

Supporting Information for

Stoichiometry of dissolved iron and aluminum as an indicator of the factors controlling the fractional solubility of aerosol iron: results of the annual observations of size-fractionated aerosol particles in Japan

Kohei Sakata^{1*}, Aya Sakaguchi², Yoshiaki Yamakawa³, Chihiro Miyamoto³, Minako Kurisu⁴, Yoshio Takahashi⁴

¹Earth System Division, National Institute for Environmental Studies, 16-2 Onogawa, Tsukuba, Ibaraki 305-8506, Japan

²Faculty of Pure and Applied Science, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Ibaraki 305-8571, Japan

³Graduate School of Science, the University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo, 113-0033, Japan

⁴Research Institute for Marine Resources Utilization, Japan Agency for Marine-Earth Science and Technology, 2-15 Natsuhshima-cho, Yokosuka, Kanagawa 237-0061, Japan

Correspondence to: Kohei Sakata (sakata.kohei@nies.go.jp)

Supplemental Figures

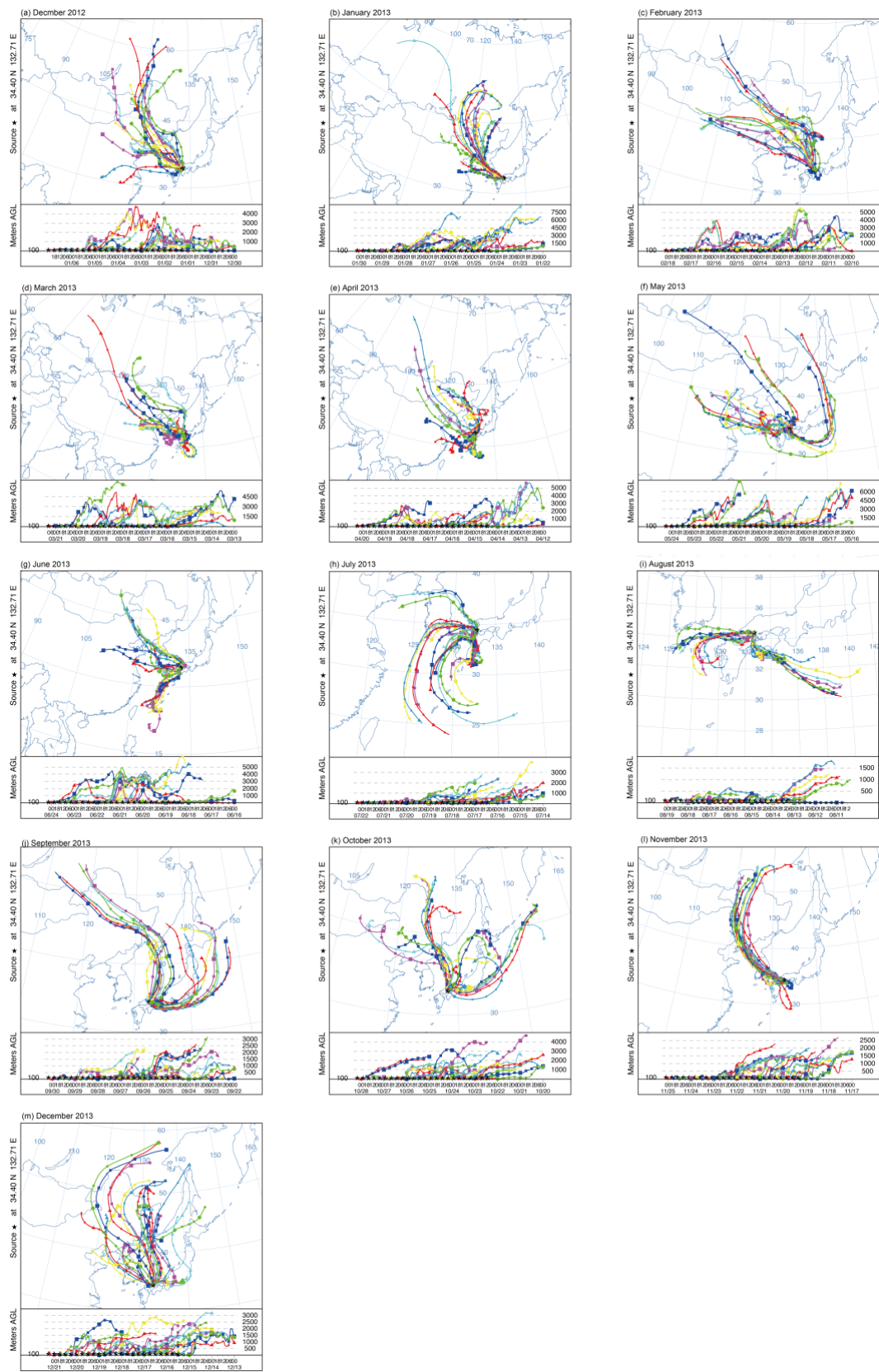


Figure S1: Backward trajectories of sampling periods of (a) December 2012, (b-m) January to December 2013.

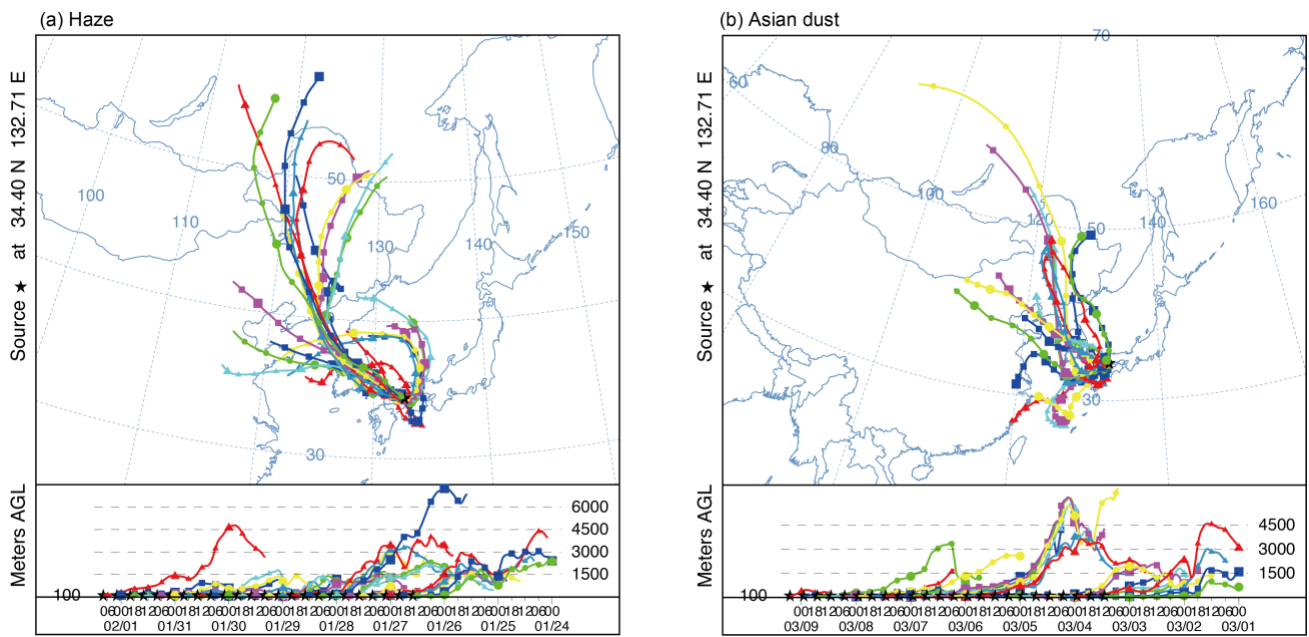


Figure S2: Backward trajectories of sampling periods of (a) Haze and (b) Asian dust events.

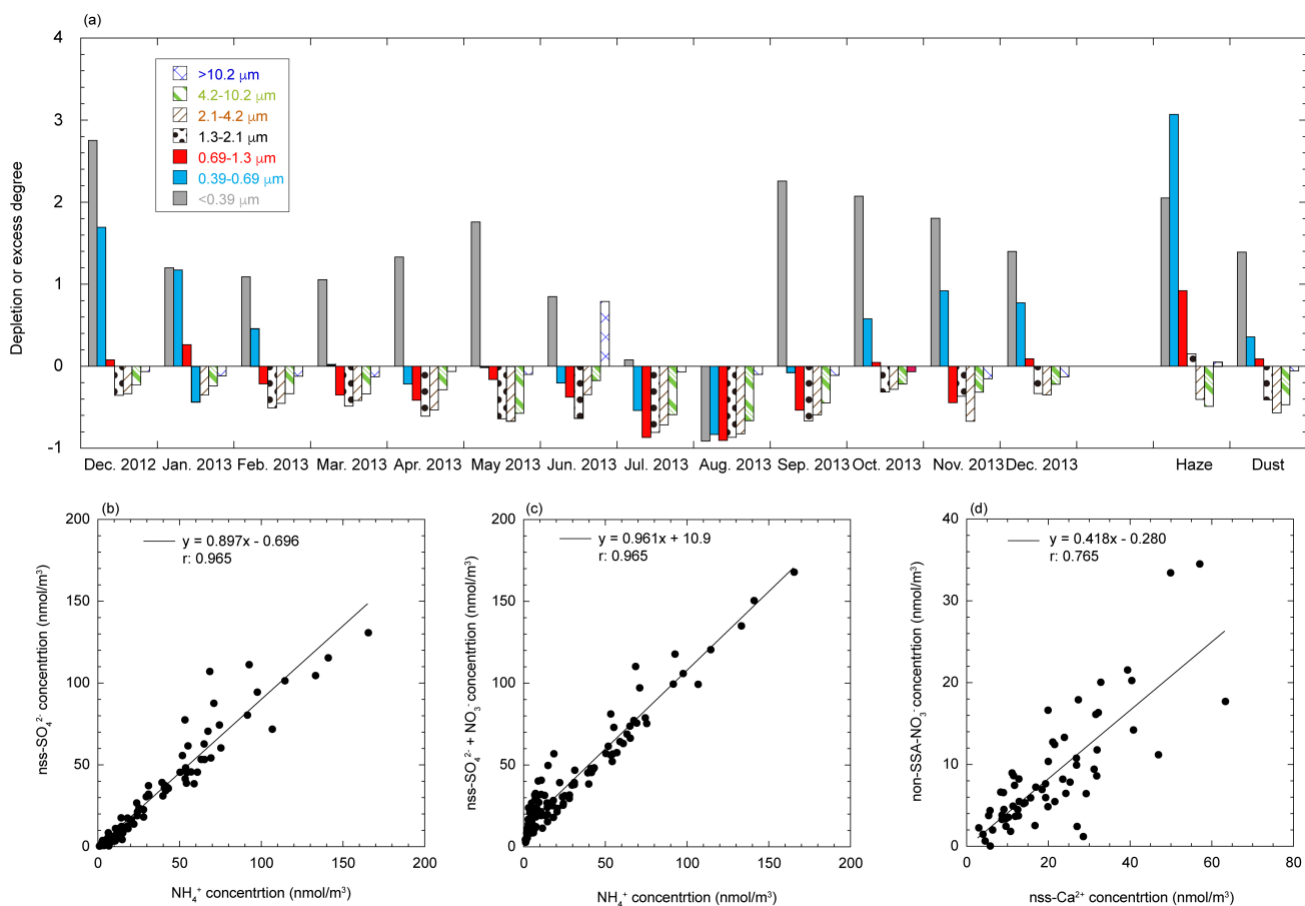


Figure S3: (a) depletion and excess degree of Cl⁻ in aerosol particles. Depletion and excess degree was calculated

