Comments to the Author:

The following alterations are still needed for the Supplement before the manuscript can be published in ACP.

Dear Prof. Dr. Maenhaut,

Thank you very much for your comments to further improve our manuscript. We addressed all comments (in italic typeset) and prepared a point-to-point response (in regular typeset).

In Table S1:

- second column, for P: replace "9" by "9.0";

Done.

- second column, for Cl: replace "3" by "3.0";

Done.

- second column, for K: replace "2" by "2.0";

Done.

- second column, for Sb: replace "9" by "9.0";

Done.

- fourth column, for Zr: replace "7" by "7.0";

Done.

- fifth column, for Si: replace "3.52" by "3.5";

Done.

- fifth column, for Se: replace "7" by "7.0";

Done.

- sixth column, for Rb: replace "9" by "9.0";

Done.

- seventh column, for Si: replace "2.43" by "2.4";

Done.

- eighth column, for Ba: replace "16" by "16.0";

Done.

- ninth column, for Ti: replace "7.09" by "7.1";

Done.

- ninth column, for Cr: replace "17" by "17.0";

Done.

- ninth column, for Mn: replace "11" by "11.0".

Done.

In Table S2:

- Delhi, third column, for Se: replace "3" by "3.0";

Done.

- Delhi, seventh column, for As: replace "10" by "10.0";

Done.

- Delhi, seventh column, for Sn: replace "18" by "18.0";

Done.

- Delhi, eighth column, for Cu: replace "13" by "13.0";

Done.

- Delhi, ninth column, for Ba: replace "15" by "15.0";

Done.

- Beijing, second column, for Br: replace "14" by "14.0";

Done.

- Beijing, seventh column, for Ba: replace "4" by "4.0";

Done.

- Beijing, ninth column, for As: replace "7" by "7.0";

Done.

- Krakow, fourth column, for Cu: replace "12" by "12.0";

Done.

- Krakow, sixth column, for Cu: replace "14" by "14.0";

Done.

- Krakow, sixth column, for Sb: replace "10" by "10.0";

Done.

- Krakow, ninth column, for Br: replace "15" by "15.0";

Done.

- London, third column, for Zn: replace "6" by "6.0";

Done.

- London, fourth column, for Cu: replace "4" by "4.0";

Done.

- London, seventh column, for Pb: replace "1" by "1.0";

Done.

- London, ninth column, for Mn: replace "3" by "3.0";

Done.

- London, ninth column, for Cu: replace "6" by "6.0".

Done.

In Table S3:

- second column, for Ca: replace "10" by "10.0";

Done.

- third column, for Fe: replace "2" by "2.0";

Done.

- fifth column, for Ca: replace "8" by "8.0";

Done.

- fifth column, for Fe: replace "10" by "10.0";

Done.

- eighth column, for Ca: replace "8" by "8.0".

Done.

Supplementary of

Highly time-resolved measurements of element concentrations in PM_{10} and $PM_{2.5}$: Comparison of Delhi, Beijing, London, and Krakow

Pragati Rai et al.

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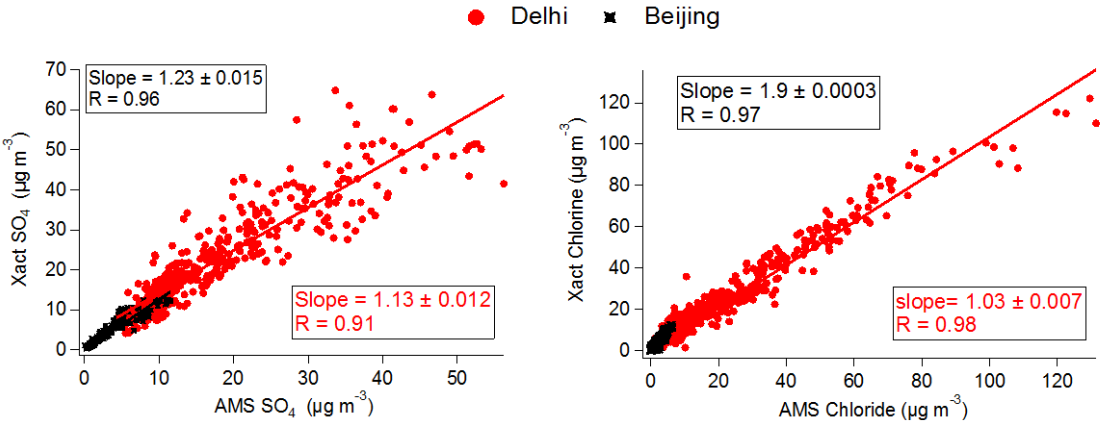
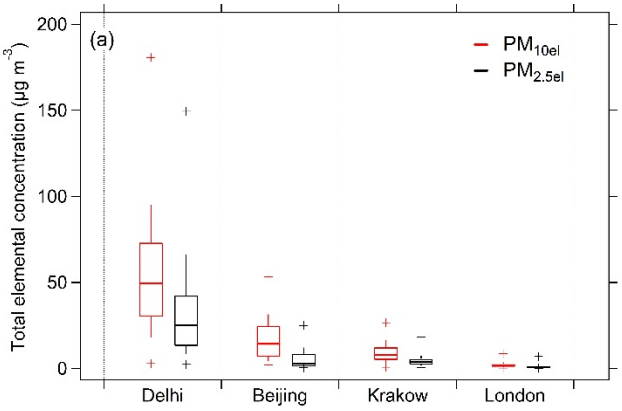


Figure S1: Comparison of Xact $PM_{2.5}$ sulfate (3×S, assuming that all S occurred in the form of sulfate.) vs. AMS sulfate (left) and Xact $PM_{2.5}$ Cl vs. AMS chloride (right) at Delhi (red; AMS PM_1) and Beijing (black; AMS $PM_{2.5}$) sites.



