Supplementary Information

Seasonal Variability of PM$_{2.5}$ Composition and Sources in the Klang Valley Urban-Industrial Environment

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Experimental quality assurance and quality control (QA/QC)

For filter sampling, the elapsed time indicator (ETI) as well as the flow/pressure recorder chart was operated and checked daily to ensure the quality of sampling. Filter weighing was repeated three times and the average of those three were used for calculation. $PM_{2.5}$ mass were corrected by deducting the monthly filter blank (FB).

Anion and cation analysis: Using ion chromatography (IC), it was ensured that all ion regression coefficients were better than 0.999 before the sample analysis was begun and that every 15th sample, the QA/QC samples were run, i.e. the ultra-pure water (UPW) and 1 ppm standard prepared the same manner as the samples. These were used as validation tests for the method and the recovery purpose. All lab ware used in the ion analyses was Class A glassware.

Trace element analysis: Two sets of solutions were prepared for two modes of inductively coupled plasma mass spectrometry (ICPMS) analysis as follows: solution (1) 50 mL stock of concentrated solution for lower weight elements; and solution (2) further diluted 1 : 4 (50 mL concentrated: UPW) for heavier weight elements. Four point calibration curves were performed for each mode of analysis as follows: mode (1) 10, 20, 30 and 50 ppb for Ag, As, Cd, Cr, Li, Be, Bi, Cs, Co, Cu,Ga, Mn, Ni, Rb, Se, Sr, U and V; and mode (2) 125, 250, 500 and 1000 ppb for Al, Ba, Fe, Pb and Zn. It was ensured that all element regression coefficients were better than 0.999 before the sample analysis was begun and that every 15th sample, the QA/QC were samples run, i.e. the UPW, 1 ppm multi-element standard and standard reference material (SRM1648a). The reference material was 10 mg of SRM1648a Urban Particulate Matter obtained from NIST (National Institute of Standards and Technology, MD, USA) while 1 ppm Multi-Element Calibration Standard 3 (Perkin Elmer Pure Plus, Perkin-Elmer; USA) both prepared the same manner as the samples to test the recovery and validation of the method. All lab ware used for trace element analyses was Teflon material, except for the syringes.

1 Table S1. Descriptive statistics of experimental data (meteorological, gaseous and ion parameters); unit: mean ± std (min - max). Remarks:

2 ud = undetected (below detection limit); SIA = secondary inorganic aerosol; NR = neutralisation ratio = $[NH_4^+] / ([SO_4^{2-}] + [NO_3^-])$, (eq m^{-3})

3 (Squizzato et al., 2013); SO_2 gas was converted from ppm to µg m^{-3} assuming 1 ppm = 2619 µg m^{-3} (25 °C,1 atm).

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Elements	Unit	ANNUAL 5 Aug 2011 - 18 July 2012 n = 81	SW 15 May - 14 Sept n = 29	INT.2 15 Sept - 30 Oct n = 7
API	-	50 ± 16 (29 - 127)	60 ± 21 (36 - 127)	49 ± 6 (40 - 59)
T	°C	28.5 ± 1.19 (26.1 - 31.6)	28.9 ± 1.36 (26.4 - 31.6)	28.5 ± 1.20 (27.1 - 30.4)
RH	%	71.2 ± 7.91 (50.4 - 86.7)	68.2 ± 9.22 (50.4 - 86. 7)	72.9 ± 8.50 (59.7 - 82.7)
WS	ms^{-1}	1.29 ± 0.194 (0.873 - 1.77)	1.39 ± 0.187 (0.966 - 1.77)	1.25 ± 0.198 (1.01 - 1.53)
WD	Degree	129 ± 31.6 (23.1 - 208)	123 ± 38.0 (23.1 - 205)	128 ± 22.0 (100 - 167)
Rainfall	mm	10.4 ± 17.5 (0.000 - 85.4)	6.27 ± 10.6 (0.000 - 34.2)	8.46 ± 16.9 (0.000 - 45.4)
CO	ppm	1.29 ± 0.31 (0.61 - 2.16)	1.26 ± 0.32 (0.61 - 1.99)	1.43 ± 0.32 (1.10 - 1.93)
O_3	ppm	0.012 ± 0.006 (0.000 - 0.029)	0.010 ± 0.007 (0.000 - 0.025)	0.017 ± 0.008 (0.010 - 0.029)
SO_2	ppm	0.003 ± 0.001 (0.001 - 0.008)	0.004 ± 0.002 (0.001 - 0.008)	0.004 ± 0.001 (0.002 - 0.005)
NO_X	ppm	0.062 ± 0.013 (0.028 - 0.109)	0.057 ± 0.012 (0.028 - 0.076)	0.072 ± 0.013 (0.059 - 0.091)
NO	ppm	0.030 ± 0.010 (0.008 - 0.067)	0.025 ± 0.008 (0.008 - 0.041)	0.033 ± 0.008 (0.025 - 0.047)
NO_2	ppm	0.032 ± 0.007 (0.016 - 0.049)	0.032 ± 0.007 (0.016 - 0.048)	0.038 ± 0.006 (0.034 - 0.049)
SO_4^{2-}	µg m^{-3}	1.3 ± 0.88	1.8 ± 1.2	1.6 ± 0.78
ss-SO_4^{2-}	µg m^{-3}	0.076 ± 0.090	0.060 ± 0.023	0.022 ± 0.0079
nss-SO_4^{2-}	µg m^{-3}	1.3 ± 0.90	1.8 ± 1.2	1.61 ± 0.79
NO_3^-	µg m^{-3}	0.21 ± 0.13	0.19 ± 0.077	0.29 ± 0.22
NH_4^+	µg m^{-3}	0.99 ± 0.85	1.5 ± 1.2	1.00 ± 0.64
SIA	µg m^{-3}	2.4 ± 1.7	3.3 ± 2.3	2.8 ± 1.6
SIA/$PM_{2.5}$	%	8.5 ± 3.0	8.7 ± 3.4	9.6 ± 3.0
NR	-	0.26	0.31	0.21
SO_4^2-SO_2	µg m^{-3}	1.3 - 8.2	1.8 - 9.5	1.6 - 10