by $\frac{(Cl^-/Na^+)_{aerosol}}{(Cl^-/Na^+)_{seawater}} - 1$. (b and c) scatter plots of [NH₄⁺] concentration with [nss-SO₄²⁻] and [nss-SO₄²⁻]+[NO₃⁻]. (d) A scatter plot between [nss-Ca²⁺] and [non-SSA-NO₃⁻].

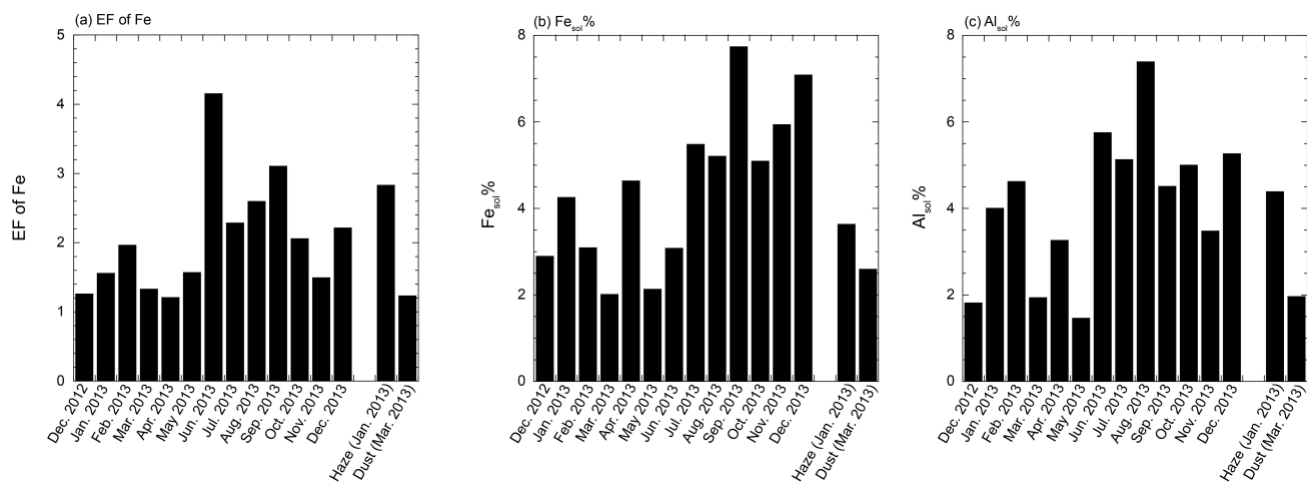


Figure S4: Monthly variations of (a) EF of Fe, (b) Fe_{sol}%, and (c) Al_{sol}% in TSP.

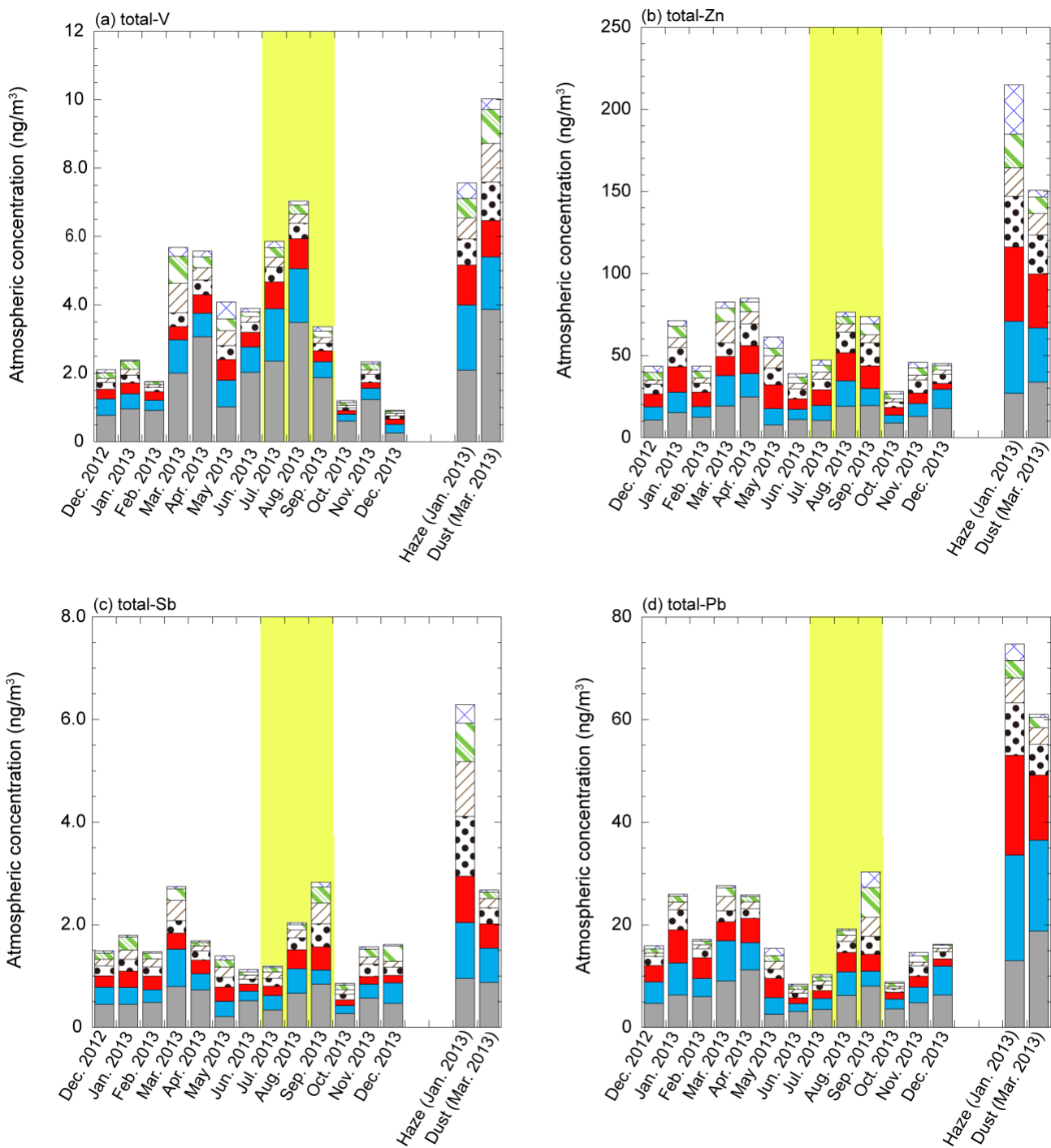


Figure S5: Monthly variation of (a) V, (b) Zn, (c) Sb, and (d) Pb concentrations in size-fractionated aerosol particles. Yellow shaded region shows the sampling period when air mass was derived from the Seto Inland Sea.