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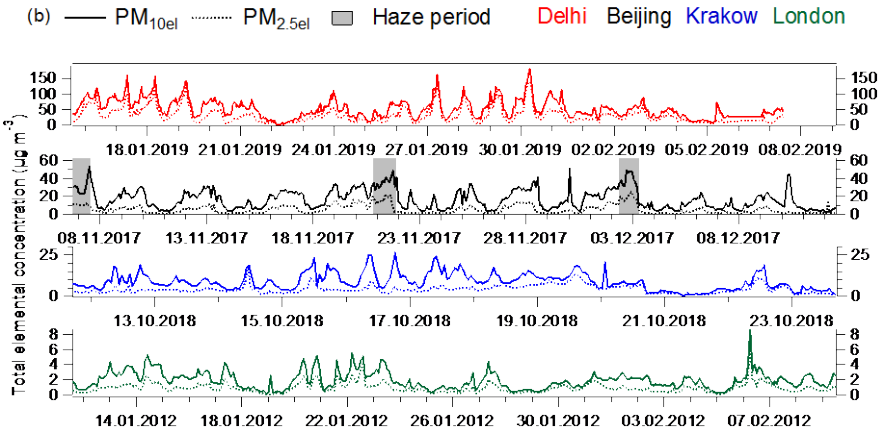


Figure S2: (a) Box-Whisker plots (top to bottom: maximum-p90-p75-p50-p25-p10-minimum) of total PM_{10el} and $PM_{2.5el}$ concentrations and (b) Time series of hourly PM_{10el} and $PM_{2.5el}$ at Delhi, Beijing, Krakow, and London.

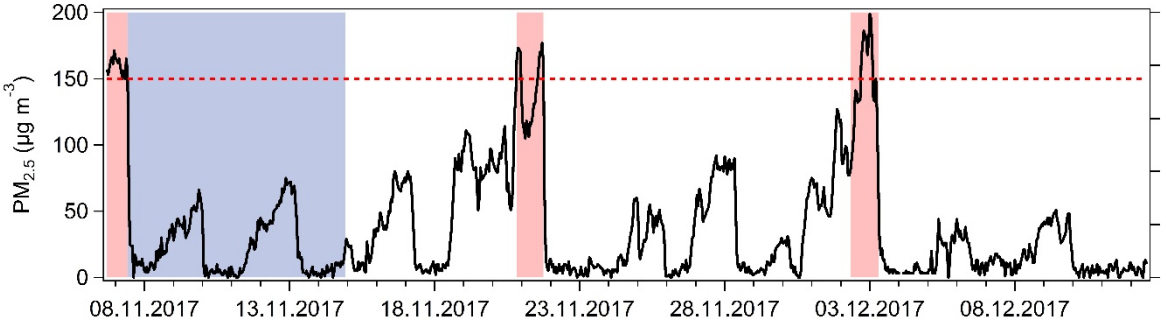


Figure S3: Time series of $PM_{2.5}$ total mass concentration in Beijing measured at the nearest national monitoring network station in Haidian District (2.5 km away from the Xact sampling site). The periods highlighted with a blue background relate to the non-heating period, while the rest belongs to the heating period. The red background colors represent haze events based on $PM_{2.5}$ concentrations ≥ 150 µg m^{-3} with a dashed red line, whereas the remaining periods are called non-haze.

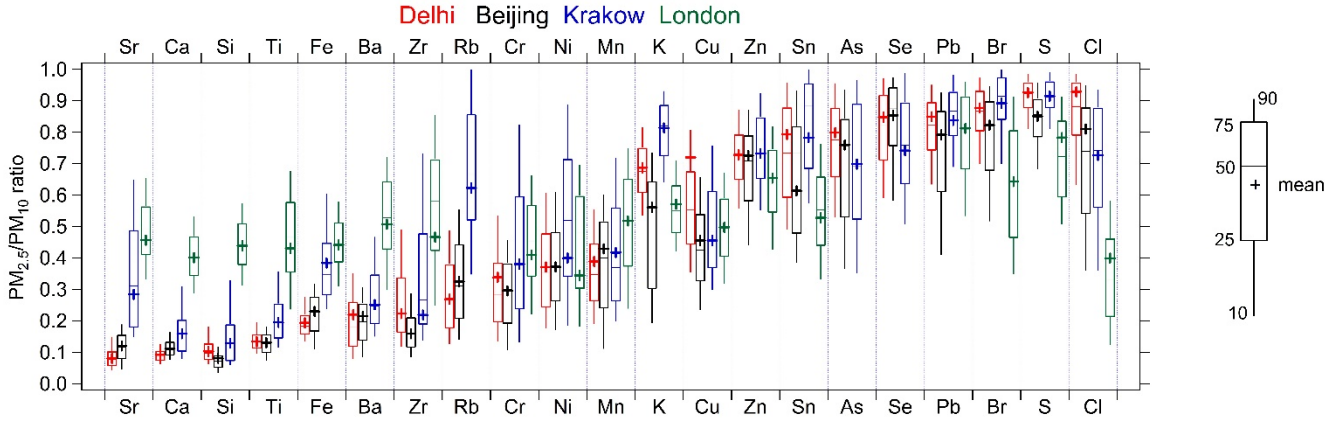


Figure S4: Box-whisker plots of the measured elemental $PM_{2.5}/PM_{10}$ ratios at all four sites. Box: First to third quartile range, −: median line, +: mean, whiskers: 10-90% percentiles. Note that Rb, As and Se were not measured in London. London measurements should be interpreted with caution due to uncertainties in the low size cut-off of the RDI, as discussed in Section 2.2.

