

Supporting Materials for

**Metal complexation inhibits the effect of oxalic acid in
aerosols as cloud condensation nuclei (CCN)**

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Table S1. The concentration (nmol/m³) of water soluble components in aerosols (a) winter, (b) summer, and (c) winter/summer.

(a) Winter (January 21 to February 12, 2002)									
Size (μm)	Oxalate	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Ca ²⁺	Na ⁺	NH ₄ ⁺	Mg ²⁺	Zn ²⁺
0.25	1.09	2.91	8.16	14.4	0.160	-	160	-	0.18
0.54	2.02	8.07	19.9	43.6	0.312	-	435	-	0.71
0.8	2.58	33.2	58.2	55.9	1.28	-	767	0.150	1.96
1.6	2.12	11.1	20.8	24.4	2.62	8.75	104	0.680	2.58
2.7	0.840	8.60	13.0	9.56	6.71	21.2	14.0	1.14	1.61
4	0.330	10.2	9.04	4.70	5.03	32.5	-	0.830	0.520
5.9	0.370	9.20	8.57	4.56	6.75	33.2	-	0.990	0.320
9	0.201	5.47	4.05	2.69	4.49	23.7	-	0.420	0.130
15	0.0623	1.41	1.03	0.720	1.55	5.65	-	0.122	0.0200
(b) Summer (July 28 to August 13, 2002)									
Size (μm)	Oxalate	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Ca ²⁺	Na ⁺	NH ₄ ⁺	Mg ²⁺	Zn ²⁺
0.25	0.0983	0.280	0.442	22.03	0.0700	-	118	-	0.140
0.54	1.17	1.11	1.28	95.0	0.270	-	475	-	0.460
0.8	1.94	0.603	3.01	146	1.07	-	453	0.740	1.40
1.6	2.78	1.27	5.44	117	2.55	8.18	245	2.30	3.07
2.7	2.60	7.10	22.5	35.3	4.71	77.9	26.1	3.71	2.04
4	1.14	12.5	29.5	12.4	5.84	103	-	3.60	0.730
5.9	0.720	15.7	32.7	10.4	9.52	96.7	-	3.46	0.530
9	0.460	7.76	22.6	6.39	8.61	47.3	-	1.74	0.310
15	0.130	2.08	2.24	3.62	3.18	6.22	-	0.380	0.0600
(c) Winter / summer									
Size (μm)	Oxalate	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Ca ²⁺	Na ⁺	NH ₄ ⁺	Mg ²⁺	Zn ²⁺
0.25	11.1	10.3	18.5	0.654	2.26	-	1.36	-	1.27
0.54	1.73	7.24	15.5	0.460	1.14	-	0.914	-	1.54
0.8	1.33	55.0	19.3	0.380	1.20	-	1.69	0.200	1.40
1.6	0.763	8.73	3.82	0.208	1.03	1.07	0.425	0.300	0.840
2.7	0.323	1.20	0.580	0.271	1.42	0.273	0.535	0.306	0.790
4	0.290	0.813	0.310	0.380	0.861	0.320	-	0.231	0.720
5.9	0.511	0.590	0.262	0.438	0.708	0.343	-	0.286	0.602
9	0.436	0.705	0.180	0.421	0.522	0.500	-	0.244	0.406
15	0.479	0.681	0.460	0.199	0.487	0.908	-	0.330	0.320

Table S2. Correlation coefficient (R^2) between some ions at various particle diameters.