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Continuation of Table S1 (2)

Elements	Unit	NE 1 Nov - 14 Mar n = 35	INT.1 15 Mar - 14 May n = 10	HAZE n = 11
API	-	44 ± 8 (29 - 58)	45 ± 9 (33 - 58)	78 ± 22 (49 - 127)
T	^{o}C	28.1 ± 1.02 (26.1 - 30.4)	28.8 ± 0.78 (27.5 - 30.2)	29.5 ± 1.33 (26.7 - 31.6)
RH	%	73.6 ± 6.79 (56.5 - 85.5)	70.5 ± 4.01 (65.1 – 77.0)	63.0 ± 9.91 (50.4 - 81.6)
WS	ms^{-1}	1.20 ± 0.167 (0.873 - 1.46)	1.32 ± 0.18 (1.08 - 1.71)	1.49 ± 0.138 (1.27 - 1.70)
WD	Degree	132 ± 31.2 (83.2- 208)	128 ± 25.1 (103 - 178)	103 ± 33.2 (23.1 - 137)
Rainfall	mm	15.1 ± 22.7 (0.000 - 85.4)	7.04 ± 9.69 (0.000 – 24.0)	2.28 ± 5.18 (0.000 - 15.8)
CO	ppm	1.29 ± 0.30 (0.92 - 2.16)	1.32 ± 0.28 (0.84 - 1.75)	1.45 ± 0.31 (0.89 - 1.99)
O_3	ppm	0.013 ± 0.005 (0.004 - 0.025)	0.014 ± 0.004 (0.003 - 0.018)	0.016 ± 0.004 (0.011 - 0.025)
SO_2	ppm	0.003 ± 0.001 (0.001 – 0.005)	0.003 ± 0.001 (0.001 – 0.005)	0.003 ± 0.001 (0.001 - 0.005)
NO_X	ppm	0.065 ± 0.014 (0.044 - 0.109)	0.059 ± 0.011 (0.039 - 0.072)	0.057 ± 0.013 (0.028 - 0.074)
NO	ppm	0.034 ± 0.010 (0.021 - 0.067)	0.029 ± 0.008 (0.013 - 0.039)	0.022 ± 0.008 (0.008 - 0.038)
NO_2	ppm	0.031 ± 0.006 (0.021 - 0.049)	0.030 ± 0.008 (0.018 - 0.044)	0.035 ± 0.008 (0.020 - 0.048)
SO_4^{2-}	µg m^{-3}	0.98 ± 0.41	1.1 ± 0.70	2.4 ± 1.2
ss-SO_4^{2-}	µg m^{-3}	0.048 ± 0.029	0.16 ± 0.15	0.059 ± 0.00
nss-SO_4^{2-}	µg m^{-3}	0.95 ± 0.42	0.90 ± 0.60	2.4 ± 1.2
NO_3^-	µg m^{-3}	0.19 ± 0.13	0.27 ± 0.13	0.22 ± 0.14
NH_4^+	µg m^{-3}	0.65 ± 0.34	0.82 ± 0.49	2.2 ± 1.5
SIA	µg m^{-3}	1.7 ± 0.81	2.0 ± 0.88	4.7 ± 2.6
SIA/$PM_{2.5}$	%	8.2 ± 2.8	8.4 ± 2.3	7.5 ± 2.7
NR	-	0.22	0.17	0.35
SO_4^2-SO_2	µg m^{-3}	0.98 - 6.6	1.1 – 8.0	2.4 - 9.1

1   Table S2. Statistical compositions of $PM_{2.5}$ during the one-year period including % recovery

2   for anions and cations using 1 ppm standard of single cation/anion standards; and for trace

3   element using standard reference material (SRM1648a). Remarks: MDL = method detection