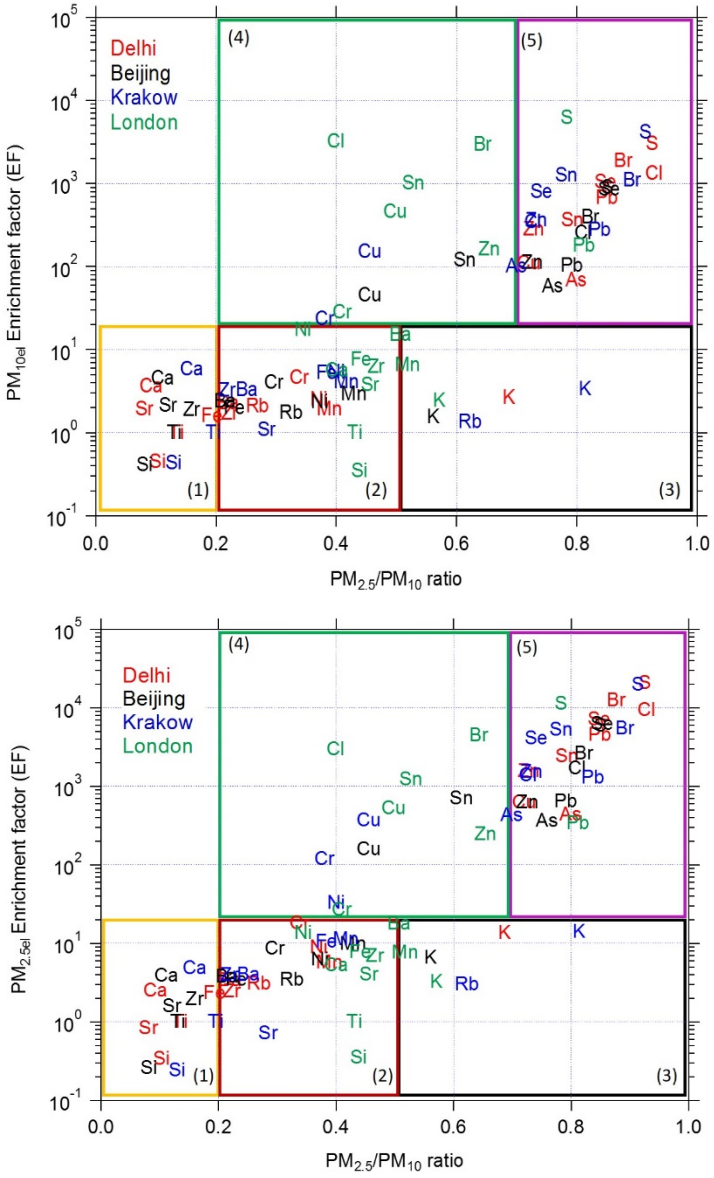


Figure S5: Enrichment factors (EF) vs $PM_{2.5}/PM_{10}$ ratios at all four sites. The upper graph is for PM_{10el} EF vs $PM_{2.5}/PM_{10}$ and the lower graph is for $PM_{2.5el}$ EF vs $PM_{2.5}/PM_{10}$. Note that Rb, As and Se were not measured