Winter								
	Oxalate	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Ca ²⁺	Na ⁺	NH ₄ ⁺	Mg ²⁺
Oxalate	1.00							
Cl ⁻	0.458	1.00						
NO ₃ ⁻	0.700	0.920	1.00					
SO ₄ ²⁻	0.884	0.565	0.796	1.00				
Ca ²⁺	0.282	8.12×10 ⁻³	0.0971	0.321	1.00			
Na ⁺	0.434	0.0370	0.196	0.434	0.850	1.00		
NH ₄ ⁺	0.695	0.642	0.831	0.921	0.340	0.425	1.00	
Mg ²⁺	0.113	1.27×10 ⁻³	0.0490	0.211	0.884	0.685	0.296	1.00
Zn ²⁺	0.561	0.379	0.445	0.320	1.60×10 ⁻⁵	0.0548	0.168	0.0621

Summer								
	Oxalate	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Ca ²⁺	Na ⁺	NH ₄ ⁺	Mg ²⁺
Oxalate	1.00							
Cl ⁻	0.0140	1.00						
NO ₃ ⁻	4.03×10 ⁻⁴	0.943	1.00					
SO ₄ ²⁻	0.432	0.333	0.287	1.00				
Ca ²⁺	0.0240	0.791	0.812	0.352	1.00			
Na ⁺	2.82×10 ⁻³	0.913	0.951	0.303	0.650	1.00		
NH ₄ ⁺	0.132	0.406	0.418	0.815	0.498	0.427	1.00	
Mg ²⁺	0.183	0.654	0.774	0.0790	0.507	0.814	0.292	1.00
Zn ²⁺	0.899	0.025	6.72×10 ⁻⁴	0.357	0.0160	1.40×10 ⁻⁴	0.0608	0.170

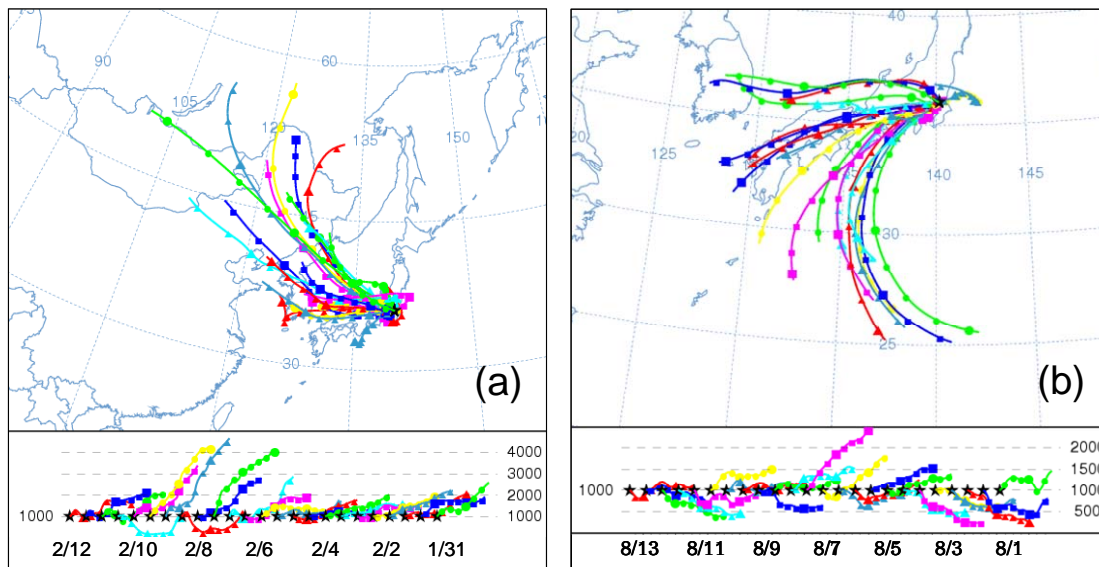


Figure S1. The three dimensional backward trajectory analysis in sampling periods; (a) winter; (b) summer. The NOAA/ARL HYSPLIT model (Draxler and Rolph, 2003) was used for the calculation. The trajectories started at the altitude of 1000 m above the sampling site in Tsukuba.