4   limit

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	Elements	Unit	ANNUAL n = 81					SW n = 29				
			min	max	med	avg	std	min	max	med	avg	std
1	$PM_{2.5}$	$\mu g/m^3$	6	118	23	28	17	14	118	29	38	24
2	F^-	$\mu g/m^3$	< MDL	0.018	0.002	0.003	0.004	< MDL	0.018	0.003	0.006	0.005
3	Cl^-	$\mu g/m^3$	< MDL	0.141	0.016	0.026	0.026	0.007	0.085	0.018	0.024	0.017
4	NO_2^-	$\mu g/m^3$	< MDL	0.009	0.004	0.004	0.002	< MDL	0.005	0.003	0.003	0.001
5	Br^-	$\mu g/m^3$	0.008	0.030	0.021	0.019	0.008	0.009	0.021	0.015	0.015	0.009
6	NO_3^-	$\mu g/m^3$	0.076	0.616	0.177	0.213	0.127	0.092	0.372	0.174	0.191	0.077
7	PO_4^{3-}	$\mu g/m^3$	< MDL	0.205	0.072	0.079	0.063	< MDL	0.074	0.048	0.054	0.017
8	SO_4^{2-}	$\mu g/m^3$	0.139	4.155	1.093	1.328	0.885	0.368	4.155	1.385	1.769	1.158
9	Na^+	$\mu g/m^3$	< MDL	2.113	0.218	0.300	0.357	0.113	0.385	0.201	0.236	0.093
10	NH_4^+	$\mu g/m^3$	0.085	5.440	0.691	0.987	0.852	0.193	5.440	1.054	1.453	1.180
11	K^+	$\mu g/m^3$	0.077	0.906	0.207	0.253	0.144	0.118	0.906	0.336	0.343	0.182
12	Ca^{2+}	$\mu g/m^3$	0.060	0.785	0.159	0.180	0.100	0.069	0.785	0.159	0.198	0.145
13	Mg^{2+}	$\mu g/m^3$	< MDL	0.391	0.024	0.033	0.044	0.015	0.115	0.029	0.035	0.021
14	Al	$\mu g/m^3$	< MDL	7.740	0.712	1.063	1.226	< MDL	7.740	0.974	1.723	1.973
15	Ba	$\mu g/m^3$	0.011	0.115	0.030	0.036	0.019	0.014	0.115	0.027	0.040	0.025
16	Fe	$\mu g/m^3$	< MDL	5.245	0.879	1.000	0.706	< MDL	5.245	0.940	1.236	1.108
17	Pb	$\mu g/m^3$	0.015	0.787	0.077	0.108	0.116	0.015	0.146	0.060	0.069	0.033
18	Zn	$\mu g/m^3$	< MDL	0.519	0.173	0.189	0.104	< MDL	0.328	0.139	0.148	0.075
19	Ag	ng/m^3	< MDL	0.360	0.112	0.145	0.104	< MDL	0.221	0.152	0.152	0.097
20	As	ng/m^3	< MDL	1.196	0.358	0.380	0.152	< MDL	0.552	0.328	0.320	0.112
21	Cd	ng/m^3	0.031	1.002	0.092	0.125	0.136	0.042	0.197	0.084	0.093	0.045
22	Cr	ng/m^3	< MDL	32.198	2.055	3.936	6.364	< MDL	32.198	1.386	6.227	10.279
23	Li	ng/m^3	< MDL	0.063	0.011	0.013	0.010	< MDL	0.063	0.010	0.014	0.015
24	Be	ng/m^3	< MDL	0.016	0.001	0.002	0.004	< MDL	0.007	0.001	0.002	0.002
25	Bi	ng/m^3	0.010	0.098	0.028	0.030	0.014	0.010	0.055	0.026	0.027	0.009
26	Cs	ng/m^3	0.009	0.124	0.030	0.035	0.021	0.012	0.096	0.035	0.039	0.022
27	Co	ng/m^3	0.004	0.117	0.012	0.015	0.014	0.004	0.117	0.011	0.018	0.022
28	Cu	ng/m^3	0.290	10.984	2.083	2.740	1.890	0.290	10.984	1.836	2.818	2.267
29	Ga	ng/m^3	0.020	0.135	0.042	0.048	0.024	0.021	0.135	0.054	0.057	0.031
30	Mn	ng/m^3	< MDL	3.841	0.822	0.934	0.643	< MDL	3.841	0.782	0.986	0.765
31	Ni	ng/m^3	< MDL	3.126	0.391	0.480	0.469	0.137	1.960	0.430	0.538	0.358
32	Rb	ng/m^3	0.349	3.185	0.865	1.008	0.501	0.443	3.185	1.188	1.251	0.653
33	Se	ng/m^3	< MDL	0.386	0.087	0.107	0.071	< MDL	0.386	0.126	0.132	0.096
34	Sr	ng/m^3	< MDL	1.192	0.201	0.265	0.223	< MDL	1.192	0.285	0.342	0.298
35	U	ng/m^3	< MDL	0.021	0.002	0.003	0.003	< MDL	0.010	0.002	0.003	0.002
36	V	ng/m^3	< MDL	2.743	0.790	0.915	0.556	< MDL	2.640	0.836	0.951	0.595
37	BC	$\mu g/m^3$	2.548	5.548	4.248	4.155	0.642	2.974	5.167	4.248	4.155	0.581