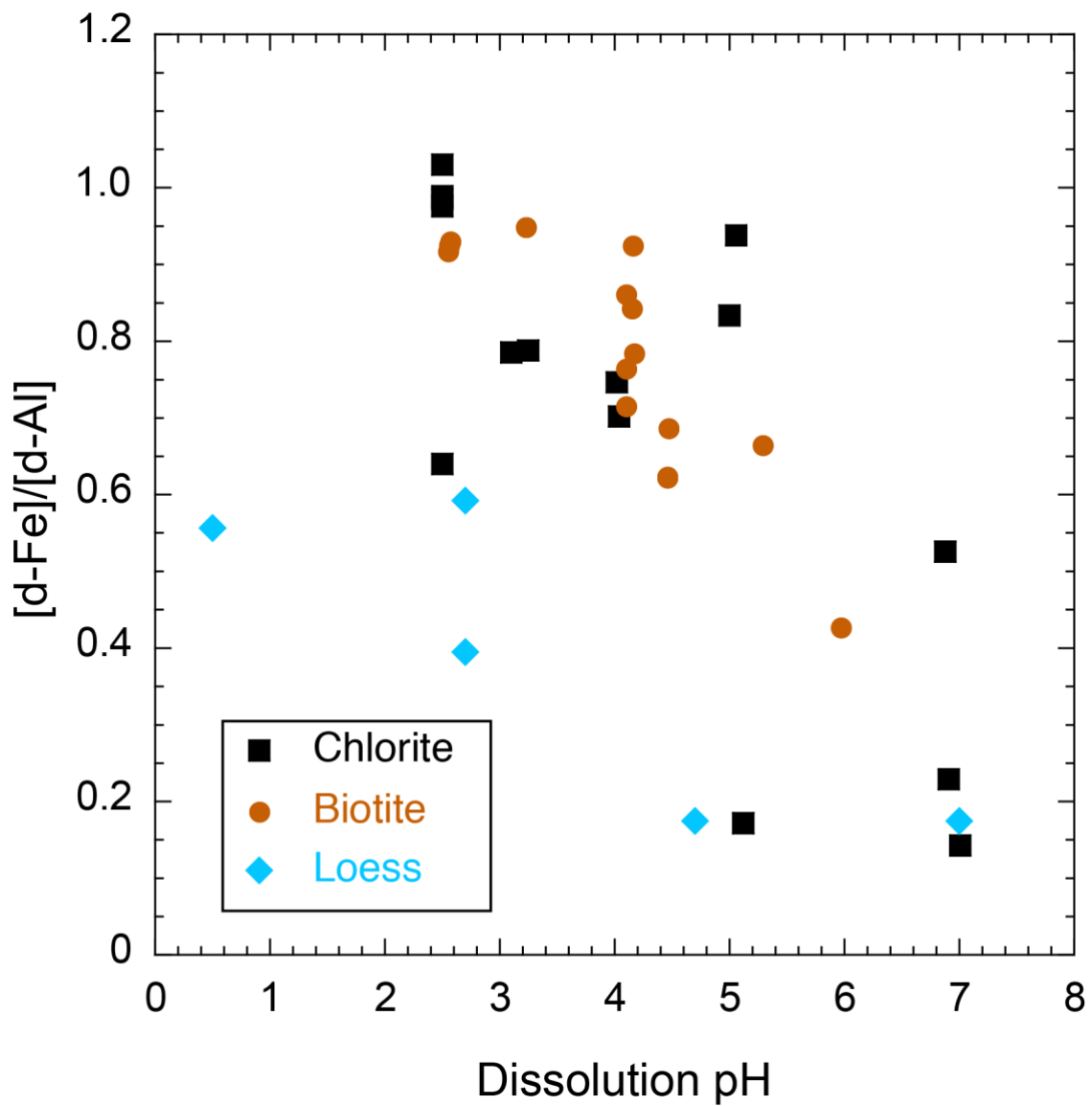


Figure S6: pH dependence of [d-Fe]/[d-Al] ratio of aluminosilicate and loess during proton-promoted dissolution. These [d-Fe]/[d-Al] ratios were referred from Kodama and Schnitzer (1973), Desboeufs et al. (2001), Lowson et al. (2005), and Bray et al. (2015).

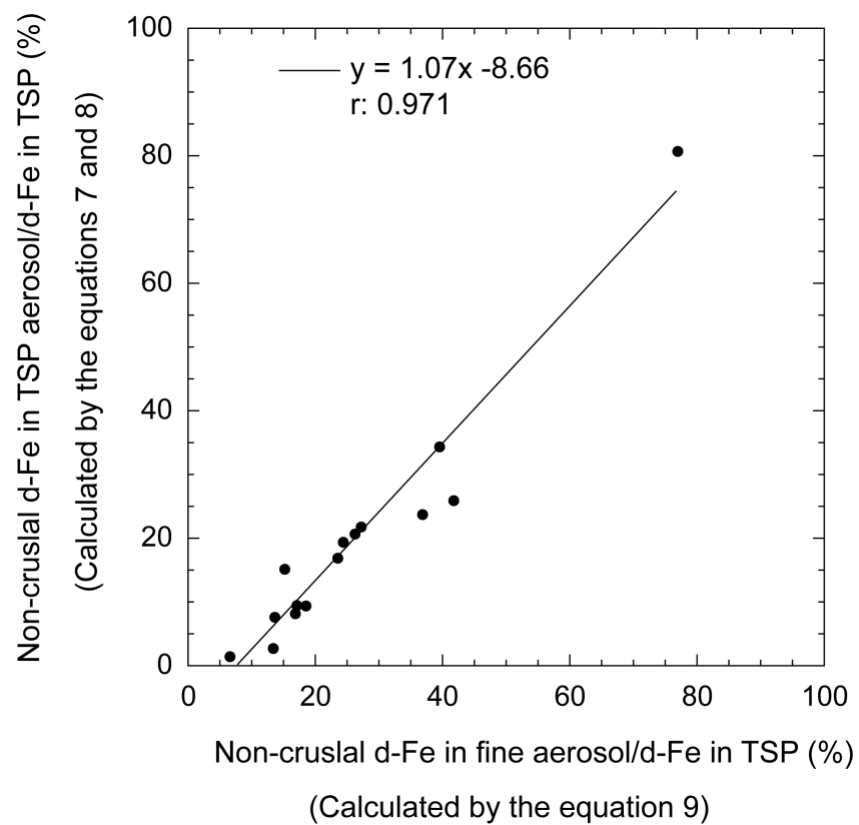


Figure S7: Comparison of the calculation results of relative abundance on non-crystal

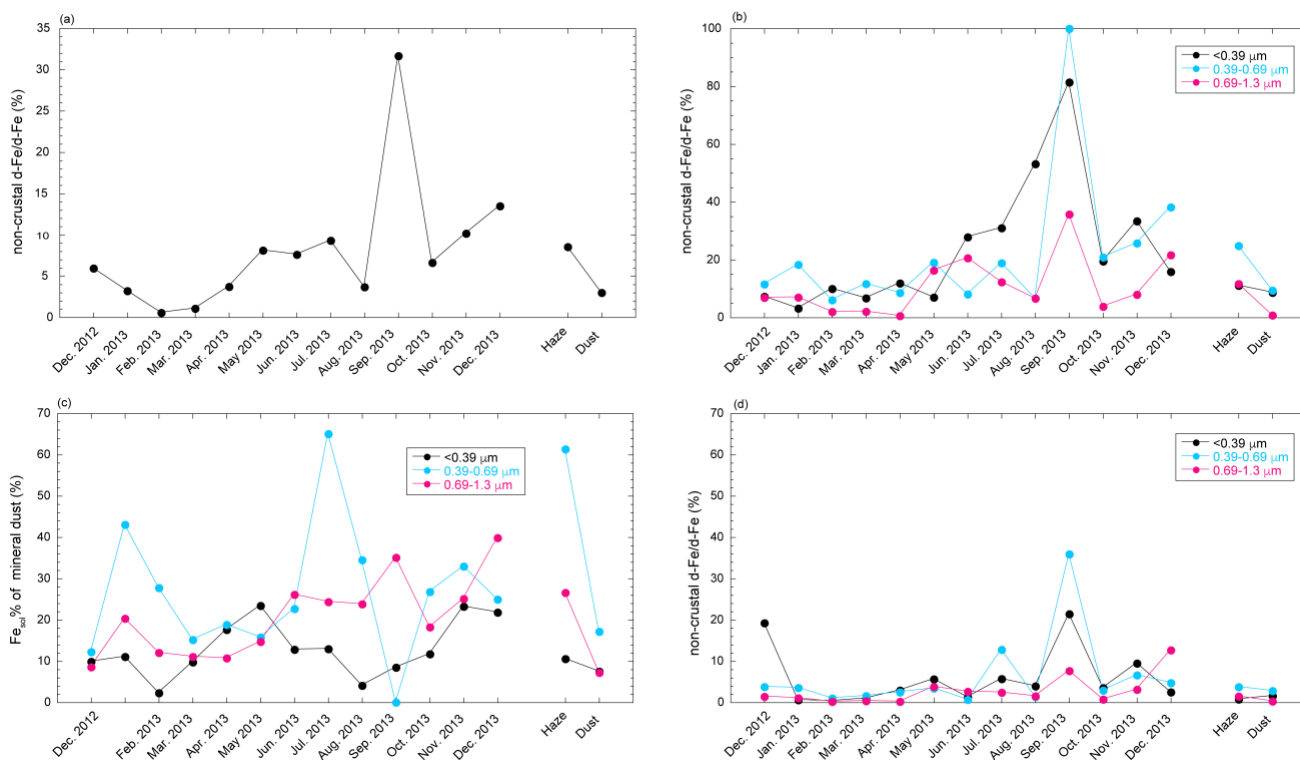


Figure S8: Relative abundance of pyrogenic Fe to d-Fe in (a) TSP and (b) fine aerosol particles when [d-Fe]/[d-Al] ratio of non-crystal Fe is 4.67. (c) crustal Fe and (d) pyrogenic Fe when [d-Fe]/[d-Al] ratio of pyrogenic Fe.

Table S1 Sampling information for each sapling period.

Sample name	Sampling period		Total flow m ³
	Start	End	
	Non-dust event		
December 2012	11:30, 25 Dec. 2012	9:20, 7 Jan. 2013	11699.5
January 2013	10:40, 21 Jan. 2013	13:29, 30 Jan. 2013	7624.5
February 2013	10:31, 4 Feb. 2013	10:52, 18 Feb. 2013	11740.6
March 2013	17:20, 21 Mar. 2013	17:20, 21 Mar. 2013	9850.1
April 2013	11:38, 8 Apr. 2013	14:30, 20 Apr. 2013	9494.7
May 2013	13:45, 13 May, 2013	14:52, 24 May, 2013	9282.3
June 2013	15:00, 11 Jun. 2013	12:16, 24 Jun. 2013	11477.6
July 2013	14:25, 8 Jul. 2013	13:00, 22 Jul. 2013	11461.2
August 2013	11:45, 6 Aug. 2013	11:25, 19 Aug. 2013	11490.1
September 2013	16:25, 17 Sep. 2013	12:40, 30 Sep. 2013	11386.3
October 2013	14:40, 15 Oct. 2013	13:10, 28 Oct. 2013	11544.3
November 2013	10:45, 12 Nov. 2013	11:05, 25 Nov. 2013	11698.3
December 2013	9:30, 7 Dec. 2013	14:40, 21 Dec. 2013	11838.8
	Dust events		
Haze	14:19, 31 Jan. 2013	17:09, 1 Feb. 2013	936.8
Dust	10:30, 4 Mar. 2013	14:30, 9 Mar. 2013	4840.2

Table S2. Major ion concentration (neq/m³) in size-fractionated aerosol samples.