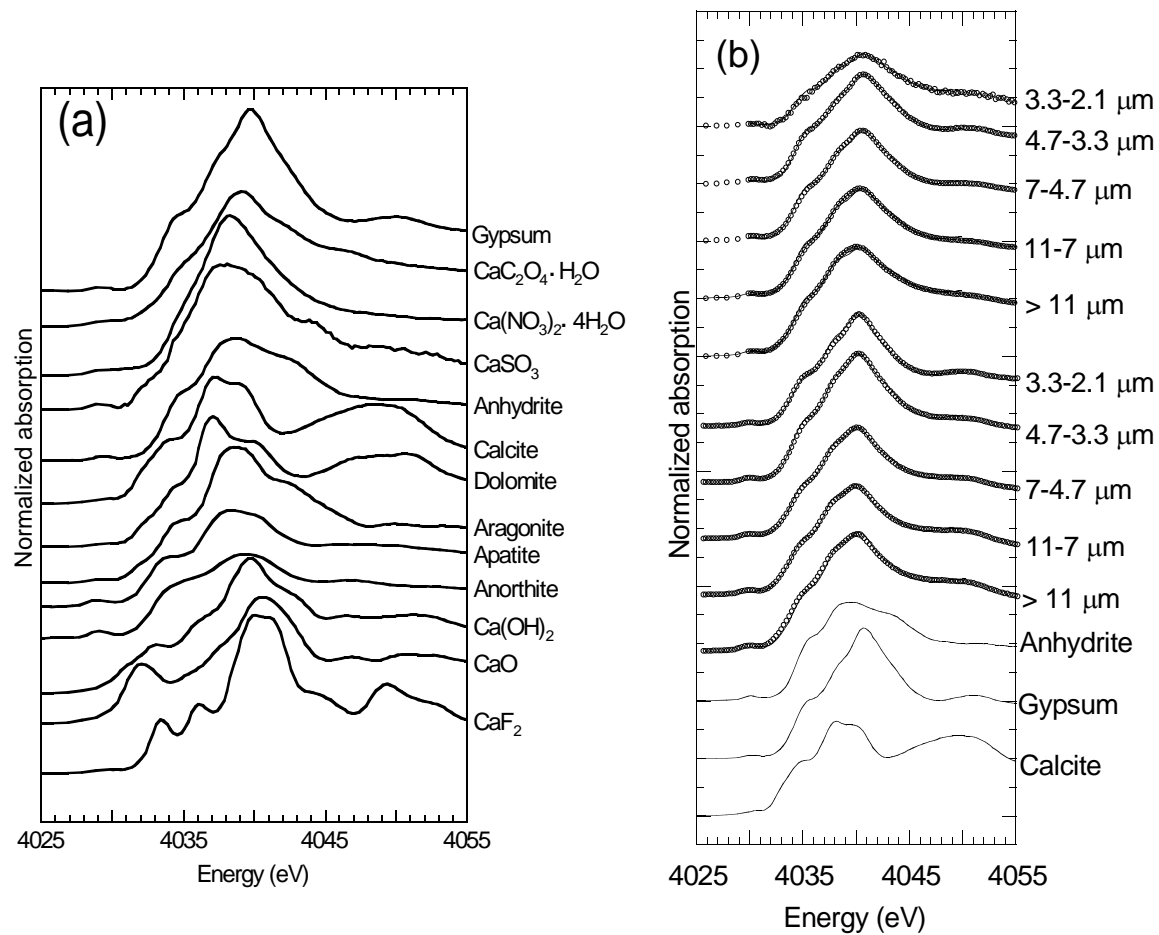


Figure S2. Calcium K-edge XANES; (a) Ca standard materials; (b) coarse particle samples (open circle: samples; lines: fitting) during winter and summer at Tsukuba with standard materials used for fitting.