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4 Continuation of Table S2 (2)
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	Elements	Unit	INT.2					NE				
			n = 7					n = 35				
			min	max	med	avg	std	min	max	med	avg	std
1	PM$_{2.5}$	µg/m^3	10	50	27	29	12	6	35	20	21	6
2	F$^-$	µg/m^3	< MDL	0.015	0.002	0.005	0.006	< MDL	0.006	0.001	0.002	0.001
3	Cl$^-$	µg/m^3	< MDL	0.077	0.012	0.027	0.031	< MDL	0.141	0.014	0.027	0.032
4	NO$_2^-$	µg/m^3	< MDL	0.009	0.006	0.006	0.002	< MDL	0.007	0.004	0.004	0.001
5	Br$^-$	µg/m^3	n/d	n/d	n/d	n/d	n/d	0.008	0.028	0.016	0.017	0.008
6	NO$_3^-$	µg/m^3	0.114	0.608	0.219	0.290	0.218	0.076	0.616	0.141	0.188	0.126
7	PO$_4^{3-}$	µg/m^3	< MDL	0.020	0.014	0.013	0.007	0.010	0.205	0.165	0.113	0.082
8	SO$_4^{2-}$	µg/m^3	0.756	3.104	1.399	1.618	0.782	0.139	1.673	0.942	0.981	0.414
9	Na$^+$	µg/m^3	< MDL	0.108	0.085	0.085	0.031	< MDL	0.399	0.134	0.189	0.115
10	NH$_4^+$	µg/m^3	0.448	2.331	0.741	1.004	0.636	0.085	1.680	0.597	0.645	0.336
11	K$^+$	µg/m^3	0.104	0.482	0.240	0.258	0.124	0.077	0.357	0.194	0.195	0.076
12	Ca^{2+}	µg/m^3	0.110	0.227	0.164	0.171	0.044	0.061	0.340	0.151	0.161	0.064
13	Mg^{2+}	µg/m^3	0.019	0.027	0.024	0.024	0.002	< MDL	0.036	0.018	0.019	0.008
14	Al	µg/m^3	< MDL	2.263	1.268	1.121	0.897	< MDL	1.757	0.721	0.806	0.387
15	Ba	µg/m^3	0.011	0.080	0.037	0.042	0.021	0.017	0.093	0.029	0.032	0.014
16	Fe	µg/m^3	0.480	1.649	0.989	1.069	0.481	0.358	1.221	0.796	0.811	0.222
17	Pb	µg/m^3	0.041	0.787	0.126	0.220	0.265	0.032	0.661	0.084	0.118	0.114
18	Zn	µg/m^3	0.112	0.515	0.330	0.300	0.152	< MDL	0.519	0.187	0.212	0.099
19	Ag	ng/m^3	n/d	n/d	n/d	n/d	n/d	< MDL	0.360	0.095	0.139	0.116
20	As	ng/m^3	0.235	0.638	0.445	0.437	0.131	0.212	1.196	0.421	0.433	0.177
21	Cd	ng/m^3	0.031	1.002	0.144	0.238	0.340	0.041	0.630	0.099	0.141	0.127
22	Cr	ng/m^3	< MDL	4.081	2.392	2.500	1.295	< MDL	5.034	1.560	1.924	1.745
23	Li	ng/m^3	< MDL	0.023	0.011	0.012	0.009	< MDL	0.032	0.011	0.012	0.007
24	Be	ng/m^3	0.001	0.002	0.001	0.001	0.001	< MDL	0.016	0.000	0.004	0.008
25	Bi	ng/m^3	0.015	0.055	0.038	0.039	0.014	0.012	0.098	0.029	0.032	0.018
26	Cs	ng/m^3	0.015	0.059	0.041	0.037	0.017	0.009	0.124	0.030	0.035	0.024
27	Co	ng/m^3	0.006	0.024	0.017	0.017	0.007	0.005	0.044	0.011	0.012	0.007
28	Cu	ng/m^3	0.593	4.718	1.999	2.542	1.556	0.969	10.188	2.223	2.835	1.845
29	Ga	ng/m^3	0.020	0.104	0.057	0.059	0.027	0.022	0.106	0.041	0.042	0.016
30	Mn	ng/m^3	< MDL	2.436	1.409	1.339	0.793	< MDL	1.438	0.862	0.811	0.324
31	Ni	ng/m^3	0.358	0.747	0.544	0.550	0.147	< MDL	3.126	0.286	0.427	0.615
32	Rb	ng/m^3	0.575	1.756	1.180	1.149	0.467	0.349	1.427	0.827	0.849	0.305
33	Se	ng/m^3	0.049	0.192	0.112	0.115	0.052	< MDL	0.242	0.096	0.098	0.051
34	Sr	ng/m^3	< MDL	1.070	0.242	0.338	0.331	< MDL	0.542	0.199	0.227	0.111
35	U	ng/m^3	< MDL	0.007	0.004	0.003	0.002	< MDL	0.021	0.002	0.003	0.003
36	V	ng/m^3	0.790	2.024	1.314	1.299	0.421	0.157	1.470	0.654	0.731	0.366
37	BC	µg/m^3	2.548	5.548	4.480	4.333	0.952	3.056	5.524	4.213	4.201	0.615

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3 Continuation of Table S2 (3)