	Diameter μm	Na ⁺ neq/m ³	NH ₄ ⁺ neq/m ³	K ⁺ neq/m ³	Mg ²⁺ neq/m ³	Ca ²⁺ neq/m ³	Cl ⁻ neq/m ³	NO ₃ neq/m ³	SO ₄ ²⁻ neq/m ³
Dec., 2012	<0.39	0.534	41.1	0.962	0.158	0.695	2.35	10.4	37.6
	0.39-0.69	0.433	24.1	0.519	0.126	0.347	1.36	6.42	19.2
	0.69-1.3	1.12	20.4	0.497	0.316	0.651	1.41	6.72	14.9
	1.3-2.1	2.74	5.07	0.176	0.656	1.24	2.07	4.96	3.66
	2.1-4.2	5.26	3.39	0.142	1.14	2.47	4.10	6.59	2.52
	4.2-10.2	10.2	1.75	0.179	1.79	3.86	9.18	6.03	2.59
	>10.2	8.11	1.38	0.120	1.21	1.91	8.87	2.11	1.87
Jan., 2013	<0.39	1.94	54.1	1.55	0.908	3.69	4.98	13.1	39.2
	0.39-0.69	1.00	56.9	1.29	0.304	0.593	2.55	11.7	46.0
	0.69-1.3	2.19	53.4	1.33	0.682	1.09	3.24	14.2	42.0
	1.3-2.1	7.85	28.0	0.630	1.66	2.51	5.18	12.6	19.2
	2.1-4.2	11.2	10.6	0.281	2.45	3.57	8.52	12.3	6.98
	4.2-10.2	13.3	5.41	0.211	2.54	3.26	11.8	9.26	3.57
	>10.2	21.3	3.43	0.292	3.56	3.97	21.9	5.43	4.15
Feb., 2013	<0.39	1.04	65.2	1.53	0.384	1.99	2.53	13.0	53.6
	0.39-0.69	1.15	50.2	1.79	0.405	0.538	1.97	11.4	45.8
	0.69-1.3	2.65	43.1	1.63	0.886	0.979	2.43	12.3	36.1
	1.3-2.1	7.54	17.6	0.669	1.99	2.22	4.35	12.3	12.9
	2.1-4.2	11.9	7.43	0.324	2.79	3.57	7.61	13.2	5.19
	4.2-10.2	14.2	4.28	0.298	3.05	3.37	11.1	10.9	3.36
	>10.2	11.5	2.00	0.182	2.05	2.74	11.8	3.65	2.71
Mar., 2013	<0.39	2.11	75.2	1.75	0.621	2.73	5.07	14.8	60.7
	0.39-0.69	2.15	64.9	1.83	0.656	1.52	2.59	10.7	63.2
	0.69-1.3	4.14	40.7	1.12	1.51	3.12	3.14	10.2	36.6
	1.3-2.1	7.68	14.4	0.496	2.33	4.30	4.63	10.9	13.4
	2.1-4.2	25.1	10.8	1.62	7.22	17.3	17.1	26.5	14.3
	4.2-10.2	29.9	8.88	0.728	8.00	17.8	23.1	32.1	8.27
	>10.2	23.5	2.67	0.425	5.20	12.5	23.9	10.8	6.11
Apr., 2013	<0.39	2.17	114	2.88	1.46	7.16	5.92	18.9	102
	0.39-0.69	1.56	53.6	1.46	0.711	2.00	1.43	7.80	48.5
	0.69-1.3	3.28	42.6	1.49	1.75	4.36	2.25	11.1	36.7
	1.3-2.1	8.88	15.2	0.550	3.16	7.23	4.07	17.1	9.72
	2.1-4.2	16.3	10.6	0.578	5.41	12.4	8.91	26.5	5.64
	4.2-10.2	23.0	7.17	0.682	7.09	20.2	19.2	27.7	5.08
	>10.2	13.0	4.48	0.361	3.98	10.3	14.3	8.43	3.29
May, 2013	<0.39	1.44	91.5	2.11	1.01	4.45	4.65	18.9	80.7
	0.39-0.69	1.95	71.0	2.28	0.623	1.52	2.24	9.28	88.0
	0.69-1.3	1.84	55.1	1.33	1.04	2.91	1.81	11.2	61.9
	1.3-2.1	4.76	31.3	0.893	2.38	6.09	1.99	14.8	32.1
	2.1-4.2	7.12	12.8	0.556	3.17	9.82	2.76	21.7	9.88
	4.2-10.2	6.97	9.16	0.403	3.04	10.8	3.47	22.6	4.70
	>10.2	3.87	4.80	0.316	2.67	15.1	4.08	17.1	2.78
Jun., 2013	<0.39	0.616	41.4	0.854	0.499	3.27	1.33	11.1	34.5
	0.39-0.69	0.627	30.9	0.838	0.244	0.487	0.583	5.85	32.3
	0.69-1.3	0.961	25.1	0.630	0.439	0.954	0.702	7.49	23.9
	1.3-2.1	2.23	13.0	0.458	0.873	2.01	0.954	7.63	11.7
	2.1-4.2	2.54	6.58	0.534	0.995	2.87	1.95	9.28	3.48
	4.2-10.2	2.18	5.20	0.367	0.974	3.88	2.10	9.11	2.37
	>10.2	0.231	1.11	0.112	0.267	1.78	0.483	2.07	0.894

Table S2. Continued.

	Diameter μm	Na ⁺ neq/m ³	NH ₄ ⁺ neq/m ³	K ⁺ neq/m ³	Mg ²⁺ neq/m ³	Ca ²⁺ neq/m ³	Cl ⁻ neq/m ³	NO ₃ neq/m ³	SO ₄ ²⁻ neq/m ³
Jul., 2013	<0.39	0.847	67.3	0.843	0.459	3.70	1.07	6.57	70.8
	0.39-0.69	0.731	30.9	0.482	0.165	0.428	0.394	1.95	37.5
	0.69-1.3	3.53	23.7	0.549	0.859	1.51	0.546	3.35	27.3
	1.3-2.1	12.8	6.35	0.564	2.72	2.79	2.88	10.3	10.2
	2.1-4.2	21.3	2.83	0.945	4.43	4.64	7.08	17.4	6.52
	4.2-10.2	15.3	2.75	0.593	3.14	4.56	7.30	13.1	3.59
Aug., 2013	>10.2	2.23	1.72	0.198	0.596	3.63	2.43	3.96	1.42
	<0.39	0.969	92.6	2.13	0.234	2.24	0.0994	6.43	112
	0.39-0.69	1.11	68.5	2.10	0.268	0.595	0.218	2.97	107
	0.69-1.3	2.35	53.2	1.86	0.693	1.50	0.262	3.41	77.9
	1.3-2.1	5.97	14.2	0.738	1.75	3.40	0.934	8.50	18.5
	2.1-4.2	8.92	6.08	0.663	2.55	6.13	1.85	16.8	6.70
Sep., 2013	4.2-10.2	6.82	5.34	0.497	2.11	8.24	2.68	17.8	3.75
	>10.2	1.22	2.71	0.318	0.841	5.95	1.28	6.78	1.53
	<0.39	0.985	63.1	2.27	0.285	1.55	3.75	15.4	53.6
	0.39-0.69	1.16	39.1	1.60	0.295	0.678	1.26	5.85	39.5
	0.69-1.3	2.62	20.5	0.689	0.663	1.13	1.43	6.61	17.0
	1.3-2.1	10.4	10.4	0.608	2.27	2.33	4.06	13.6	7.931
Oct., 2013	2.1-4.2	22.8	8.16	0.857	5.01	5.14	10.9	25.2	5.94
	4.2-10.2	30.2	6.67	0.916	6.26	6.03	19.5	27.6	4.26
	>10.2	29.8	3.70	0.773	6.78	10.8	31.1	16.6	4.45
	<0.39	0.871	27.9	1.58	0.434	1.90	3.13	6.84	22.9
	0.39-0.69	0.628	14.5	0.759	0.213	0.392	1.16	2.95	12.5
	0.69-1.3	1.64	11.4	0.262	0.377	0.410	2.01	3.94	7.61
Nov., 2013	1.3-2.1	6.07	5.54	0.230	1.06	0.846	4.87	5.62	3.95
	2.1-4.2	16.2	3.45	0.443	2.98	1.86	13.6	9.10	3.54
	4.2-10.2	19.2	3.08	0.472	3.65	2.00	17.6	8.54	3.36
	>10.2	17.3	0.918	0.266	2.77	1.24	18.9	2.00	2.52
	<0.39	1.08	54.4	1.88	0.393	1.04	3.55	11.3	45.6
	0.39-0.69	0.716	29.5	1.18	0.254	0.545	1.61	7.04	30.8
Dec., 2013	0.69-1.3	2.96	5.91	0.270	1.13	1.96	1.93	5.07	3.70
	1.3-2.1	3.43	6.78	0.314	1.30	2.27	2.54	6.40	4.93
	2.1-4.2	7.54	4.88	0.299	2.32	5.14	2.90	11.9	3.80
	4.2-10.2	14.9	2.65	0.353	3.53	6.14	12.0	10.8	3.15
	>10.2	39.7	4.62	1.04	9.28	9.51	39.3	12.0	7.91
	<0.39	1.06	39.7	1.37	0.414	1.54	2.97	7.24	31.3
Haze	0.39-0.69	0.865	24.2	1.25	0.253	0.411	1.79	4.99	21.6
	0.69-1.3	1.87	18.2	0.993	0.631	0.831	2.39	5.04	13.1
	1.3-2.1	5.23	7.45	0.385	1.21	1.02	4.09	6.54	5.93
	2.1-4.2	9.47	4.19	0.235	2.00	1.61	7.19	7.47	2.70
	4.2-10.2	19.9	2.44	0.418	4.17	3.48	18.1	6.99	3.74
	>10.2	17.0	1.91	0.316	3.29	2.22	17.3	2.62	3.15
Asian dust	<0.39	1.82	107	2.07	0.601	1.93	6.50	27.3	72.2
	0.39-0.69	1.34	165	3.06	0.602	1.52	6.39	36.9	131
	0.69-1.3	2.41	133	1.97	0.991	2.99	5.42	30.1	105
	1.3-2.1	2.43	60.9	1.00	1.20	3.77	3.27	17.3	46.0
	2.1-4.2	2.78	14.5	0.257	1.00	3.70	1.94	11.7	8.24
	4.2-10.2	7.76	7.93	0.277	1.77	8.49	4.64	17.8	6.14
Asian dust	>10.2	3.38	4.91	<D.L.	0.691	4.76	4.15	6.40	2.59
	<0.39	2.07	141	3.71	1.20	5.17	5.78	34.8	116
	0.39-0.69	1.71	69.1	2.03	0.825	2.43	2.71	21.2	54.6
	0.69-1.3	2.99	58.8	1.70	1.63	4.55	3.82	25.4	39.0
	1.3-2.1	6.39	22.0	0.862	3.40	11.8	4.42	24.6	14.7
	2.1-4.2	14.3	18.4	0.673	5.96	30.5	7.23	44.1	13.0
Asian dust	4.2-10.2	17.8	14.8	0.663	5.47	28.4	11.0	43.2	6.66
	>10.2	2.46	2.22	0.121	0.907	4.69	2.71	4.57	1.14

<D.L.: Below detection limits

Table S3 Total and dissolved Fe and Al concentrations and their fractional solubilities and [d-Fe]/[d-Al] ratio in size-fractionated aerosol particles.