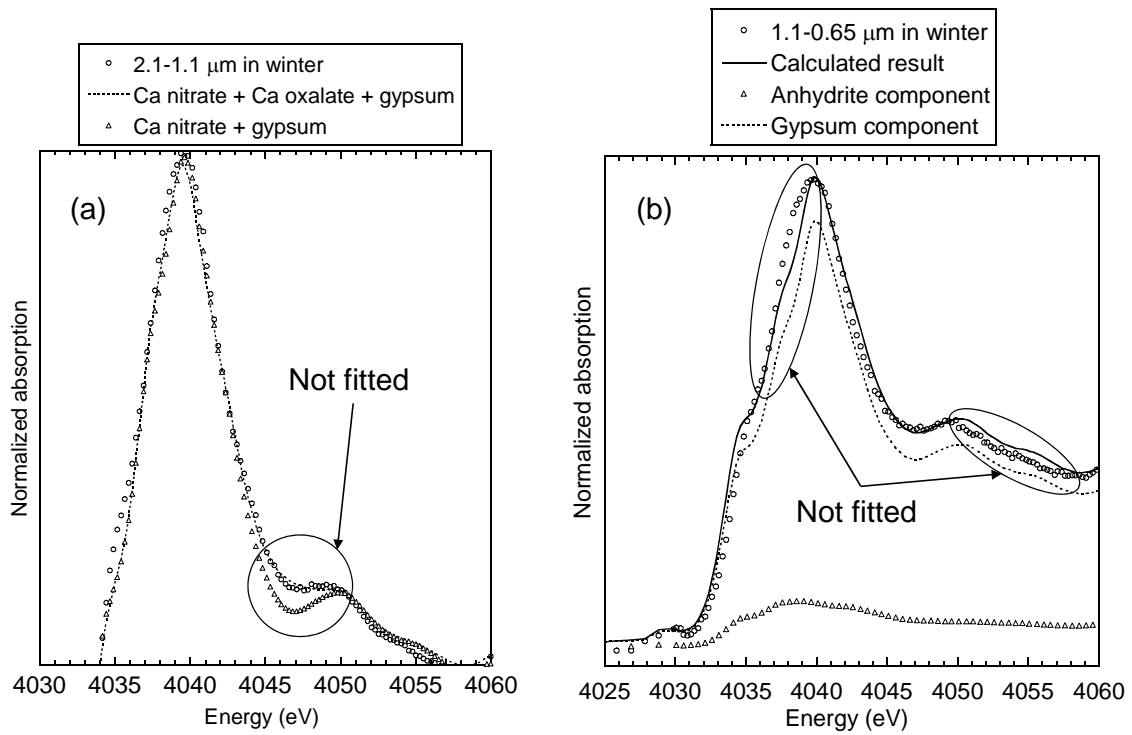


Figure S3. Fitting results of the samples with various standard materials. A circle and ellipses represent the regions of misfit. (a) fitting by Ca nitrate + Ca-oxalate + gypsum (dashed line), Ca nitrate + gypsum (triangle), and 2.1-1.1 μm in winter (circle). (b) fitting result (line) by gypsum (triangle) + anhydrite (triangle) with the spectrum of 1.1-0.65 μm in winter (circle).