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	Elements	Unit	INT.1 n = 10					HAZE n = 11					% Recovery
			min	max	med	avg	std	min	max	med	avg	std	
1	$PM_{2.5}$	$\mu g/m^3$	14	39	22	23	8	40	118	54	61	24	
2	F^-	$\mu g/m^3$	< MDL	0.011	0.004	0.004	0.004	0.003	0.018	0.011	0.011	0.006	105
3	Cl^-	$\mu g/m^3$	0.007	0.072	0.018	0.027	0.021	0.007	0.085	0.021	0.030	0.022	112
4	NO_2^-	$\mu g/m^3$	< MDL	0.003	0.003	0.003	0.000	0.003	0.004	0.004	0.004	0.001	107
5	Br^-	$\mu g/m^3$	0.025	0.030	0.028	0.028	0.002	n/d	n/d	n/d	n/d	n/d	99
6	NO_3^-	$\mu g/m^3$	0.170	0.559	0.215	0.267	0.125	0.092	0.372	0.194	0.219	0.142	131
7	PO_4^{3-}	$\mu g/m^3$	< MDL	0.114	0.075	0.080	0.014	n/d	n/d	n/d	n/d	n/d	103
8	SO_4^{2-}	$\mu g/m^3$	0.269	2.631	0.949	1.057	0.698	0.719	4.155	2.276	2.403	1.235	106
9	Na^+	$\mu g/m^3$	0.343	2.113	0.367	0.637	0.611	0.233	0.233	0.233	0.233	0.000	110
10	NH_4^+	$\mu g/m^3$	0.245	1.591	0.769	0.821	0.485	0.541	5.440	1.999	2.211	1.468	86
11	K^+	$\mu g/m^3$	0.087	0.290	0.176	0.181	0.063	0.336	0.906	0.452	0.507	0.171	96
12	Ca^{2+}	$\mu g/m^3$	0.060	0.259	0.211	0.196	0.062	0.124	0.785	0.170	0.282	0.206	93
13	Mg^{2+}	$\mu g/m^3$	0.015	0.391	0.048	0.081	0.111	0.028	0.115	0.042	0.050	0.027	98
14	Al	$\mu g/m^3$	0.025	0.990	0.387	0.408	0.292	< MDL	7.740	2.050	2.788	2.564	36
15	Ba	$\mu g/m^3$	0.018	0.052	0.032	0.033	0.011	0.022	0.115	0.043	0.046	0.026	
16	Fe	$\mu g/m^3$	0.511	1.452	0.900	0.980	0.355	0.371	5.245	1.351	1.897	1.500	69
17	Pb	$\mu g/m^3$	0.045	0.293	0.090	0.108	0.069	0.059	0.146	0.095	0.094	0.030	101
18	Zn	$\mu g/m^3$	0.050	0.256	0.142	0.137	0.065	< MDL	0.328	0.185	0.175	0.088	69
19	Ag	ng/m^3	< MDL	0.249	0.176	0.161	0.096	n/d	n/d	n/d	n/d	n/d	74
20	As	ng/m^3	0.211	0.454	0.313	0.326	0.093	0.214	0.428	0.328	0.337	0.065	51
21	Cd	ng/m^3	0.034	0.135	0.075	0.080	0.033	0.051	0.197	0.139	0.130	0.047	47
22	Cr	ng/m^3	< MDL	10.545	2.865	3.370	3.063	< MDL	4.214	2.866	2.866	1.907	17
23	Li	ng/m^3	< MDL	0.030	0.014	0.015	0.006	0.002	0.063	0.010	0.021	0.021	
24	Be	ng/m^3	0.001	0.001	0.001	0.001	0.000	0.275	2.484	1.395	1.456	0.749	
25	Bi	ng/m^3	0.010	0.035	0.026	0.023	0.008	0.017	0.042	0.027	0.028	0.008	
26	Cs	ng/m^3	0.013	0.054	0.025	0.026	0.012	0.033	0.096	0.059	0.060	0.019	38
27	Co	ng/m^3	0.005	0.025	0.012	0.015	0.006	0.004	0.117	0.018	0.029	0.032	60
28	Cu	ng/m^3	1.468	4.383	2.040	2.323	1.002	1.517	6.840	3.264	3.491	1.962	65
29	Ga	ng/m^3	0.021	0.062	0.033	0.038	0.013	0.024	0.135	0.057	0.056	0.032	
30	Mn	ng/m^3	0.222	2.932	0.578	0.937	0.930	< MDL	3.841	1.113	1.388	1.064	72
31	Ni	ng/m^3	< MDL	0.928	0.402	0.456	0.283	0.196	1.960	0.425	0.588	0.496	65
32	Rb	ng/m^3	0.379	1.324	0.818	0.785	0.282	1.048	3.185	1.733	1.777	0.612	29
33	Se	ng/m^3	< MDL	0.087	0.058	0.055	0.026	0.066	0.386	0.203	0.207	0.085	42
34	Sr	ng/m^3	< MDL	0.228	0.133	0.132	0.058	< MDL	1.192	0.311	0.456	0.386	58
35	U	ng/m^3	0.001	0.004	0.002	0.002	0.001	0.515	9.890	2.730	3.820	2.829	
36	V	ng/m^3	0.240	2.743	1.035	1.186	0.832	0.612	2.640	0.841	1.024	0.574	64
37	BC	$\mu g/m^3$	3.091	4.799	3.591	3.865	0.676	4.202	5.167	4.529	4.611	0.332	

6

1   Table S3. The input variables during the execution of PMF 5.0 procedure for the apportionment of $PM_{2.5}$ sources

Sequence Number	Seed	Q robust	Q true	Q theory	Q true/Qexp	Extra Uncertainty (%)	Number of factor	Standard Error	R^2
1	1	1581.27	1581.27	1925	0.93843919	5	5	4.547911	0.887934
2	2	1581.27	1581.27	1925	0.93843919	5	5	4.548350	0.887933
3	3	1581.26	1581.26	1925	0.93843323	5	5	4.549417	0.887884
4	4	1581.27	1581.27	1925	0.93843919	5	5	4.547655	0.887952
5	5	1581.27	1581.27	1925	0.93843919	5	5	4.547842	0.887945
6	6	1581.26	1581.26	1925	0.93843323	5	5	4.548071	0.887947
7	7	1581.26	1581.26	1925	0.93843323	5	5	4.547759	0.887944
8	8	1581.27	1581.26	1925	0.93843323	5	5	4.548212	0.887924
9	9	1581.27	1581.27	1925	0.93843919	5	5	4.547940	0.887949

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Table S4. Seasonal results of Pearson correlation matrices between $PM_{2.5}$ mass, chemical mass closure (CMC) components and positive matrix factorisation (PMF) factors identified towards: a) meteorological parameters, and b) gaseous parameters. Remarks: BC = black carbon while PMF factors were: factor 1= combustion of engine oil; factor 2= mineral dust; factor 3= mixed secondary inorganic aerosol (SIA) and biomass burning; factor 4= mixed traffic and industrial; and factor 5= sea salt. For meteorological parameters, API is air pollution index; T = temperature; RH = relative humidity; WS = wind speed; and WD = wind direction.