	Diameter μm	Total Al ng/m^3	d-Al ng/m^3	Al _{sol} % %	Total Fe ng/m^3	d-Fe ng/m^3	Fe _{sol} % %	[d-Fe]/[d-Al]
Dec., 2012	<0.39	56.8	2.78	4.90	40.4	4.14	10.3	0.719
	0.39-0.69	22.6	1.15	5.08	21.9	2.13	9.74	0.901
	0.69-1.3	39.0	1.69	4.34	38.6	2.46	6.39	0.703
	1.3-2.1	77.7	1.53	1.98	63.5	1.49	2.35	0.469
	2.1-4.2	68.2	0.902	1.32	61.0	0.914	1.50	0.489
	4.2-10.2	122	0.125	0.102	101.8	0.108	0.106	0.416
	>10.2	71.2	0.117	0.165	66.8	0.120	0.179	0.492
Jan., 2013	<0.39	82.1	5.71	6.95	90.1	6.43	7.14	0.544
	0.39-0.69	17.6	2.58	14.7	44.6	6.34	14.2	1.19
	0.69-1.3	34.2	3.49	10.2	56.5	5.11	9.06	0.708
	1.3-2.1	88.7	3.88	4.37	82.7	3.69	4.46	0.460
	2.1-4.2	99.0	4.14	4.18	99.0	1.72	1.74	0.201
	4.2-10.2	159	1.30	0.821	152.3	0.717	0.470	0.266
	>10.2	65.6	0.762	1.16	56.1	0.651	1.16	0.413
Feb., 2013	<0.39	67.6	0.692	1.02	79.5	1.20	1.50	0.834
	0.39-0.69	15.0	2.20	14.7	28.2	3.04	10.8	0.667
	0.69-1.3	34.1	2.81	8.23	51.3	2.87	5.60	0.495
	1.3-2.1	46.7	3.81	8.15	60.0	3.07	5.13	0.390
	2.1-4.2	59.7	2.93	4.91	80.5	1.95	2.42	0.321
	4.2-10.2	61.8	1.39	2.25	88.2	0.723	0.819	0.251
	>10.2	36.5	1.02	2.80	44.1	0.453	1.03	0.214
Mar., 2013	<0.39	140	6.96	4.98	163	10.0	6.14	0.695
	0.39-0.69	66.6	4.18	6.28	102	7.83	7.66	0.906
	0.69-1.3	93.4	7.01	7.51	104	7.23	6.95	0.498
	1.3-2.1	544	5.47	1.01	427	3.20	0.749	0.282
	2.1-4.2	475	8.20	1.73	398	2.47	0.622	0.146
	4.2-10.2	222	1.00	0.45	202	0.424	0.210	0.204
	>10.2	175	0.415	0.24	165	0.104	0.0632	0.121
Apr., 2013	<0.39	121	8.76	7.23	149	16.6	11.10	0.915
	0.39-0.69	50.9	4.48	8.80	59.4	7.18	12.09	0.775
	0.69-1.3	103	8.48	8.22	92.4	7.63	8.25	0.435
	1.3-2.1	197	5.25	2.67	60.9	3.30	5.42	0.304
	2.1-4.2	177	3.27	1.85	158	1.70	1.07	0.251
	4.2-10.2	203	0.77	0.38	183	0.379	0.207	0.237
	>10.2	116	0.537	0.46	96.7	0.257	0.266	0.231
May, 2013	<0.39	26.0	3.06	11.8	23.3	4.48	19.2	0.707
	0.39-0.69	70.9	3.75	5.28	101	9.46	9.39	1.22
	0.69-1.3	73.2	3.86	5.27	87.8	8.82	10.0	1.10
	1.3-2.1	121	5.08	4.21	125.5	7.21	5.74	0.685
	2.1-4.2	209	4.63	2.22	221	2.25	1.02	0.235
	4.2-10.2	110	0.415	0.38	113	0.251	0.222	0.292
	>10.2	819	0.0548	0.01	863	0.0901	0.0104	0.794
Jun., 2013	<0.39	23.8	0.88	3.69	78.8	2.90	3.68	1.60
	0.39-0.69	37.3	4.04	10.8	98.0	6.30	6.43	0.753
	0.69-1.3	22.0	1.86	8.46	53.6	4.96	9.25	1.29
	1.3-2.1	26.6	2.37	8.91	74.6	3.87	5.19	0.791
	2.1-4.2	61.9	2.10	3.40	184	1.83	1.00	0.421
	4.2-10.2	23.9	1.67	6.97	80.4	1.59	1.97	0.460
	>10.2	68.8	2.28	3.32	181	1.59	0.878	0.337

Table S3 Continued.

	Diameter μm	Total Al ng/m^3	d-Al ng/m^3	Al _{sol} % %	Total Fe ng/m^3	d-Fe ng/m^3	Fe _{sol} % %	[d-Fe]/[d-Al]
Jul., 2013	<0.39	74.5	2.68	3.59	103	9.58	9.32	1.73
	0.39-0.69	32.0	7.00	21.9	47.8	17.6	36.8	1.21
	0.69-1.3	56.4	5.55	9.85	92.6	10.7	11.6	0.933
	1.3-2.1	67.5	5.64	8.35	116	6.85	5.89	0.587
	2.1-4.2	95.2	4.08	4.28	160	2.47	1.54	0.292
	4.2-10.2	122	3.99	3.26	226	1.82	0.806	0.221
	>10.2	132	0.809	0.61	160	0.541	0.338	0.323
Aug., 2013	<0.39	25.9	1.01	3.90	65.7	5.60	8.53	2.67
	0.39-0.69	14.8	6.21	41.9	33.4	8.90	26.6	0.693
	0.69-1.3	142	7.74	5.46	235	11.1	4.71	0.691
	1.3-2.1	74.3	14.4	19.33	103	13.9	13.5	0.466
	2.1-4.2	83.1	4.56	5.49	132	4.40	3.33	0.466
	4.2-10.2	149	4.11	2.75	227	2.14	0.941	0.251
	>10.2	60.1	2.60	4.32	117	1.54	1.31	0.286
Sep., 2013	<0.39	86.8	3.37	3.89	162	27.1	16.7	3.88
	0.39-0.69	15.3	1.42	9.31	48.7	13.8	28.2	4.67
	0.69-1.3	28.5	2.67	9.35	69.3	10.7	15.4	1.93
	1.3-2.1	40.1	3.48	8.68	94.8	6.68	7.05	0.927
	2.1-4.2	55.4	4.17	7.53	125	5.08	4.08	0.588
	4.2-10.2	102	2.48	2.44	199	1.52	0.764	0.296
	>10.2	67.9	0.262	0.39	143	0.210	0.147	0.387
Oct., 2013	<0.39	43.5	1.69	3.89	53.6	4.34	8.10	1.24
	0.39-0.69	7.01	0.605	8.63	16.3	1.62	9.94	1.30
	0.69-1.3	17.9	1.97	11.0	23.8	2.33	9.79	0.571
	1.3-2.1	23.5	1.29	5.50	29.7	1.59	5.35	0.594
	2.1-4.2	29.6	1.14	3.83	39.5	1.22	3.09	0.520
	4.2-10.2	40.8	1.91	4.68	59.7	1.41	2.37	0.357
	>10.2	19.7	0.498	2.53	33.5	0.505	1.51	0.490
Nov., 2013	<0.39	31.4	1.98	6.30	47.9	7.51	15.7	1.83
	0.39-0.69	14.1	1.38	9.75	26.3	4.30	16.3	1.51
	0.69-1.3	24.9	3.01	12.1	28.9	4.65	16.1	0.748
	1.3-2.1	102	2.61	2.57	95.8	2.81	2.94	0.520
	2.1-4.2	64.0	2.40	3.75	56.7	2.17	3.83	0.437
	4.2-10.2	105	1.37	1.31	93.1	1.11	1.19	0.392
	>10.2	36.7	0.394	1.07	37.5	0.34	0.916	0.422
Dec., 2013	<0.39	34.6	2.73	7.90	62.5	6.14	9.82	1.09
	0.39-0.69	11.0	0.72	6.56	31.8	3.03	9.53	2.04
	0.69-1.3	11.5	1.46	12.6	14.8	4.02	27.2	1.33
	1.3-2.1	24.5	1.71	6.97	35.4	2.76	7.80	0.782
	2.1-4.2	31.9	1.17	3.66	45.2	1.27	2.80	0.523
	4.2-10.2	42.2	0.879	2.08	52.3	0.742	1.42	0.408
	>10.2	14.7	0.303	2.07	16.0	0.294	1.84	0.469
Haze	<0.39	84.3	3.76	4.46	155	6.86	4.42	0.881
	0.39-0.69	45.5	8.37	18.4	199	25.4	12.8	1.47
	0.69-1.3	93.4	10.3	11.0	215	19.3	8.96	0.906
	1.3-2.1	160	10.7	6.68	299	13.1	4.37	0.589
	2.1-4.2	208	6.47	3.11	393	6.61	1.68	0.494
	4.2-10.2	278	8.59	3.09	536	7.62	1.42	0.429
	>10.2	304	3.31	1.09	476	3.53	0.741	0.515
Dust	<0.39	336	11.8	3.50	334	19.0	5.67	0.778
	0.39-0.69	141	10.9	7.75	157	18.3	11.6	0.809
	0.69-1.3	292	15.9	5.44	242	14.6	6.03	0.443
	1.3-2.1	537	11.5	2.15	452	6.20	1.37	0.260
	2.1-4.2	668	2.34	0.35	513	1.90	0.370	0.393
4.2-10.2	652	2.41	0.37	517	1.07	0.208	0.215	
>10.2	194	0.478	0.25	160	0.41	0.258	0.417	

<D.L.: Below detection limits

Table S4 Total metal concentration of target metals in size-fractionated aerosol samples.