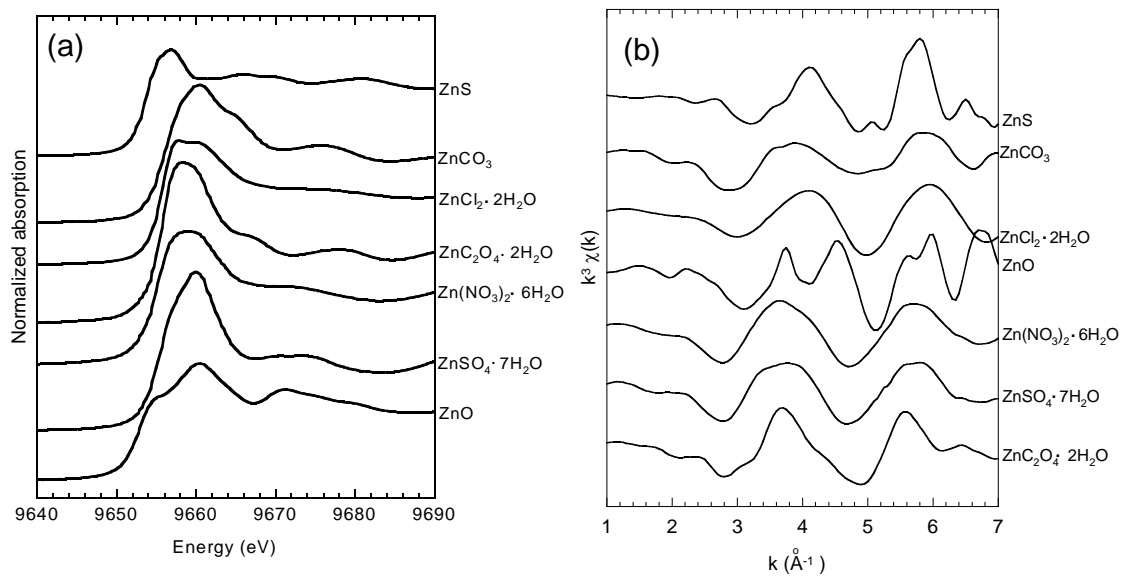


Figure S4. Zinc K-edge XANES (a) and EXAFS (b) for Zn standard materials.

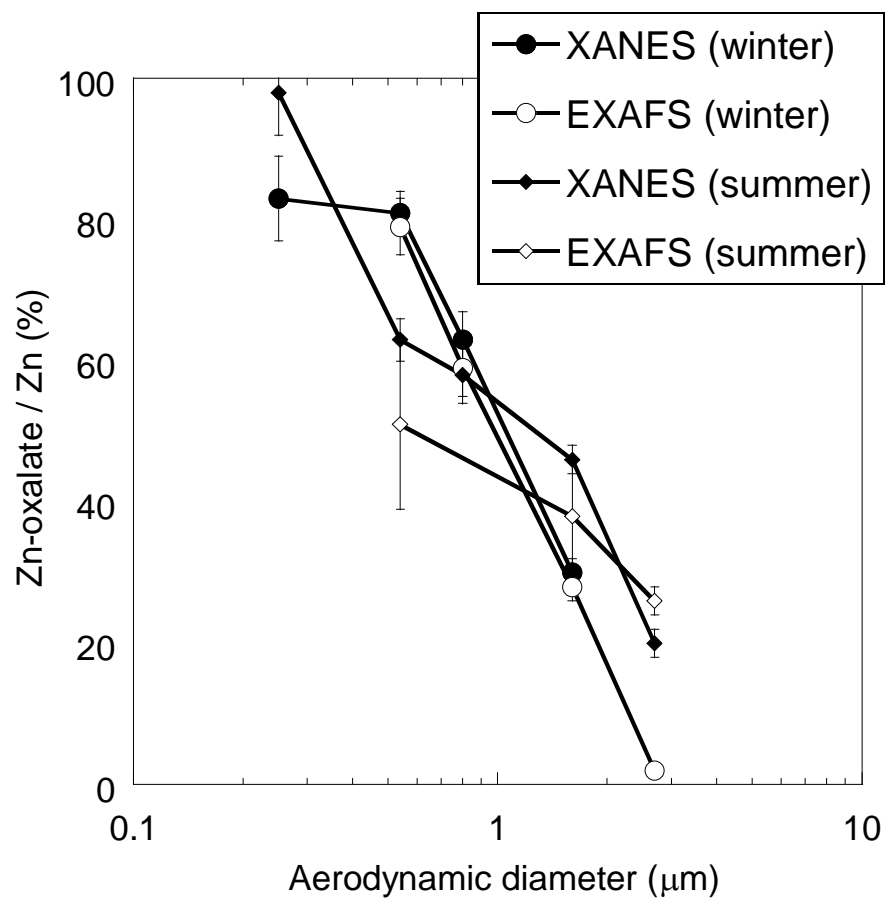


Figure S5. Comparison of the fraction of Zn-oxalate resulting from XANES fitting and EXAFS fitting, which shows the consistency between them.