8

a)

	Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
API	PM$_{2.5}$ mass	**0.763**	**0.748**	0.299	**0.473**	**0.705**	0.531
	CMC SO$_4^{2-}$	**0.629**	**0.565**	0.683	**0.469**	0.585	0.480
	CMC NO$_3^-$	-0.172	-0.350	0.370	0.079	0.623	-0.175
	CMC NH$_4^+$	**0.712**	**0.667**	0.750	**0.561**	0.460	**0.606**
	CMC Sea salt	-0.087	-0.294	0.214	-0.088	0.547	-0.102
	CMC Dust	**0.227**	0.220	-0.033	-0.101	0.302	-0.114
	CMC BC	**0.353**	**0.580**	0.046	0.234	**0.772**	0.274
	PMF Factor 1	0.133	-0.062	-0.084	**0.366**	-0.313	0.076
	PMF Factor 2	**0.354**	0.275	-0.041	-0.103	0.325	0.005
	PMF Factor 3	**0.725**	**0.716**	**0.767**	0.158	**0.817**	0.476
	PMF Factor 4	**-0.235**	0.081	-0.006	-0.169	0.416	0.014
	PMF Factor 5	0.011	-0.079	-0.279	-0.025	0.612	-0.124
T	PM$_{2.5}$ mass	**0.310**	0.236	0.572	0.201	0.030	-0.050
	CMC SO$_4^{2-}$	0.149	-0.063	0.089	**0.414**	0.112	-0.547
	CMC NO$_3^-$	0.052	0.194	0.521	-0.151	-0.097	0.384
	CMC NH$_4^+$	0.145	0.046	0.087	0.176	-0.542	-0.259
	CMC Sea salt	0.155	**0.407**	0.128	**0.379**	0.008	0.562
	CMC Dust	**0.576**	**0.641**	0.692	**0.581**	-0.216	**0.761**
	CMC BC	-0.023	0.188	0.689	**-0.421**	-0.079	0.489
	PMF Factor 1	0.063	0.022	-0.393	0.083	-0.110	0.086
	PMF Factor 2	**0.599**	**0.673**	0.703	**0.464**	0.734	**0.789**
	PMF Factor 3	**0.236**	0.229	0.342	0.177	-0.184	-0.094
	PMF Factor 4	-0.189	0.122	0.709	-0.280	0.403	0.089
	PMF Factor 5	0.213	0.313	0.140	0.163	-0.405	**0.687**
RH	PM$_{2.5}$ mass	**-0.314**	-0.252	-0.495	-0.174	0.152	0.108
	CMC SO$_4^{2-}$	-0.054	0.186	-0.007	**-0.352**	0.148	**0.646**
	CMC NO$_3^-$	-0.035	-0.358	-0.489	0.231	0.338	-0.534
	CMC NH$_4^+$	-0.112	0.011	-0.010	-0.113	0.504	0.332
	CMC Sea salt	-0.117	**-0.379**	-0.097	**-0.519**	0.227	-0.461
	CMC Dust	**-0.615**	**-0.716**	-0.697	**-0.543**	0.437	**-0.742**
	CMC BC	0.022	-0.189	-0.640	**0.396**	0.228	-0.472
	PMF Factor 1	0.056	0.186	0.439	-0.116	0.139	-0.039
	PMF Factor 2	**-0.652**	**-0.738**	-0.778	**-0.547**	-0.600	**-0.759**
	PMF Factor 3	-0.226	-0.192	-0.275	-0.111	0.053	0.229
	PMF Factor 4	0.212	-0.219	-0.655	0.334	-0.567	-0.205
	PMF Factor 5	-0.220	-0.353	-0.131	-0.219	0.600	-0.570

	Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
WS	PM$_{2.5}$ mass	**0.274**	0.164	0.245	-0.030	0.192	-0.446
	CMC SO$_4^{2-}$	0.174	-0.047	-0.125	0.224	0.208	**-0.692**
	CMC NO$_3^-$	-0.113	0.161	0.378	**-0.429**	-0.055	**0.719**
	CMC NH$_4^+$	0.140	0.015	-0.103	-0.034	-0.421	-0.530
	CMC Sea salt	0.013	0.007	-0.035	0.241	-0.195	0.111
	CMC Dust	**0.385**	**0.421**	0.588	**0.408**	-0.154	0.430
	CMC BC	-0.070	0.162	0.445	**-0.403**	-0.034	-0.060
	PMF Factor 1	0.084	-0.066	-0.599	0.107	0.557	0.104
	PMF Factor 2	**0.429**	**0.456**	0.875	**0.401**	0.249	0.494
	PMF Factor 3	0.213	0.107	0.182	-0.186	0.228	**-0.678**
	PMF Factor 4	**-0.391**	0.155	0.378	**-0.507**	0.628	0.138
	PMF Factor 5	0.132	0.048	0.128	0.144	-0.267	0.306
WD	PM$_{2.5}$ mass	-0.131	-0.181	0.409	0.056	0.047	0.413
	CMC SO$_4^{2-}$	-0.054	-0.006	0.047	0.083	-0.029	0.162
	CMC NO$_3^-$	0.002	0.170	0.471	-0.226	-0.124	0.451
	CMC NH$_4^+$	-0.105	-0.012	0.034	-0.029	-0.524	0.319
	CMC Sea salt	0.096	0.064	-0.190	0.309	0.145	-0.039
	CMC Dust	0.014	-0.085	0.625	**0.406**	**-0.699**	0.197
	CMC BC	-0.063	-0.130	0.579	-0.113	-0.172	0.381
	PMF Factor 1	0.205	**0.581**	0.129	-0.147	-0.15	**0.677**
	PMF Factor 2	-0.016	-0.011	0.354	**0.373**	-0.195	0.319
	PMF Factor 3	-0.161	-0.257	-0.045	0.144	0.633	0.259
	PMF Factor 4	0.163	-0.185	0.638	0.048	0.464	0.161
	PMF Factor 5	-0.067	-0.152	0.339	0.121	-0.312	-0.011
Rainfall	PM$_{2.5}$ mass	-0.212	-0.246	**-0.733**	-0.052	-0.051	-0.178
	CMC SO$_4^{2-}$	-0.136	-0.133	-0.422	0.025	0.001	0.236
	CMC NO$_3^-$	-0.030	-0.030	-0.414	0.037	0.079	-0.257
	CMC NH$_4^+$	-0.092	-0.188	-0.335	0.207	0.618	0.012
	CMC Sea salt	-0.106	-0.274	0.109	-0.093	-0.251	-0.213
	CMC Dust	**-0.228**	-0.353	-0.296	-0.265	0.551	-0.391
	CMC BC	-0.116	-0.141	**-0.893**	-0.029	0.027	-0.591
	PMF Factor 1	-0.037	-0.084	-0.291	0.191	0.530	0.064
	PMF Factor 2	**-0.257**	-0.184	-0.156	**-0.513**	-0.581	-0.399
	PMF Factor 3	-0.207	-0.227	-0.227	-0.265	0.047	-0.097
	PMF Factor 4	-0.110	-0.102	-0.743	-0.296	-0.524	-0.343
	PMF Factor 5	-0.050	-0.128	**-0.816**	0.144	0.338	-0.101