Sampling date	Diameter µm	Ti ng/m ³	V ng/m ³	Mn ng/m ³	Co ng/m ³	Ni ng/m ³	Cu ng/m ³	Zn ng/m ³	Cd ng/m ³	Sb ng/m ³	Pb ng/m ³
Dec., 2012	<0.39	4.08	0.773	1.80	0.0359	0.534	0.911	10.8	0.155	0.448	4.68
	0.39-0.69	1.81	0.472	1.87	0.0195	0.258	0.882	7.91	0.115	0.331	4.19
	0.69-1.3	3.86	0.285	2.17	0.0261	0.203	0.831	7.80	0.0810	0.225	3.19
	1.3-2.1	6.12	0.193	1.93	0.0352	0.142	1.06	6.00	0.0582	0.194	1.78
	2.1-4.2	5.28	0.127	1.34	0.0271	0.087	0.893	2.39	0.0224	0.126	0.681
	4.2-10.2	8.72	0.168	2.32	0.0663	0.110	1.20	4.84	0.0174	0.123	0.819
	>10.2	4.53	0.0852	1.26	0.0350	0.0504	0.380	3.68	0.00545	0.0477	0.583
Jan., 2013	<0.39	7.71	0.957	3.90	0.0648	0.798	1.37	15.1	0.155	0.448	6.31
	0.39-0.69	2.85	0.444	3.91	0.0253	0.371	1.08	12.5	0.135	0.326	6.24
	0.69-1.3	4.47	0.316	4.30	0.0282	0.230	1.12	15.5	0.156	0.321	6.50
	1.3-2.1	9.72	0.227	3.62	0.0398	0.157	1.40	11.8	0.119	0.230	3.90
	2.1-4.2	10.0	0.176	2.42	0.0351	0.189	1.44	6.06	0.0337	0.179	1.48
	4.2-10.2	14.0	0.235	3.26	0.0619	0.303	1.65	6.84	0.0300	0.256	1.13
	>10.2	5.23	0.0324	1.32	0.0323	0.106	0.447	3.42	0.0255	0.0340	0.417
Feb., 2013	<0.39	5.06	0.913	3.22	0.0323	0.741	1.31	12.4	0.189	0.485	6.00
	0.39-0.69	1.39	0.296	2.19	0.0077	0.221	0.640	6.57	0.107	0.246	3.54
	0.69-1.3	3.07	0.248	3.61	0.0155	0.156	0.834	8.66	0.121	0.266	4.05
	1.3-2.1	4.19	0.130	2.56	0.0101	0.0852	0.815	5.54	0.0541	0.181	1.75
	2.1-4.2	4.44	0.0855	1.99	0.0106	0.0468	0.877	3.16	0.0287	0.158	0.783
	4.2-10.2	4.37	0.0774	2.16	0.0214	0.0741	0.97	4.07	0.0253	0.110	0.674
	>10.2	2.48	0.0108	1.08	0.0098	<D.L.	0.404	3.14	0.0127	0.0274	0.334
Mar., 2013	<0.39	9.75	2.01	6.03	0.0872	0.959	2.28	19.3	0.343	0.793	9.06
	0.39-0.69	5.40	0.968	6.21	0.0489	0.523	1.61	18.4	0.177	0.729	7.86
	0.69-1.3	7.20	0.391	4.28	0.0314	0.251	0.956	11.6	0.117	0.313	3.67
	1.3-2.1	13.4	0.402	4.06	0.0503	0.289	1.02	8.29	0.0733	0.248	2.17
	2.1-4.2	29.1	0.859	8.70	0.129	0.529	2.15	13.1	0.0788	0.394	2.78
	4.2-10.2	32.8	0.795	10.4	0.138	0.529	2.07	8.16	0.0771	0.221	1.67
	>10.2	11.4	0.263	3.69	0.0486	0.182	0.60	3.71	0.0092	0.0502	0.467
Apr., 2013	<0.39	8.31	3.07	6.84	0.0671	1.54	2.52	24.8	0.350	0.730	11.2
	0.39-0.69	3.43	0.687	3.66	0.0167	0.327	1.13	14.1	0.165	0.312	5.32
	0.69-1.3	6.64	0.543	4.05	0.0397	0.352	1.24	17.2	0.145	0.267	4.75
	1.3-2.1	11.1	0.428	3.50	0.0562	0.353	1.04	13.3	0.0892	0.183	1.85
	2.1-4.2	11.9	0.360	3.46	0.0322	0.171	0.861	7.38	0.0487	0.098	1.38
	4.2-10.2	11.7	0.318	4.66	0.0689	0.236	0.785	5.93	0.0375	0.0649	1.01
	>10.2	6.72	0.167	2.29	0.0237	0.0770	0.291	2.27	0.0104	0.0332	0.329
May, 2013	<0.39	3.28	1.02	2.49	0.0219	0.50	0.776	7.72	0.098	0.213	2.56
	0.39-0.69	2.57	0.776	2.71	0.0252	0.382	0.760	10.0	0.118	0.292	3.24
	0.69-1.3	4.76	0.606	3.63	0.0310	0.396	1.09	14.5	0.112	0.277	3.76
	1.3-2.1	8.52	0.404	2.90	0.0374	0.301	0.970	10.1	0.0576	0.192	1.82
	2.1-4.2	13.7	0.432	4.18	0.0485	0.354	1.46	7.41	0.0555	0.200	1.49
	4.2-10.2	12.4	0.352	4.47	0.0592	0.283	1.09	4.66	0.0207	0.139	1.12
	>10.2	17.7	0.492	7.75	0.0727	0.405	1.28	6.92	0.0275	0.0778	1.42
Jun., 2013	<0.39	1.37	2.03	3.31	0.0307	0.658	1.27	11.0	0.152	0.517	3.12
	0.39-0.69	0.442	0.745	1.92	0.0208	0.183	0.562	6.23	0.0632	0.185	1.51
	0.69-1.3	1.23	0.426	1.96	0.0188	0.304	0.593	6.43	0.0480	0.137	1.14
	1.3-2.1	2.43	0.297	1.82	0.0107	<D.L.	0.694	6.13	0.0484	0.100	0.921
	2.1-4.2	2.61	0.164	2.06	0.0184	0.0277	0.804	3.20	0.0301	0.0812	0.723
	4.2-10.2	2.14	0.140	3.23	0.0253	<D.L.	0.728	3.80	0.0338	0.0619	0.645
	>10.2	1.69	0.103	2.58	0.0097	<D.L.	0.412	2.07	0.00896	0.0435	0.381

Table S4 Continued.