in London. London measurements should be interpreted with caution (especially with respect to the $PM_{2.5}/PM_{10}$ ratio) due to uncertainties in the low size cut-off of the RDI, as discussed in Section 2.2.

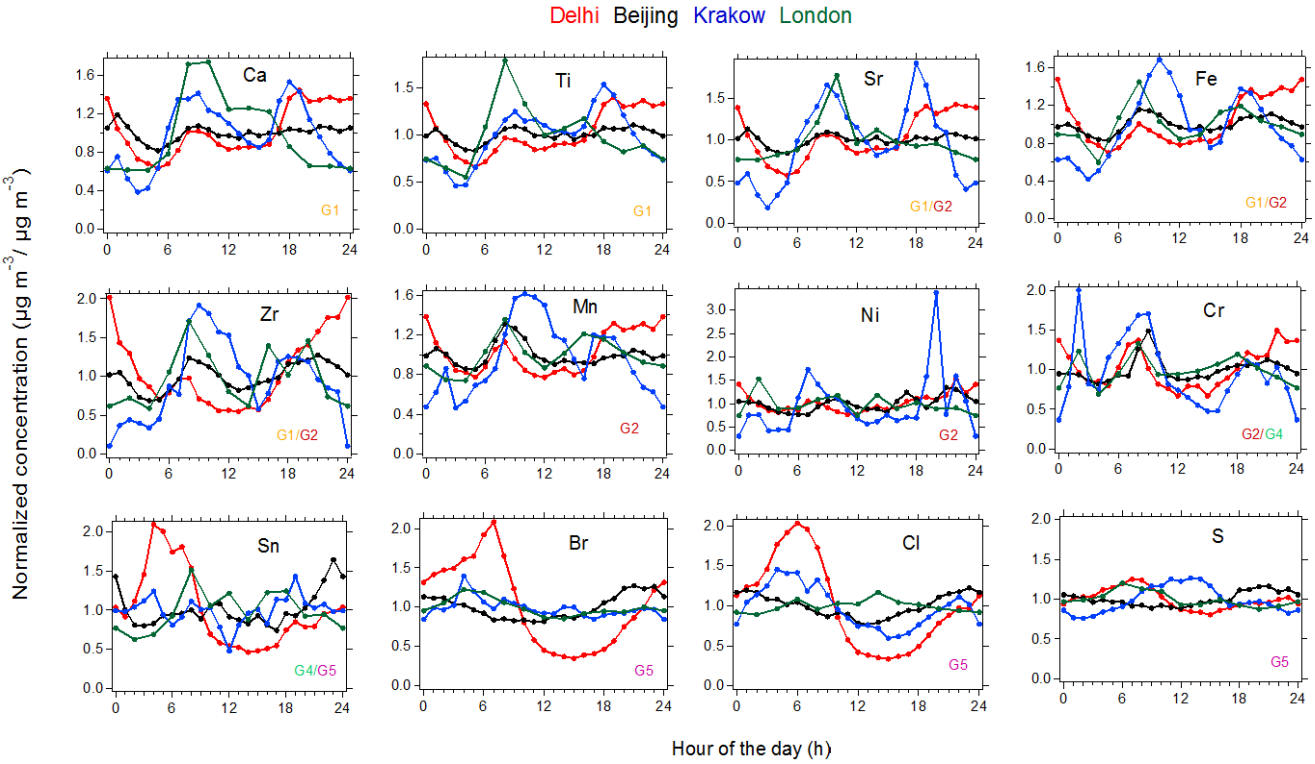
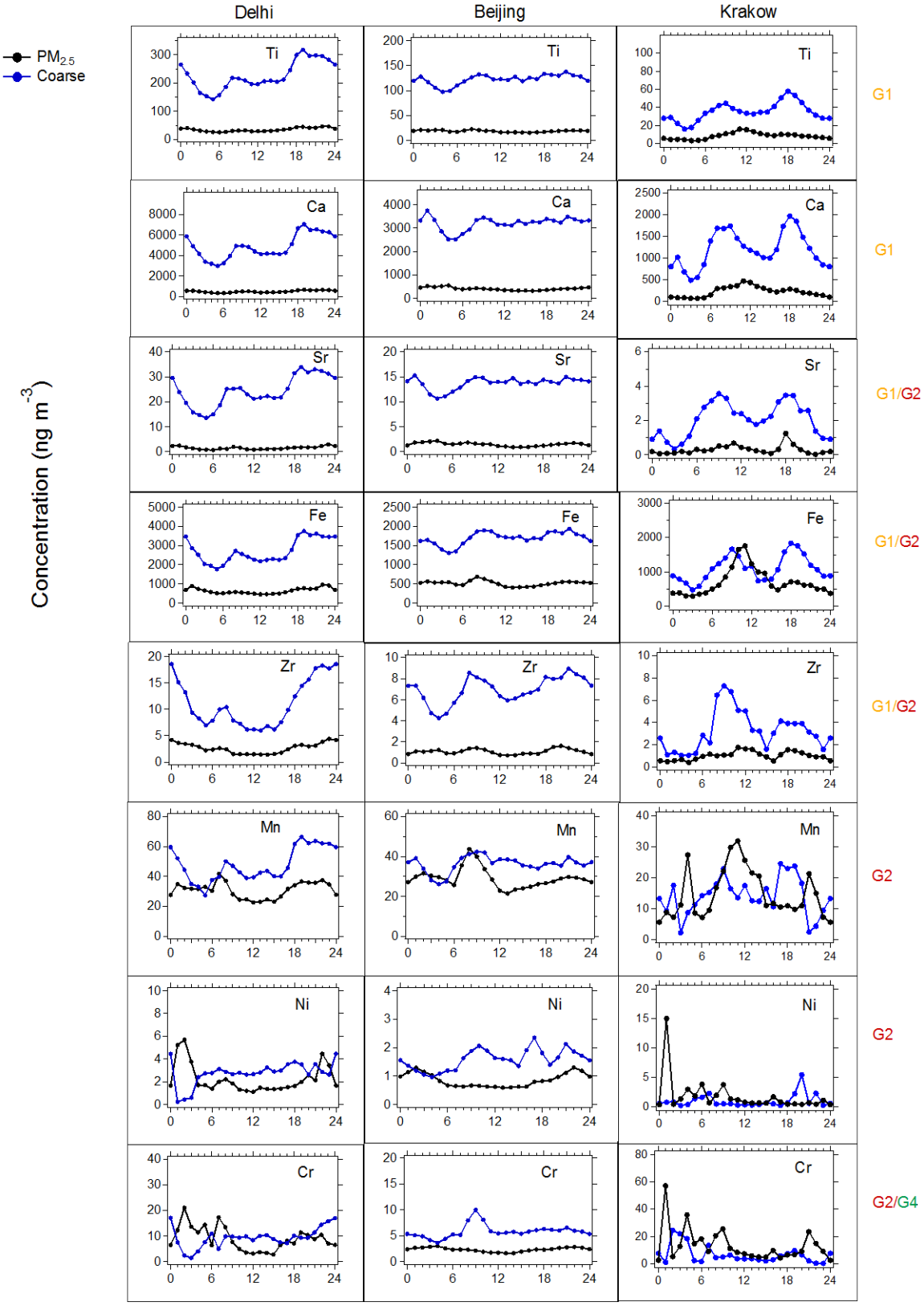


