

Table S1 Total mass concentrations of the elements in the soluble and insoluble fractions of size-classified APM

Element		Nov. 2004			Apl. 2005			Aug. 2005			Oct. 2005		
		<2 μm	2-11 μm	>11 μm	<2 μm	2-11 μm	>11 μm	<2 μm	2-11 μm	>11 μm	<2 μm	2-11 μm	>11 μm
Major (wt%)	Na	0.490	4.79	3.43	0.811	4.47	1.65	0.699	7.97	2.45	0.610	5.73	3.47
	Mg	0.103	1.26	1.21	0.345	1.30	0.797	0.225	1.41	0.939	0.169	1.30	1.22
	Al	0.366	2.86	3.57	1.46	3.49	3.13	0.756	1.66	6.75	0.363	2.38	4.19
	K	0.744	0.930	0.673	0.861	0.989	0.690	0.693	0.854	0.786	0.917	0.989	1.09
	Ca	0.456	4.32	6.22	0.940	2.94	2.79	0.817	3.00	5.75	0.639	3.01	6.36
	Fe	0.953	5.23	4.73	1.59	3.20	2.97	1.15	3.55	4.08	0.991	3.47	4.62
Trace ($\mu\text{g/g}$)	Li	8.61	14.6	12.6	11.3	17.7	10.9	5.60	6.73	10.6	9.14	14.0	8.41
	Be	0.164	0.674	0.590	0.409	0.939	0.620	0.043	0.012	N.D.	0.074	0.225	N.D.
	Ti	333	2500	2620	976	2190	1920	696	1570	2370	369	2200	2550
	V	108	82.5	65.1	200	77.9	65.9	280	82.9	60.8	147	66.8	63.7
	Cr	108	351	417	112	165	149	94.2	166	196	63.6	175	34.3
	Mn	898	1200	1150	690	927	741	477	725	970	1210	954	1120
	Co	4.82	16.0	13.9	7.14	12.3	9.82	3.40	7.96	2.47	4.53	12.9	20.8
	Ni	96.3	120	136	122	84.5	70.5	294	192	17.5	81.0	46.4	34.2
	Cu	399	1510	762	326	531	217	488	1000	416	468	1020	545
	Zn	6220	6890	3410	3830	2860	847	3370	3670	1850	9110	7140	2400
	As	69.5	31.5	7.54	74.7	38.7	10.7	68.9	23.8	12.0	107	41.8	12.2
	Se	62.0	11.4	N.D.	53.6	5.86	N.D.	52.3	13.4	N.D.	66.5	11.6	N.D.
	Mo	75.0	112	88.2	41.6	17.7	6.25	187	59.3	N.D.	43.8	25.8	74.6
	Cd	30.9	10.2	N.D.	27.7	10.4	1.26	23.7	12.3	3.24	41.2	6.39	N.D.
	Sb	130	233	76.2	96.0	77.7	24.9	105	138	7.48	160	150	67.9
	Ba	243	1630	889	294	822	329	566	1200	761	257	1250	1270
	Pb	1150	679	291	990	319	103	715	291	184	1590	564	285

Table S2 Minimum, maximum, median and mean mass concentrations of the elements in size-classified APM obtained during the long-term monitoring in Tokyo (May, 1995 – December, 2006)

Element	< 2 μm				2-11 μm				> 11 μm				
	minimum	maximum	median	mean	minimum	maximum	median	mean	minimum	maximum	median	mean	
Major (wt%)	Na	0.0388	3.57	0.666	0.747	1.12	19.3	4.62	4.94	0.531	5.07	1.96	2.20
	Mg	0.0028	0.643	0.127	0.156	0.222	2.00	1.21	1.15	0.142	2.59	0.931	0.923
	Al	0.128	2.07	0.509	0.628	0.917	5.29	2.52	2.64	0.405	14.3	4.00	4.11
	K	0.398	1.41	0.790	0.804	0.128	2.57	0.996	1.01	0.476	2.96	0.720	0.748
	Ca	0.211	2.78	0.595	0.670	1.07	6.33	3.68	3.78	0.694	15.9	5.99	6.07
	Fe	0.434	2.50	0.932	1.00	1.33	9.28	4.16	4.10	0.714	11.2	4.11	4.19
Trace ($\mu\text{g/g}$)	Li	1.23	47.1	8.36	9.10	1.67	41.6	13.1	14.2	0.0073	20.1	12.0	11.5
	Be	0.0084	1.79	0.145	0.244	0.105	5.65	0.644	0.786	0.0191	4.03	0.678	0.818
	Ti	145	1540	361	448	461	5110	1060	2190	535	9600	2600	2670
	V	35.5	947	172	211	13.9	240	100	104	17.5	307	94.5	97.8
	Cr	18.6	1100	96.3	112	42.2	1200	263	278	103	3280	299	352
	Mn	99.7	1200	662	687	104	2920	1020	1030	232	3030	1030	1090
	Co	0.325	25.5	5.14	5.84	2.68	37.6	14.9	15.3	3.18	56.8	16.8	17.2
	Ni	23.3	1240	118	141	17.8	1120	122	138	15.1	2730	171	225
	Cu	102	805	426	440	119	2990	982	1060	143	1370	492	515
	Zn	449	10900	4450	4930	246	17500	3890	4240	14.4	6470	2100	2180
	As	31.0	129	66.6	66.8	10.7	79.3	29.8	31.2	0.80	35.5	10.3	10.6
	Se	19.9	99.9	58.4	57.4	2.28	43.9	14.0	14.5	0.193	12.4	3.30	3.61
	Mo	7.26	211	67.9	72.3	1.79	376	83.4	88.2	0.28	529	64.9	86.6
	Cd	11.1	103	39.0	42.4	2.00	56.2	12.2	14.4	0.0567	26.1	3.39	4.23
	Sb	38.7	459	167	181	15.4	461	166	181	5.75	146	52.4	52.6
	Ba	29.7	735	209	236	112	2880	1010	1150	67.5	2450	508	583
	Pb	422	5110	1260	1420	141	2260	566	649	70.0	641	259	266