	Diameter µm	Ti ng/m ³	V ng/m ³	Mn ng/m ³	Co ng/m ³	Ni ng/m ³	Cu ng/m ³	Zn ng/m ³	Cd ng/m ³	Sb ng/m ³	Pb ng/m ³
Jul., 2013	<0.39	4.72	2.35	3.59	0.0447	1.04	0.865	10	0.12	0.339	3.50
	0.39-0.69	1.83	1.53	2.43	0.0220	0.685	0.780	9.17	0.08	0.278	2.15
	0.69-1.3	3.03	0.785	2.54	0.0389	1.03	0.903	9.52	0.06	0.186	1.52
	1.3-2.1	5.91	0.433	2.34	0.0637	0.455	0.946	6.48	0.05	0.154	1.07
	2.1-4.2	6.67	0.289	2.64	0.0634	0.301	0.935	4.22	0.03	0.121	0.822
	4.2-10.2	8.47	0.287	3.99	0.0406	0.291	0.880	4.22	0.03	0.0867	0.815
	>10.2	5.29	0.175	3.16	0.0311	0.165	0.487	3.16	0.01	0.0257	0.410
Aug-2., 2013	<0.39	1.34	3.48	3.72	0.0908	1.02	2.44	19.0	0.26	0.661	6.24
	0.39-0.69	0.672	1.58	2.83	0.0364	0.612	2.04	15.5	0.17	0.481	4.61
	0.69-1.3	2.00	0.873	2.62	0.0274	1.53	1.94	17.1	0.15	0.369	3.75
	1.3-2.1	4.13	0.450	2.06	0.0376	0.250	1.31	12.7	0.10	0.233	2.17
	2.1-4.2	5.26	0.265	2.22	0.0335	0.162	1.16	5.11	0.04	0.149	1.16
	4.2-10.2	7.36	0.271	3.83	0.0612	0.0459	1.21	4.46	0.03	0.110	0.96
	>10.2	2.76	0.110	2.83	0.0170	<D.L.	0.460	2.54	0.01	0.0315	0.299
Sep., 2013	<0.39	4.86	1.87	6.51	0.0499	0.709	2.37	19.5	0.33	0.839	8.05
	0.39-0.69	0.899	0.461	3.90	0.0056	0.138	0.769	10.4	0.15	0.277	2.93
	0.69-1.3	2.56	0.323	4.47	0.0139	0.105	0.893	13.7	0.19	0.451	3.30
	1.3-2.1	4.35	0.221	3.10	0.0219	0.290	1.23	14.1	0.20	0.455	3.45
	2.1-4.2	4.77	0.171	2.95	0.0266	0.115	1.35	4.89	0.20	0.402	3.77
	4.2-10.2	6.58	0.182	5.02	0.0402	0.02	1.76	6.79	0.25	0.312	5.75
	>10.2	3.42	0.130	4.45	0.0367	<D.L.	1.02	4.43	0.10	0.0908	3.07
Oct., 2013	<0.39	4.06	0.601	1.98	0.0179	0.548	0.898	8.94	0.12	0.267	3.61
	0.39-0.69	1.02	0.204	1.16	0.0189	0.534	0.373	4.74	0.06	0.158	1.90
	0.69-1.3	1.89	0.108	1.24	0.0123	0.681	0.399	4.66	0.04	0.112	1.36
	1.3-2.1	2.14	0.0735	0.914	8.28.E-03	0.252	0.479	3.40	0.02	0.107	0.713
	2.1-4.2	2.89	0.0714	0.86	0.0107	0.176	0.615	2.05	0.01	0.0917	0.453
	4.2-10.2	3.47	0.0834	1.85	0.0178	0.287	0.956	2.78	0.02	0.0921	0.572
	>10.2	1.68	0.0561	0.915	8.00.E-04	0.170	0.333	1.49	0.01	0.0298	0.264
Nov., 2013	<0.39	4.72	1.23	3.00	0.0378	0.875	1.19	12.9	0.14	0.567	4.82
	0.39-0.69	2.12	0.334	2.25	0.0081	0.477	0.650	7.79	0.10	0.272	3.03
	0.69-1.3	2.93	0.166	1.65	0.0167	0.290	0.566	6.46	0.04	0.150	2.13
	1.3-2.1	8.30	0.236	2.13	0.0281	0.638	1.23	7.98	0.08	0.243	2.04
	2.1-4.2	5.50	0.136	1.22	0.0234	0.431	0.890	2.79	0.03	0.137	0.726
	4.2-10.2	8.12	0.170	1.87	0.0316	0.495	1.36	4.71	0.03	0.154	1.18
	>10.2	3.38	0.0599	0.875	0.0159	0.394	0.588	3.12	0.01	0.0488	0.68
Dec., 2013	<0.39	5.85	0.254	3.20	0.0362	1.49	1.49	17.9	0.16	0.467	6.33
	0.39-0.69	2.17	0.254	2.31	0.0272	0.885	0.945	11.5	0.16	0.399	5.62
	0.69-1.3	1.99	0.155	0.59	0.0172	0.261	0.365	3.52	0.04	0.146	1.42
	1.3-2.1	3.49	0.0925	1.09	0.0199	0.067	0.786	5.51	0.04	0.160	1.39
	2.1-4.2	4.13	0.0693	0.69	0.0217	0.352	0.923	2.74	0.01	0.114	0.661
	4.2-10.2	4.68	0.0712	0.89	0.0198	0.525	0.791	2.58	0.02	0.295	0.610
	>10.2	1.81	0.0185	0.22	2.66.E-03	0.269	0.165	1.38	0.00	0.0310	0.220
Haze	<0.39	6.01	2.09	8.04	0.171	1.43	0.601	26.9	0.41	0.954	13.0
	0.39-0.69	4.90	1.90	16.4	0.170	1.47	3.36	43.9	0.63	1.09	20.6
	0.69-1.3	9.75	1.17	16.5	0.224	1.01	3.25	45.3	0.54	0.907	19.4
	1.3-2.1	15.4	0.765	9.31	0.170	0.372	4.11	30.9	0.27	1.17	10.2
	2.1-4.2	17.2	0.610	7.48	0.234	0.835	5.13	17.4	0.12	1.07	4.88
	4.2-10.2	19.2	0.568	7.62	0.220	0.399	6.33	20.5	0.09	0.748	3.40
	>10.2	19.1	0.459	9.79	0.496	0.932	3.84	29.9	0.12	0.363	3.24
Asian dust	<0.39	21.5	3.86	10.8	0.158	2.59	3.37	33.8	0.47	0.878	18.8
	0.39-0.69	9.33	1.54	9.46	0.0868	1.51	3.08	33.1	0.38	0.663	17.7
	0.69-1.3	16.8	1.07	10.4	0.107	1.12	2.51	32.8	0.28	0.478	12.7
	1.3-2.1	31.4	1.12	10.5	0.177	0.829	2.07	23.9	0.20	0.314	6.04
	2.1-4.2	35.9	1.13	11.2	0.197	0.728	1.94	13.0	0.09	0.179	3.22
	4.2-10.2	35.5	1.00	12.0	0.174	0.936	1.52	9.97	0.08	0.124	2.00
	>10.2	10.1	0.300	4.15	0.0717	0.232	0.480	4.27	0.03	0.0432	0.586

<D.L.: Below detection limits

Table S5 Dissolved metal concentration in size-fractionated aerosol particles.

	µm	Ti ng/m ³	V ng/m ³	Mn ng/m ³	Co ng/m ³	Ni ng/m ³	Cu ng/m ³	Zn ng/m ³	Cd ng/m ³	Sb ng/m ³	Pb ng/m ³
Dec., 2012	<0.39	<D.L.	0.541	0.827	0.0216	0.209	0.539	7.57	0.117	0.305	2.34
	0.39-0.69	<D.L.	0.163	0.810	<D.L.	<D.L.	0.251	3.41	0.0716	0.0982	1.09
	0.69-1.3	<D.L.	0.134	1.00	1.60.E-03	0.0101	0.331	4.31	0.0545	0.0963	0.975
	1.3-2.1	<D.L.	0.0449	0.737	2.64.E-04	<D.L.	0.491	2.77	0.0327	0.0766	0.306
	2.1-4.2	<D.L.	0.0252	0.590	4.59.E-03	0.0174	0.438	1.54	0.0123	0.0576	0.0825
	4.2-10.2	<D.L.	0.0152	0.519	3.73.E-03	0.0123	0.191	1.18	5.38.E-03	0.0221	9.12.E-03
	>10.2	<D.L.	0.0139	0.375	2.38.E-03	0.0109	0.0925	1.25	3.89.E-03	0.0105	0.0103
Jan., 2013	<0.39	<D.L.	0.735	1.94	0.0275	0.424	0.618	13.1	0.117	0.176	3.53
	0.39-0.69	0.0264	0.455	2.58	0.0128	0.207	0.584	13.9	0.179	0.130	3.74
	0.69-1.3	0.0171	0.198	2.63	0.0082	0.0903	0.516	12.4	0.132	0.0874	3.08
	1.3-2.1	0.0272	0.0988	2.12	0.0087	0.0613	0.680	9.94	0.103	0.0679	1.45
	2.1-4.2	<D.L.	0.0324	1.46	0.0112	0.0361	0.737	5.26	0.0428	0.0391	0.342
	4.2-10.2	<D.L.	0.0089	1.07	0.0066	0.0296	0.395	3.69	0.0123	0.0168	0.108
	>10.2	<D.L.	<D.L.	0.483	1.53.E-03	0.0120	0.106	2.15	4.54.E-03	5.94.E-03	0.0518
Feb., 2013	<0.39	1.53.E-04	0.125	0.269	4.12.E-03	0.0505	0.0840	1.54	0.151	<D.L.	0.540
	0.39-0.69	<D.L.	0.174	0.942	5.57.E-03	0.0608	0.172	3.38	0.095	<D.L.	1.26
	0.69-1.3	<D.L.	0.113	1.22	4.37.E-03	0.0336	0.211	3.62	0.134	<D.L.	1.09
	1.3-2.1	<D.L.	0.0824	1.44	5.42.E-03	0.0054	0.356	3.65	0.162	<D.L.	0.654
	2.1-4.2	<D.L.	0.0373	1.12	6.32.E-03	9.23.E-03	0.518	2.70	0.227	<D.L.	0.222
	4.2-10.2	<D.L.	0.0226	1.07	5.93.E-03	0.0159	0.390	3.22	0.245	<D.L.	0.0678
	>10.2	<D.L.	0.0199	0.654	4.37.E-03	<D.L.	0.150	2.64	0.655	<D.L.	0.0426
Mar., 2013	<0.39	0.0277	1.47	3.30	0.0424	0.550	0.910	17.3	0.217	0.304	4.65
	0.39-0.69	0.0366	0.624	2.96	0.0157	0.202	0.492	14.1	0.154	0.157	2.83
	0.69-1.3	0.0387	0.317	3.31	0.0145	0.110	0.517	14.9	0.124	0.116	2.82
	1.3-2.1	<D.L.	0.0918	2.55	0.0132	0.0512	0.540	7.46	0.0661	0.0486	0.572
	2.1-4.2	0.01	0.0866	4.34	0.0268	0.0756	0.729	9.81	0.0815	0.0404	0.482
	4.2-10.2	<D.L.	0.0336	4.88	0.0209	0.0644	0.471	4.05	0.0311	0.0207	0.0311
	>10.2	<D.L.	0.0072	1.17	3.82.E-03	0.0171	0.0740	1.15	8.55.E-03	4.47.E-03	0.0207
Apr., 2013	<0.39	0.154	2.19	3.53	0.0247	0.857	1.21	20.2	0.256	0.363	7.52
	0.39-0.69	0.0809	0.461	1.98	6.23.E-03	0.111	0.646	11.5	0.121	0.151	3.35
	0.69-1.3	0.128	0.290	2.41	2.45.E-03	0.0727	0.685	14.5	0.113	0.122	2.36
	1.3-2.1	0.0773	0.116	1.62	0.0110	0.0792	0.503	9.89	0.0590	0.0678	0.509
	2.1-4.2	0.0504	0.0516	1.94	0.0204	9.74.E-03	0.341	4.97	0.0444	0.0271	0.174
	4.2-10.2	0.0559	0.0429	2.21	0.0126	0.0128	0.200	2.68	0.0156	0.0126	0.0257
	>10.2	0.169	0.0167	0.745	<D.L.	<D.L.	0.0622	0.796	9.1.E-03	7.97.E-03	9.20.E-03
May, 2013	<0.39	0.0105	0.594	1.12	0.0126	0.217	0.262	5.75	0.0606	<D.L.	0.917
	0.39-0.69	<D.L.	0.737	1.98	0.0144	0.233	0.422	11.0	0.112	<D.L.	2.55
	0.69-1.3	<D.L.	0.359	1.68	9.89.E-03	0.118	0.343	10.6	0.0774	<D.L.	1.92
	1.3-2.1	<D.L.	0.150	1.33	0.0107	0.0282	0.374	7.56	0.0435	<D.L.	0.607
	2.1-4.2	<D.L.	0.0576	1.16	9.55.E-03	0.0163	0.353	3.51	0.0218	<D.L.	0.149
	4.2-10.2	<D.L.	0.0287	1.22	8.81.E-03	0.0161	0.151	1.76	9.14.E-03	<D.L.	0.0122
	>10.2	<D.L.	0.0156	0.526	3.81.E-03	<D.L.	0.0445	0.57	1.61.E-03	<D.L.	4.80.E-03
Jun., 2013	<0.39	2.45.E-03	0.361	0.534	7.26.E-03	0.1112	0.155	2.14	0.0251	<D.L.	0.378
	0.39-0.69	<D.L.	0.477	1.13	0.0104	0.1380	0.223	4.36	0.0371	<D.L.	0.683
	0.69-1.3	<D.L.	0.307	1.09	6.27.E-03	0.2240	0.244	4.82	0.0323	<D.L.	0.559
	1.3-2.1	<D.L.	0.156	0.902	4.90.E-03	0.118	0.302	4.25	0.0241	<D.L.	0.337
	2.1-4.2	<D.L.	0.0566	0.947	4.03.E-03	0.0138	0.339	2.38	0.0208	<D.L.	0.204
	4.2-10.2	<D.L.	0.0277	1.27	6.51.E-03	0.0334	0.277	2.04	0.0145	<D.L.	0.118
	>10.2	<D.L.	0.0311	1.62	4.91.E-03	<D.L.	0.208	2.09	0.0092	<D.L.	0.100

<D.L.: Below detection limits

Table S5 Continued.