Figure S6: Diurnal patterns (mean) of elements in each group (G1: Group 1, G2: Group 2, G3: Group 3, G4: Group 4, G5: Group 5) in PM_{10} normalized by the mean values of the elements in PM_{10} at Delhi, Beijing, Krakow, and London. Note that due to the time resolution of the original data the London data are 2-hour averages, while the other data are one-hour averages.


Delhi
Beijing
Krakow
PM2.5
Coarse
Concentration (ng m-3)
Ti
Ca
Sr
Fe
Zr
Mn
Ni
Cr
G1
G1
G1/G2
G1/G2
G1/G2
G2
G2
G2/G4

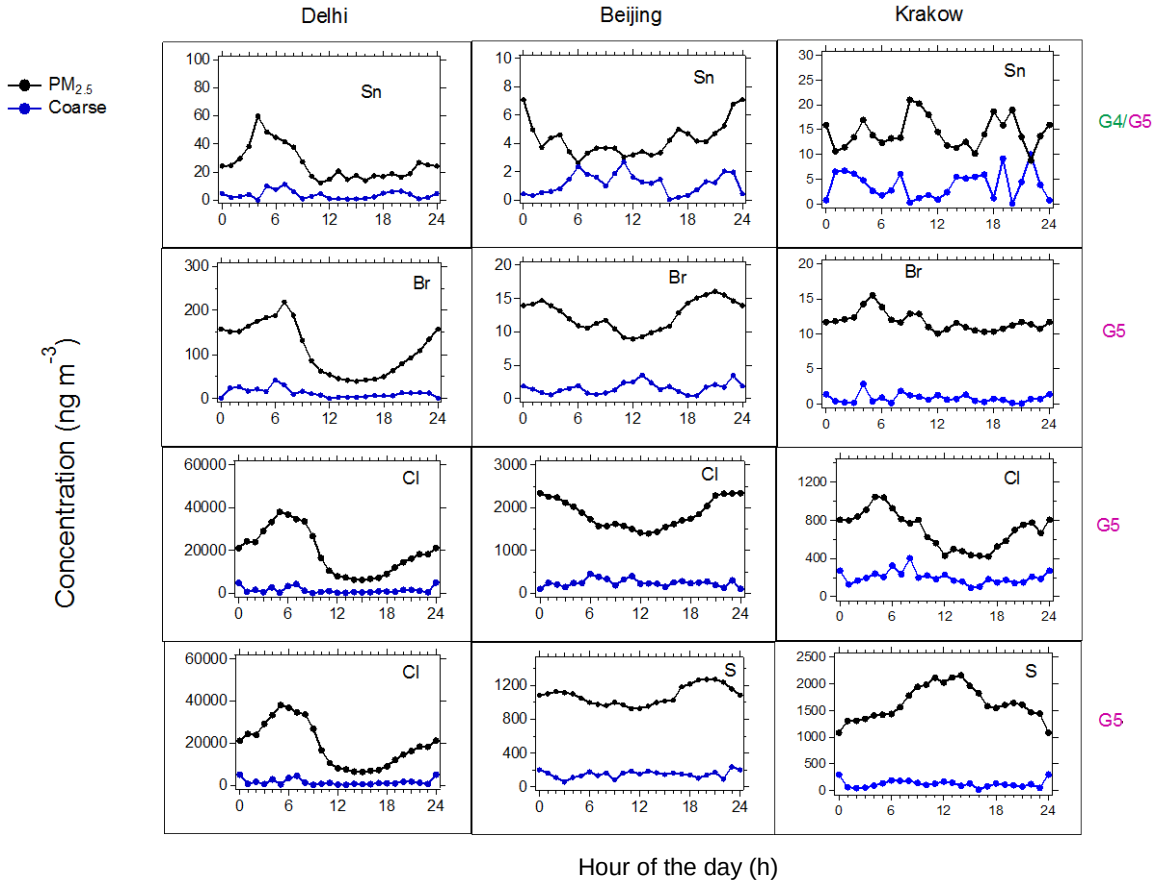


Figure S7: Diurnal patterns (mean) of elements in $PM_{2.5}$ and coarse size fractions (PM_{10}–$PM_{2.5}$) at Delhi, Beijing, and Krakow, in each group (G1: Group 1, G2: Group 2, G3: Group 3, G4: Group 4, G5: Group 5).

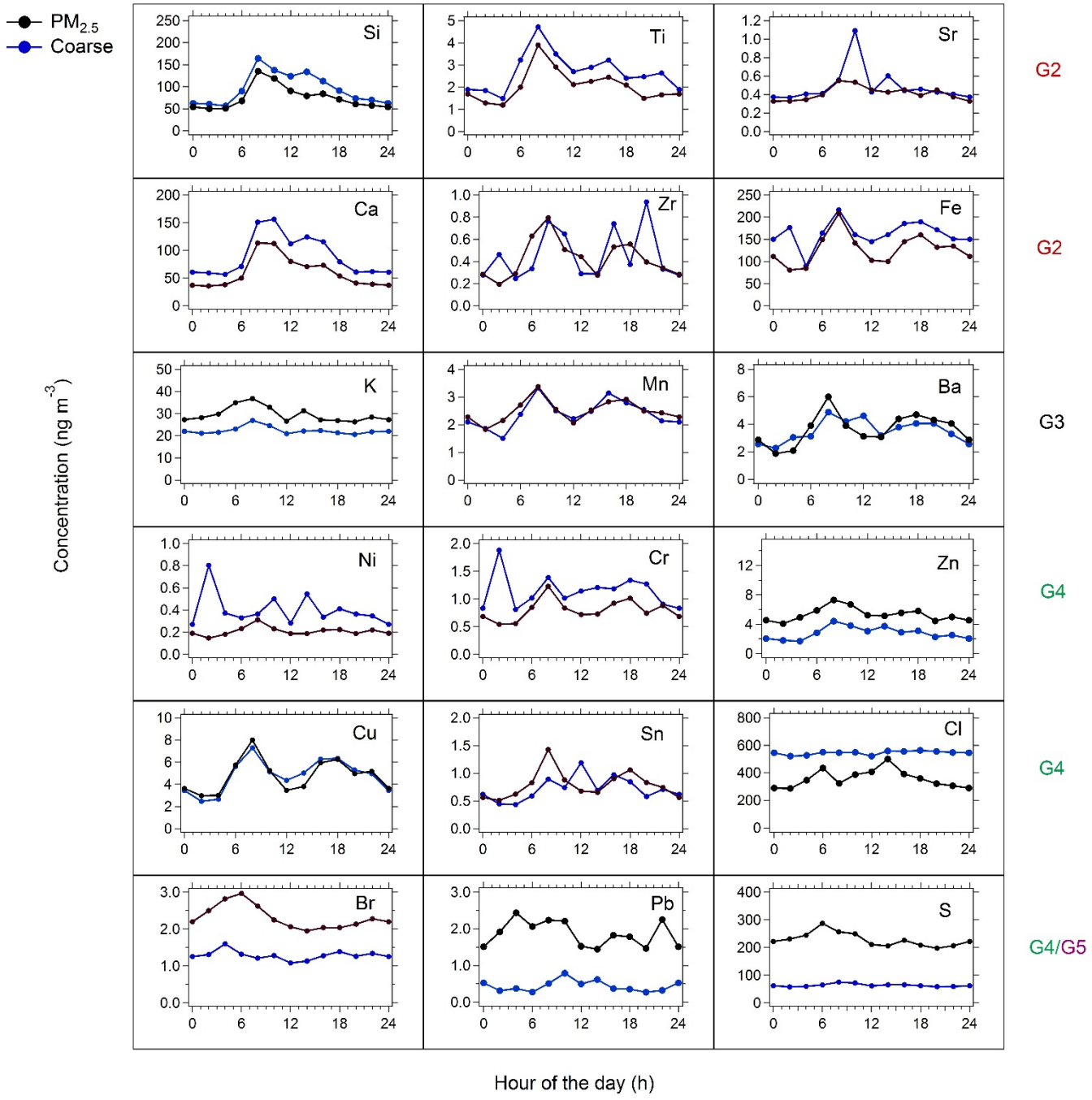


Figure S8: Diurnal patterns (mean) of elements in $PM_{2.5}$ and coarse size fractions (PM_{10}–$PM_{2.5}$) at London, in each group (G2: Group 2, G3: Group 3, G4: Group 4, G5: Group 5). Note that Group 1 is absent in London. Note that due to the time resolution of the original data of London, hour of the day on the x-axis represents 2-hour averages. London measurements should be interpreted with caution due to uncertainties in the low size cut-off of the RDI, as discussed in Section 2.2.

