAs #15 and #27 are not included here due to the recombination of the particle after hygroscopic process).

Figure S1. Optical images of the second field (particles #17 - #26) during humidifying (A-F, \uparrow) and dehydration (G-K, \downarrow) processes and the SEI of the same field (L).



Figure S2. Optical images of the third field (particles #27 - #39) during humidifying (A-F, \uparrow) and dehydration (G-K, \downarrow) processes and the SEI of the same field (L).



Figure S3. 2-D area ratio plots of (Na, Mg)(Cl, NO₃) aerosols as a function of RH and secondary electron images (SEIs) of the effloresced ones.



Figure S4. X-Ray elemental maps for C, N, O, Na, Mg, S, Cl, and Ca and secondary electron images (SEIs) of the effloresced ambient SSAs: (a) SSA #5 and (b) SSA #19 with Cl-rich compositions of $X_{(Na,Mg)Cl} = 0.75$ and 0.72, respectively.



Figure S5. X-Ray elemental maps for C, N, O, Na, Mg, S, Cl, and Ca and secondary electron images (SEIs) of the effloresced ambient SSAs: (a) SSA #23 and (b) SSA #11 with equimolar and Cl-depleted compositions of $X_{(Na,Mg)Cl} = 0.52$ and 0.23, respectively.





Figure S6. 2-D area ratio plot, X-ray spectrum, and SEM image with diameter of a reacted Ca-containing particle (particle #20).

Figure S7. 2-D area ratio plot, X-ray spectrum, and SEM image with diameter of an ammonium sulfate aerosol mixed with organics (particle #36).

Figure S8. 2-D area ratio plots, X-ray spectra, and SEM images with diameters of partially reacted Ca-containing particles (particles #17 and #25).

Figure S9. 2-D area ratio plot, X-ray spectrum, and SEM image with diameter of partially aged SiO₂ particle (particle #37).

Figure S10. 2-D area ratio plot, X-ray spectrum, and SEM images with diameters of aluminosilicate particles (particles #29 and #32).

Figure S11. 2-D area ratio plot, X-ray spectrum, and SEM images with diameters of Fe-rich particles (particles #2 and #7).