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Continuation of Table S4 (2)

b)

	Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE		Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
CO	$PM_{2.5}$ mass	**0.471**	**0.687**	0.713	**0.488**	**0.654**	**0.749**	NO_X	$PM_{2.5}$ mass	0.058	0.112	**0.800**	**0.380**	0.588	0.192
	CMC SO_4^{2-}	**0.455**	**0.579**	**0.793**	**0.428**	0.582	0.412		CMC SO_4^{2-}	0.050	0.038	0.709	0.180	0.492	-0.333
	CMC NO_3^-	0.171	-0.292	0.334	0.296	**0.687**	0.014		CMC NO_3^-	**0.284**	0.085	0.545	0.295	0.609	0.429
	CMC NH_4^+	**0.462**	**0.664**	0.711	**0.576**	0.177	**0.674**		CMC NH_4^+	0.050	0.127	0.663	**0.351**	0.226	0.030
	CMC Sea salt	0.020	-0.273	-0.085	**-0.482**	0.626	0.107		CMC Sea salt	-0.063	-0.04	0.005	**-0.527**	0.472	0.307
	CMC Dust	0.060	0.232	0.001	-0.285	0.331	0.205		CMC Dust	-0.019	0.244	0.364	**-0.419**	0.358	0.589
	CMC BC	**0.549**	**0.533**	0.554	**0.544**	**0.748**	**0.697**		CMC BC	**0.467**	0.122	0.642	**0.621**	**0.724**	0.588
	PMF Factor 1	0.206	0.134	0.722	**0.498**	-0.167	0.306		PMF Factor 1	0.170	0.218	0.482	**0.471**	-0.043	0.158
	PMF Factor 2	0.197	**0.455**	-0.637	-0.295	0.198	0.322		PMF Factor 2	0.122	**0.504**	-0.449	-0.280	0.081	**0.650**
	PMF Factor 3	**0.302**	**0.612**	0.420	-0.083	0.072	0.600		PMF Factor 3	-0.139	0.014	0.504	-0.220	0.000	0.120
	PMF Factor 4	0.199	0.157	0.577	0.040	-0.015	0.007		PMF Factor 4	**0.459**	0.186	**0.774**	0.219	-0.007	0.239
	PMF Factor 5	-0.025	-0.163	0.438	-0.167	**0.694**	0.010		PMF Factor 5	-0.122	0.029	0.180	-0.288	**0.643**	0.362
O_3	$PM_{2.5}$ mass	**0.298**	**0.535**	0.427	**0.433**	0.378	0.449	NO	$PM_{2.5}$ mass	**-0.262**	-0.309	0.701	0.086	-0.126	-0.285
	CMC SO_4^{2-}	0.198	0.191	0.674	**0.531**	0.239	**0.803**		CMC SO_4^{2-}	**-0.252**	-0.270	0.573	-0.088	-0.227	**-0.702**
	CMC NO_3^-	0.079	0.032	-0.055	-0.087	0.345	-0.413		CMC NO_3^-	0.185	0.177	0.353	0.126	-0.006	0.432
	CMC NH_4^+	**0.241**	0.343	0.553	**0.521**	0.627	**0.685**		CMC NH_4^+	**-0.243**	-0.285	0.491	0.053	0.134	-0.475
	CMC Sea salt	0.121	-0.014	-0.210	0.333	0.237	-0.332		CMC Sea salt	-0.062	0.215	-0.073	**-0.475**	0.013	0.434
	CMC Dust	0.115	0.193	-0.467	0.207	0.316	-0.532		CMC Dust	-0.124	0.159	0.225	**-0.469**	-0.013	**0.687**
	CMC BC	0.049	0.131	0.316	-0.263	0.454	-0.071		CMC BC	**0.237**	-0.207	0.583	**0.472**	0.142	0.274
	PMF Factor 1	**-0.287**	**-0.605**	**0.796**	0.118	0.040	0.358		PMF Factor 1	0.083	0.247	0.651	0.326	-0.246	0.050
	PMF Factor 2	-0.009	0.105	-0.832	-0.007	-0.434	-0.427		PMF Factor 2	-0.034	0.369	-0.629	-0.247	-0.360	**0.698**
	PMF Factor 3	**0.304**	**0.597**	0.204	**0.383**	0.703	0.538		PMF Factor 3	**-0.390**	-0.376	0.263	-0.343	-0.476	-0.282
	PMF Factor 4	0.035	0.024	0.188	**-0.351**	0.126	-0.122		PMF Factor 4	**0.428**	0.028	0.719	0.195	-0.370	0.239
	PMF Factor 5	0.147	0.113	0.638	0.305	0.509	-0.379		PMF Factor 5	-0.118	0.181	0.327	-0.325	0.188	0.589
SO_2	$PM_{2.5}$ mass	**0.324**	0.141	-0.250	**0.654**	0.627	0.445	NO_2	$PM_{2.5}$ mass	**0.473**	**0.528**	**0.851**	**0.711**	**0.874**	0.599
	CMC SO_4^{2-}	**0.345**	0.201	-0.559	**0.407**	**0.670**	-0.017		CMC SO_4^{2-}	**0.444**	0.359	**0.819**	**0.545**	**0.846**	0.155
	CMC NO_3^-	0.030	0.009	0.041	0.061	0.523	0.193		CMC NO_3^-	**0.287**	-0.052	0.751	**0.454**	**0.778**	0.269
	CMC NH_4^+	**0.310**	0.267	-0.442	**0.460**	-0.404	0.353		CMC NH_4^+	**0.433**	**0.528**	**0.828**	**0.700**	0.173	0.526
	CMC Sea salt	-0.028	-0.057	0.611	-0.191	0.261	0.279		CMC Sea salt	-0.041	-0.309	0.112	**-0.408**	0.583	0.069
	CMC Dust	0.189	0.207	0.331	-0.022	0.031	0.425		CMC Dust	0.137	0.234	0.515	-0.173	0.482	0.276
	CMC BC	0.167	0.010	-0.217	**0.461**	0.455	**0.709**		CMC BC	**0.579**	0.435	0.661	**0.633**	**0.785**	**0.689**
	PMF Factor 1	**0.380**	0.301	-0.349	**0.436**	0.395	0.276		PMF Factor 1	0.213	0.098	0.192	**0.529**	0.175	0.202
	PMF Factor 2	**0.345**	0.319	0.224	0.060	0.583	0.471		PMF Factor 2	**0.260**	0.422	-0.145	-0.201	0.388	0.345
	PMF Factor 3	0.181	0.108	-0.260	-0.008	-0.168	0.375		PMF Factor 3	**0.291**	**0.422**	**0.783**	0.057	0.736	0.340
	PMF Factor 4	-0.008	0.200	-0.032	0.114	0.330	0.465		PMF Factor 4	**0.311**	0.290	**0.766**	0.172	0.337	0.239
	PMF Factor 5	-0.026	-0.082	-0.581	-0.211	0.320	0.267		PMF Factor 5	-0.073	-0.152	-0.017	-0.118	**0.641**	0.007