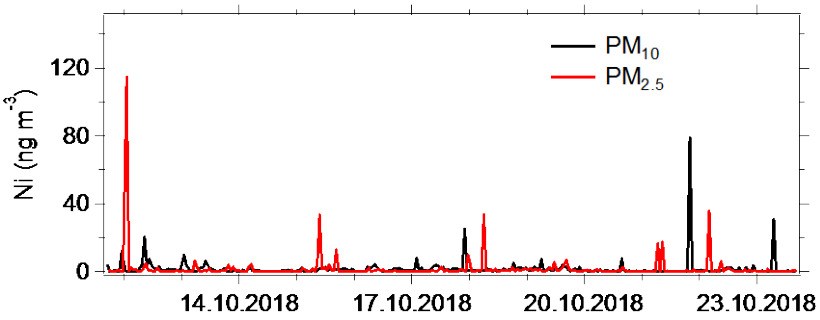


Figure S9: Time series of Ni in PM_{10} (black) and $PM_{2.5}$ (red) in Krakow.

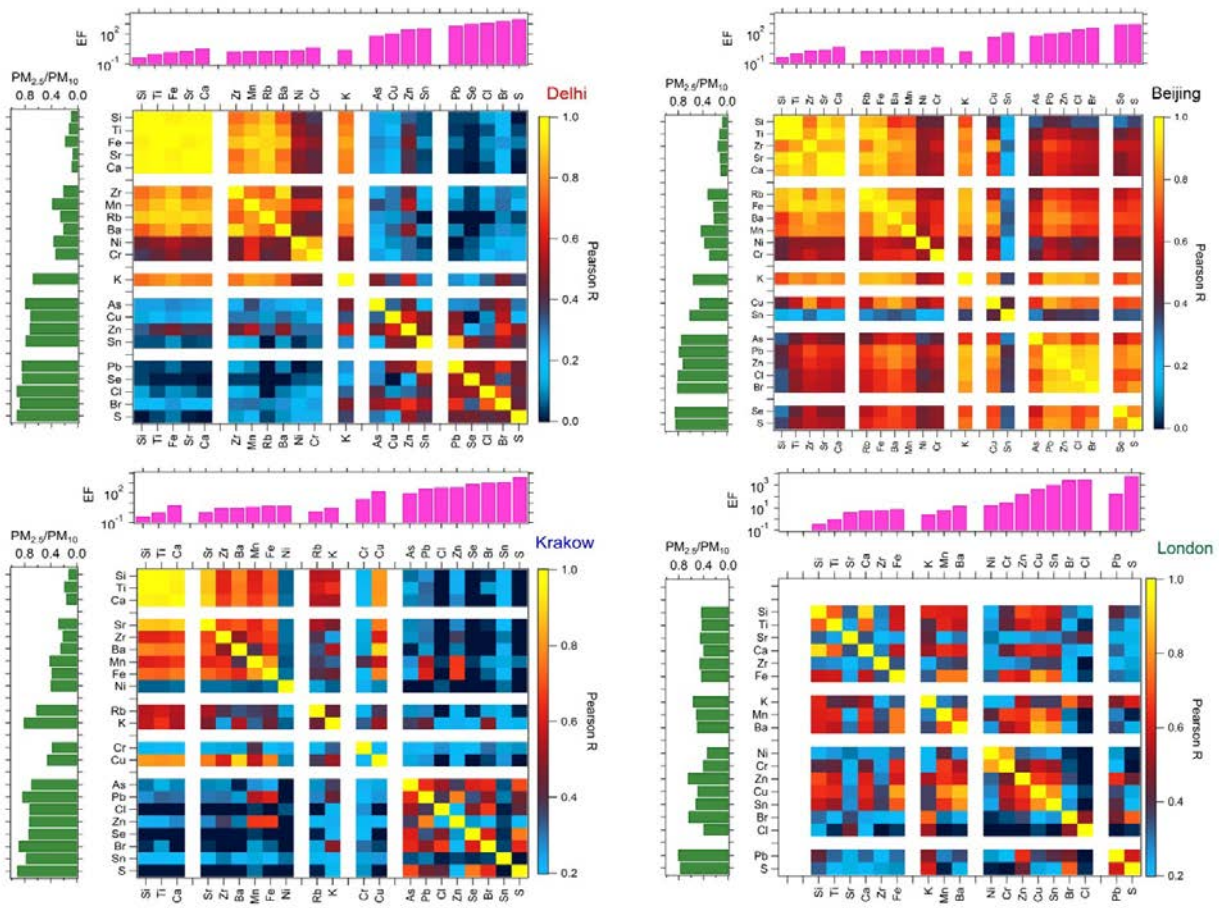


Figure S10: Correlation (Pearson's *R*) matrix of measured elements in elemental PM_{10} at all four sites (white color represents gap between each group elements). Elements are sorted by group along each axis. Note that in London, Group 1 (represented as white gap) is absent as well as Rb, As and Se were not measured. London measurements should be interpreted with caution due to uncertainties in the low size cut-off of the RDI, as discussed in Section 2.2.

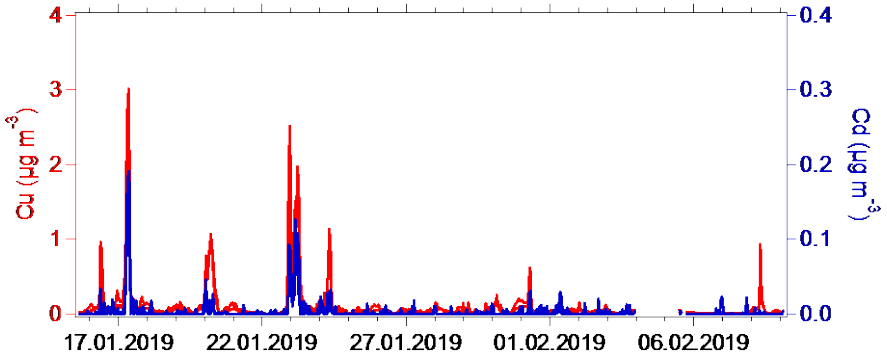


Figure S11: Time series of $PM_{2.5}$ Cu and Cd in Delhi.

