

**Supplementary Table S1** Analytical results of National Institute of Standard and Technology (NIST) SRM 1648

Element	Analytical mode <sup>a</sup>	Mass / Charge	Detection limit <sup>b</sup>	Found <sup>c</sup> (mean ± sd)			Certified / Reference <sup>d</sup>	Recovery
				m/z	pg ml <sup>-1</sup>	μg g <sup>-1</sup>		
Be	NG	9	0.228		1.27 ± 0.11	0.11	—	—
Al	He	27	185		32500 ± 1100	34200	95.1	
V	He	51	2.06		130 ± 4	127	102.4	
Cr	He	53	2.65		400 ± 24	403	99.4	
Mn	H <sub>2</sub>	55	1.71		778 ± 34	786	99.0	
Fe	H <sub>2</sub>	57	119		38100 ± 1600	39100	97.5	
Co	He	59	0.734		17.0 ± 0.8	18	<sup>d</sup>	94.2
Ni	He	60	2.89		66.0 ± 1.2	82		80.4
Cu	He	63	16.7		563 ± 45	609		92.5
Zn	He	66	88.4		4780 ± 148	4760		100.4
As	He	75	0.819		114 ± 3	115		99.0
Se	H <sub>2</sub>	78	2.17		24.7 ± 1.1	27		91.5
Mo	He	95	2.29		14.6 ± 0.6	—		—
Ag	He	107	0.331		6.05 ± 0.16	6	<sup>d</sup>	100.9
Cd	He	111	0.461		71.8 ± 4.4	75		95.7
Sb	He	121	1.27		42.7 ± 1.5	45	<sup>d</sup>	95.0
Ba	He	137	29.3		702 ± 20	737	<sup>d</sup>	95.3
Tl	He	205	0.0632		2.34 ± 0.09	—		—
Pb	He	208	17.1		6650 ± 156	6550		101.6
Th	He	232	0.120		7.01 ± 0.05	7.4	<sup>d</sup>	94.7
U	He	238	0.0367		5.30 ± 0.14	5.5		96.4

<sup>a</sup> Determination was performed under three different analytical modes. NG means non-gas mode which was performed without octopole reaction cell system. He means collision mode by using helium gas. H<sub>2</sub> means reaction mode by using hydrogen gas. <sup>b</sup> Values were defined as three times the standard deviation of 10 calibration blank analyses. <sup>c</sup> Values were derived from the analyses of three samples. <sup>d</sup> Reference value.

**Supplementary Table S2** Elemental concentrations<sup>a</sup> in size-classified airborne particulate matter (APM)

Element	Concentration / ng m <sup>-3</sup>					
	Roadside site (Tatebayashi)			Residential site (Maebashi)		
	< 2.1 μm	2.1–11 μm	> 11 μm	< 2.1 μm	2.1–11 μm	> 11 μm
Be	0.0023	0.018	0.011	0.0045	0.011	0.0036
Al	91	830	620	93	620	250
V	1.6	1.5	1.0	1.2	1.1	0.43
Cr	13	4.2	1.5	3.0	3.5	0.78
Mn	18	17	10	6.9	9.5	3.0
Fe	250	720	410	87	370	130
Co	0.17	0.30	0.18	0.13	0.29	0.087
Ni	4.0	2.0	0.80	2.0	4.5	0.35
Cu	10	23	4.9	5.0	11	3.5
Zn	79	48	41	32	20	5.7
As	0.93	0.35	0.12	0.74	0.25	0.045
Se	0.69	0.070	0.0092	0.37	0.050	0.0039
Mo	2.8	1.5	0.32	2.4	1.1	0.12
Ag	0.23	0.052	0.015	0.086	0.031	0.0039
Cd	0.40	0.063	0.015	0.48	0.041	0.0090
Sb	3.4	3.9	0.75	2.2	0.99	0.13
Ba	9.8	33	8.7	5.4	11	2.1
Tl	0.058	0.016	0.0057	0.037	0.012	0.0015
Pb	13	3.9	1.4	11	2.7	0.52
Th	0.0093	0.090	0.063	0.0062	0.048	0.015
U	0.0056	0.033	0.021	0.018	0.029	0.0086

<sup>a</sup> Elemental concentrations in 13 size fractions were merged into 3 size ranges of  $D_p < 2.1$ , 2.1–11, and > 11 μm.