	μm	Ti ng/m ³	V ng/m ³	Mn ng/m ³	Co ng/m ³	Ni ng/m ³	Cu ng/m ³	Zn ng/m ³	Cd ng/m ³	Sb ng/m ³	Pb ng/m ³
Jul., 2013	<0.39	0.0211	1.25	1.33	0.0264	0.384	0.364	5.12	0.0652	0.114	1.18
	0.39-0.69	0.0315	1.45	2.33	0.0209	0.557	0.611	8.76	0.0993	0.154	1.32
	0.69-1.3	0.0259	0.534	1.52	0.0130	0.403	0.521	6.50	0.0515	0.0696	0.653
	1.3-2.1	0.0161	0.290	1.18	0.0143	0.155	0.750	5.50	0.0369	0.0454	0.451
	2.1-4.2	0.0133	0.104	1.02	0.0105	0.099	0.532	2.59	0.0271	0.0255	0.218
	4.2-10.2	0.0442	0.0677	1.72	0.0154	0.102	0.436	3.14	0.0283	0.0182	0.102
	>10.2	0.0114	0.0219	0.863	0.0135	0.143	0.141	1.59	5.20.E-03	0.0106	0.0152
Aug., 2013	<0.39	7.05.E-03	1.02	0.855	0.0206	0.279	0.392	5.18	0.0649	<D.L.	1.47
	0.39-0.69	<D.L.	0.727	1.20	0.0124	0.239	0.667	9.13	0.0844	<D.L.	1.79
	0.69-1.3	<D.L.	0.540	1.46	0.0135	0.845	0.880	12.3	0.096	<D.L.	1.72
	1.3-2.1	<D.L.	0.459	2.09	0.0218	0.113	1.28	17.4	0.133	<D.L.	1.57
	2.1-4.2	<D.L.	0.142	1.46	0.0192	0.0399	0.809	5.21	0.0515	<D.L.	0.44
	4.2-10.2	<D.L.	0.0761	2.16	0.0269	0.0543	0.610	3.82	0.0314	<D.L.	0.132
	>10.2	<D.L.	0.0726	2.51	0.0129	<D.L.	0.340	3.42	0.0197	<D.L.	0.0570
Sep., 2013	<0.39	0.0558	1.80	4.40	0.0194	0.623	1.15	22.3	0.365	0.492	5.27
	0.39-0.69	0.0316	0.405	2.94	5.99.E-03	0.147	0.555	11.1	0.135	0.146	1.81
	0.69-1.3	0.0232	0.265	3.24	7.73.E-03	0.0837	0.575	13.1	0.176	0.169	2.08
	1.3-2.1	0.0188	0.166	2.24	0.0114	0.0577	1.00	9.77	0.236	0.208	2.10
	2.1-4.2	0.0229	0.106	2.55	0.0178	0.0758	1.34	6.21	0.247	0.205	2.13
	4.2-10.2	9.06.E-03	0.0449	2.87	0.0145	0.0530	0.757	4.15	0.197	0.111	1.15
	>10.2	6.88.E-03	0.0219	1.58	6.52.E-03	0.0452	0.176	2.69	0.0492	0.0249	0.146
Oct., 2013	<0.39	5.97.E-03	0.450	0.951	6.30.E-03	0.138	0.378	6.87	0.099	0.224	1.60
	0.39-0.69	4.28.E-03	0.0866	0.495	2.04.E-03	7.25.E-03	0.105	2.40	0.0241	0.0549	0.544
	0.69-1.3	0.0118	0.0622	0.704	2.78.E-03	0.163	0.220	3.87	0.0274	0.0569	0.542
	1.3-2.1	6.46.E-03	0.0295	0.576	3.19.E-03	4.29.E-03	0.343	3.04	0.0165	0.0384	0.231
	2.1-4.2	0.0061	0.0182	0.599	3.07.E-03	8.15.E-03	0.376	1.49	9.90.E-03	0.0325	0.109
	4.2-10.2	0.0134	0.0200	1.16	4.63.E-03	0.0234	0.544	1.85	0.0124	0.0389	0.114
	>10.2	4.43.E-03	7.32.E-03	0.418	1.40.E-03	0.0132	0.123	0.900	2.14.E-03	9.04.E-03	0.0371
Nov., 2013	<0.39	9.07.E-03	1.11	1.86	0.0312	0.406	0.555	11.8	0.138	0.286	3.10
	0.39-0.69	7.66.E-03	0.266	1.34	6.75.E-03	0.0780	0.237	6.24	0.0616	0.099	1.45
	0.69-1.3	0.0138	0.145	1.61	7.67.E-03	0.0825	0.391	8.76	0.0635	0.0937	1.33
	1.3-2.1	8.02.E-03	0.0653	1.17	7.66.E-03	0.0230	0.494	5.55	0.0455	0.0680	0.517
	2.1-4.2	9.57.E-03	0.0340	1.17	0.0122	0.0281	0.761	3.45	0.0261	0.0677	0.223
	4.2-10.2	7.21.E-03	0.0199	0.951	0.0112	0.0242	0.502	2.74	0.0173	0.0352	0.129
	>10.2	1.74.E-03	0.0153	0.396	6.61.E-03	4.97.E-03	0.144	2.05	6.63.E-03	0.0179	0.0625
Dec., 2013	<0.39	0.0110	0.438	1.19	0.0240	0.172	0.505	10.7	0.112	0.291	2.63
	0.39-0.69	6.29.E-03	0.117	0.832	7.66.E-03	0.0433	0.236	5.33	0.0690	0.126	1.47
	0.69-1.3	0.0125	0.0766	1.17	0.0119	0.0360	0.358	6.71	0.0744	0.105	0.815
	1.3-2.1	8.91.E-03	0.0396	0.837	0.0058	0.0157	0.520	5.23	0.0392	0.0703	0.513
	2.1-4.2	5.98.E-03	0.0186	0.524	0.0060	0.0053	0.468	2.35	0.0159	0.0489	0.154
	4.2-10.2	8.78.E-03	0.0152	0.725	0.0083	0.0255	0.473	2.51	0.0123	0.0366	0.0619
	>10.2	<D.L.	0.0101	0.359	3.97.E-03	0.0138	0.105	1.78	6.05.E-03	0.0107	0.0316
Haze	<0.39	7.27.E-03	1.12	2.40	0.0467	0.783	1.07	13.1	0.271	0.372	1.66
	0.39-0.69	0.581	2.09	13.0	0.0873	1.41	2.02	47.3	0.887	0.892	9.49
	0.69-1.3	0.0403	0.888	10.8	0.0661	0.912	2.14	41.4	0.456	0.775	7.16
	1.3-2.1	0.0249	0.376	6.58	0.0719	0.753	3.02	27.7	0.300	0.898	2.87
	2.1-4.2	<D.L.	0.0974	2.68	0.0367	0.497	2.34	9.92	0.0915	0.622	0.703
	4.2-10.2	0.152	0.0868	4.16	0.0642	0.532	3.29	14.3	0.0979	0.375	0.578
	>10.2	<D.L.	<D.L.	2.67	0.0571	0.416	0.928	11.2	8.77.E-03	0.110	0.241
Asian dust	<0.39	0.0967	3.26	5.00	0.0724	1.21	2.33	28.8	0.481	0.511	5.83
	0.39-0.69	0.0649	1.13	6.90	0.0489	0.674	2.10	34.4	0.369	0.456	5.44
	0.69-1.3	0.0782	0.710	6.75	0.0411	0.339	1.52	31.4	0.366	0.240	4.29
	1.3-2.1	0.102	0.318	7.05	0.0609	0.423	1.23	23.0	0.201	0.136	0.788
	2.1-4.2	0.0357	0.194	9.07	0.0603	0.254	0.893	10.2	0.0830	0.0688	0.0861
	4.2-10.2	0.0326	0.105	6.16	0.0501	0.150	0.477	48.9	0.0157	0.0351	0.0388
	>10.2	0.0117	0.0254	1.40	9.94.E-03	0.0920	0.104	1.55	3.4.E-03	9.04.E-03	0.0143

<D.L.: Below detection limits

References

- Bray, A. W., Oelkers, E. H., Bonneville, S., Wolff-Boenisch, D., Potts, N. J., Fones, G., and Benning, L. G.: The effect of pH, grain size, and organic ligands on biotite weathering rates, *Geochim. Cosmochim. Acta*, 164, 127–145, <http://dx.doi.org/10.1016/j.gca.2015.04.048>, 2015.
- Desboeufs, K. V., Losno, R., Colin, J. L.: Factors influencing aerosol solubility during cloud processes, *Atmos. Environ.*, 35, 3529–3537, [https://doi.org/10.1016/S1352-2310\(00\)00472-6](https://doi.org/10.1016/S1352-2310(00)00472-6), 2001.
- Kodama, H., Schnitzer, M.: Dissolution of chlorite minerals by fulvic acid, *Can. J. Soil Sci.*, 53, 240–243, <https://doi.org/10.4141/cjss73-036>, 1973.
- Lowson, R. T., Comarmond, J., Rajaratnam, G., Brown, P. L.: The kinetics of the dissolution of chlorite as a function of pH and at 25°C, *Geochim. Cosmochim. Acta*, 69, 1687–1699, <https://doi.org/10.1016/j.gca.2004.09.028>, 2005.