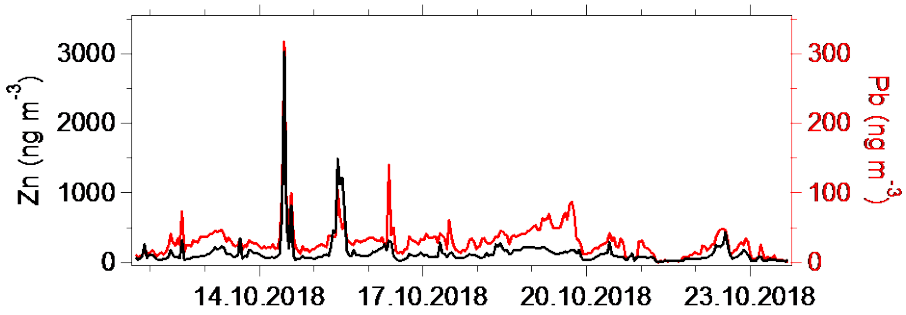


Figure S12: Time series of $PM_{2.5}$ Zn and Pb in Krakow.

Table S1: Xact 625i minimum detection limits (MDL) of elements with 30 min and 60 min time resolution. The percentage of data points below MDL is reported for both sizes in Delhi, Beijing and Krakow.

Element	Xact MDL		Data points below MDL (%)					
	60 min ($ng\ m^{-3}$)	30 min ($ng\ m^{-3}$)	Delhi		Beijing		Krakow	
			PM_{10el}	$PM_{2.5el}$	PM_{10el}	$PM_{2.5el}$	PM_{10el}	$PM_{2.5el}$
Al	170	500	18.8	61	25	86	87	99
Si	31	89	0	3.52	0	2.43	12.1	56
P	9.0	26	100	100	100	100	100	100
S	5.5	15.7	0	0	0	0	0	0
Cl	3.0	8.6	0	0	0	0	0	0
K	2.0	5.8	0	0	0	0	0	0
Ca	0.52	1.50	0	0	0	0.24	0	0
Ti	0.28	0.79	0	0	0	0.24	0.35	7.109
V	0.21	0.60	4.9	45	12.6	86	85	95
Cr	0.20	0.58	0	10.8	0.49	16.3	10.6	17.0
Mn	0.25	0.71	0	0	0	0	5.3	11.0
Fe	0.30	0.85	0	0	0	0.49	0	0
Co	0.24	0.68	99	97	93	98	99	100
Ni	0.17	0.47	3.5	27	5.8	30	39	59
Cu	0.14	0.39	0	0	0	0	0.35	0.35
Zn	0.12	0.33	0	0	0	0.24	0	0
Ga	0.10	0.29	100	100	68	84	100	99
Ge	0.10	0.28	100	100	82	81	98	98
As	0.11	0.31	0.20	100	0.73	4.1	6.4	10.3
Se	0.14	0.40	6.4	7.0	40	45	59	60
Br	0.18	0.52	0	0	0	0.73	0	0.35
Rb	0.33	0.95	10.9	58	9.0	47	66	87
Sr	0.38	1.10	2.9	56	0	43	46	92
Y	0.48	1.40	95	98	86	99	98	99
Zr	0.57	1.60	7.0	46	5.6	47	41	72
Cd	4.4	12.4	93	95	100	99	99	100
In	5.4	15.4	93	92	97	98	100	99
Sn	7.1	20	50	54	70	79	65	70
Sb	9.0	26	75	77	90	95	89	90
Ba	0.67	1.90	5.1	37	0.50	25	16.0	54
Hg	0.21	0.60	100	100	100	100	100	100
Tl	0.20	0.57	95	99	99	99	100	100
Pb	0.22	0.63	0	0	0	0.49	1.77	0.71
Bi	0.23	0.64	100	100	94	95	100	100

Table S2: Means, medians and 25–75th percentiles of PM_{10el} and $PM_{2.5el}$ concentrations (ng m^{-3}) in Delhi, Beijing, Krakow and London (*BDL: below detection limit).

Element	PM_{10el}				$PM_{2.5el}$			
	mean	median	q25	q75	mean	median	q25	q75
Delhi								
Al	3100	2600	920	4600	740	199	BDL	1060
Si	12200	11000	7400	16100	1240	1180	690	1720
S	7700	6400	4200	10300	7100	6100	3900	9400
Cl	20000	13100	5100	26000	19100	12200	4500	25000
K	3100	2600	1810	3800	2100	1740	1100	2700
Ca	5400	4700	3100	7200	490	410	260	640
Ti	260	240	163	340	35	31	20	45
V	5.9	5.1	3.1	8.3	1.29	0.78	BDL	1.96
Cr	17.4	12.2	7.2	24	9.1	3.5	1.42	8.5
Mn	79	64	42	100	32	21	12.6	38
Fe	3300	2900	1990	4300	650	500	360	720
Ni	4.9	3.9	2.4	6.5	2.3	1.12	0.40	2.3
Cu	128	48	25	91	96	27	13.0	54
Zn	780	610	380	1020	580	440	260	760
As	13.6	11.7	7.3	17.1	11.5	10.0	6.1	15.2
Se	3.9	3.0	1.50	4.6	3.5	2.8	1.42	4.2
Br	124	77	39	160	111	66	35	147
Rb	7.2	6.2	2.9	10.2	1.41	0.58	BDL	2.2
Sr	26	23	13.8	35	1.47	0.80	BDL	2.3
Zr	13.5	9.9	5.3	18.1	2.6	1.89	0.57	3.6
Sn	30	20	12.2	35	27	18.0	9.7	32
Sb	24	14.1	3.7	27	18.4	12.8	2.2	25
Ba	59	43	20	78	11.8	4.8	0.16	15.0
Pb	480	220	106	420	420	181	89	360
Beijing								
Al	1520	1160	184	2500	68	BDL	BDL	BDL
Si	6100	5600	2900	7900	510	400	163	650
S	1220	710	440	1350	1080	580	380	1160
Cl	2100	680	200	3100	1840	480	134	2700
K	960	730	320	1530	550	260	100	910
Ca	3600	3200	1500	5200	410	340	174	590
Ti	141	128	68	195	18.8	15.6	7.0	26
V	1.98	1.54	0.65	2.7	0.11	BDL	BDL	BDL
Cr	8.2	7.2	3.1	11.6	2.4	1.73	0.41	3.9
Mn	65	56	25	95	29	20	6.4	44.2
Fe	2200	1950	980	3200	520	390	165	750
Ni	2.4	2.0	0.88	3.4	0.86	0.60	0.10	1.29
Cu	28	22	8.7	43	13.2	9.2	3.2	20
Zn	165	94	36	260	124	69	22	200
As	6.2	2.8	1.22	8.1	5.2	1.90	0.76	7.0
Se	1.7	0.36	BDL	2.5	1.67	0.25	BDL	2.4
Br	14.0	5.1	2.1	21	12.5	5.08	1.62	18.1
Rb	3.2	2.7	1.13	4.8	0.85	0.42	BDL	1.3
Sr	15.1	13.5	6.4	22	1.50	0.64	BDL	2.5
Zr	8.1	6.9	2.5	11.7	1.10	0.66	0.05	1.7
Sn	5.3	4.1	0.83	7.8	4.2	2.8	BDL	6.2
Sb	2.7	0.03	BDL	4.7	1.89	BDL	BDL	2.6
Ba	33	28	10.8	50	6.9	4.0	0.68	11.1
Pb	39	20	7.4	63	33	15.2	5.2	55

Element	PM_{10el}				$PM_{2.5el}$			
	mean	median	q25	q75	mean	median	q25	q75
Krakow								
Al	146	BDL	BDL	BDL	28	BDL	BDL	BDL
Si	1980	1460	380	2900	220	70	14.2	310
S	1780	1190	870	1930	1660	1100	800	1830
Cl	880	630	290	1100	690	430	195	860
K	650	640	440	880	530	510	340	720
Ca	1460	1060	460	1970	230	130	65	330
Ti	44	37	18.1	65	8.6	6.7	3.2	12.6
V	0.25	BDL	BDL	0.17	0.10	BDL	BDL	BDL
Cr	14.9	7.7	2.7	17.6	14.7	3.8	1.01	11.9
Mn	28	22	9.6	36.5	15.2	8.1	4.0	17.3
Fe	1830	1480	710	2400	720	520	250	880
Ni	1.73	0.73	0.24	1.72	2.1	0.36	BDL	0.94
Cu	29	22	12.0	41	14.0	10.9	5.9	19.2
Zn	168	129	77	191	140	91	61	150
As	3.3	3.1	1.95	4.4	2.8	2.5	1.19	3.6
Se	0.50	0.23	BDL	0.79	0.50	0.27	BDL	0.74
Br	12.2	10.6	6.2	15.3	11.7	9.8	5.8	15.0
Rb	0.79	0.50	BDL	1.30	0.34	BDL	BDL	0.55
Sr	2.4	1.39	BDL	3.66	0.31	BDL	BDL	0.16
Zr	4.3	2.3	0.77	5.61	1.04	0.66	BDL	1.71
Sn	17.4	15.5	7.9	25	14.3	12.1	4.3	23
Sb	10.8	8.7	1.19	16.7	10.0	6.4	BDL	17.1
Ba	14.3	10.5	2.1	20	3.4	1.59	BDL	4.9
Pb	33	31	18.2	42	30	26	15.7	37
London								
Al	96	82	60	121	46	43	29	55
Si	174	140	77	220	76	58	36	104
S	290	210	136	320	230	139	81	260
Cl	910	630	230	1390	360	162	57	480
K	52	48	30	69	30	26	15.6	39
Ca	154	114	72	195	62	46	27	79
Ti	4.8	3.4	1.87	6.2	2.1	1.52	0.75	2.8
V	1.18	0.83	0.52	1.42	0.56	0.43	0.25	0.72
Cr	1.97	1.37	0.72	2.5	0.81	0.62	0.31	1.03
Mn	4.9	3.8	2.3	5.8	2.5	1.96	0.98	3.0
Fe	290	220	130	360	129	92	52	161
Ni	0.62	0.35	0.18	0.70	0.21	0.16	0.08	0.28
Cu	9.8	7.2	4.0	12.2	4.9	3.1	1.84	6.0
Zn	8.2	6.0	2.7	11.3	5.4	3.5	1.69	7.9
Br	3.6	3.2	1.76	4.9	2.3	1.73	1.03	2.8
Sr	0.92	0.75	0.47	1.16	0.42	0.34	0.21	0.53
Zr	0.91	0.46	0.20	1.07	0.44	0.26	0.12	0.52
Sn	1.53	1.07	0.52	1.87	0.81	0.55	0.27	1.04
Sb	1.18	0.84	0.43	1.44	0.63	0.42	0.22	0.81
Ba	7.3	4.9	2.7	8.6	3.7	2.4	1.33	4.6
Pb	2.3	1.31	0.63	2.8	1.89	1.0	0.45	2.3

Table S3: Relative fractions (%) of elements in both sizes for all four sites.

Element	Delhi		Beijing		Krakow		London	
	PM_{10el}	$PM_{2.5el}$	PM_{10el}	$PM_{2.5el}$	PM_{10el}	$PM_{2.5el}$	PM_{10el}	$PM_{2.5el}$
Si	23	3.9	36	9.9	22	5.1	9.1	8.4
S	14.2	22	7.3	21	20	39	15.2	25
Cl	38	60	12.5	36	9.8	16	47	40
K	5.7	6.6	5.8	10.6	7.3	12.4	2.7	3.3
Ca	10.0	1.55	22	8.0	16.3	5.4	8.0	6.8
Ti	0.49	0.11	0.85	0.36	0.49	0.20	0.25	0.23
Cr	0.03	0.03	0.05	0.05	0.17	0.34	0.10	0.09
Mn	0.15	0.10	0.39	0.56	0.32	0.35	0.26	0.28
Fe	6.2	2.0	13.2	10.0	21	16.7	15.2	14.1
Ni	0.01	0.01	0.01	0.02	0.02	0.05	0.03	0.02
Cu	0.24	0.30	0.17	0.26	0.33	0.32	0.51	0.53
Zn	1.45	1.81	0.99	2.4	1.88	3.2	0.43	0.59
Br	0.23	0.35	0.08	0.24	0.14	0.27	0.19	0.25
Sr	0.05	0	0.09	0.03	0.03	0.01	0.05	0.05
Zr	0.03	0.01	0.05	0.02	0.05	0.02	0.05	0.05
Sn	0.06	0.08	0.03	0.08	0.19	0.33	0.08	0.09
Ba	0.11	0.04	0.20	0.13	0.16	0.08	0.38	0.40
Pb	0.88	1.32	0.23	0.63	0.36	0.68	0.12	0.21