Remarks: Values in bold are different from 0 with a significance level alpha=0.05

1   Table S5. Relative contribution from five identified sources for each HAZE episode.

Source contribution, µg m^{-3} (%)	HAZE 2011 n = 3	HAZE 2012 n = 8
Factor 1: Combustion of engine oil	5.66 (10%)	3.71 (6%)
Factor 2: Mineral dust	1.17 (2%)	15.1 (25%)
Factor 3: Mixed SIA and biomass burning	47.0 (81%)	33.2 (56%)
Factor 4: Mixed traffic and industrial	1.12 (2%)	2.12 (4%)
Factor 5: Sea salt	2.87 (5%)	5.28 (9%)

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Table S6. Pearson correlation matrix results between chemical mass closure (CMC) components and positive matrix factorisation (PMF) factor for: a) HAZE 2011, and b) HAZE 2012 episodes. Remarks: SIA = secondary inorganic aerosol.

a) HAZE 2011

Variables	PMF Factor 3	CMC SO_4^{2-}	CMC NH_4^+	CMC K^+	CMC nss-SO_4^{2-}	CMC SIA
PMF Factor 3	1	0.899	0.873	0.915	0.899	0.888
CMC SO_4^{2-}		1	**0.998**	0.647	**1.000**	**1.000**
CMC NH_4^+			1	0.603	**0.998**	**1.000**
CMC K^+				1	0.647	0.627
CMC nss- SO_4^{2-}					1	**1.000**
CMC SIA						1

b) HAZE 2012

Variables	PMF Factor 3	CMC SO_4^{2-}	CMC NH_4^+	CMC K^+	CMC nss-SO_4^{2-}	CMC SIA
PMF Factor 3	1	**0.963**	**0.944**	0.695	**0.965**	**0.952**
CMC SO_4^{2-}		1	**0.989**	**0.781**	**1.000**	**0.995**
CMC NH_4^+			1	**0.849**	**0.988**	**0.997**
CMC K^+				1	**0.777**	**0.829**
CMC nss- SO_4^{2-}					1	**0.995**
CMC SIA						1

Values in bold are different from 0 with a significance level alpha=0.05.

1 Table S7. HAZE episodes results for Pearson correlation matrices between PM2.5 mass,

2 chemical mass closure (CMC) components and positive matrix factorisation (PMF) factors

3 identified towards: a) meteorological parameters, and b) gaseous parameters. Remarks: BC =

4 black carbon while PMF factor details are as follows: factor 1= combustion of engine oil;

5 factor 2= mineral dust; factor 3= mixed secondary inorganic aerosol (SIA) and biomass

6 burning; factor 4= mixed traffic and industrial; and factor 5= sea salt. For meteorological

7 parameters, API is air pollution index; T = temperature; RH = relative humidity; WS = wind

8 speed; and WD = wind direction.

9

a)

	Variables	HAZE 2011	HAZE 2012
API	$PM_{2.5}$ mass	0.082	0.613
	SO_4^{2-}	0.491	**0.736**
	NO_3^-		-0.313
	NH_4^+	0.539	0.670
	Sea salt	0.386	-0.204
	Dust	0.145	-0.298
	BC	0.027	0.245
	Factor 1	0.990	0.093
	Factor 2	**1**	-0.231
	Factor 3	0.061	0.611
	Factor 4	-0.530	-0.075
	Factor 5	**1**	-0.370
T	$PM_{2.5}$ mass	0.387	-0.188
	SO_4^{2-}	0.737	-0.612
	NO_3^-		0.287
	NH_4^+	0.774	-0.517
	Sea salt	0.653	0.693
	Dust	0.445	**0.863**
	BC	0.336	0.371
	Factor 1	0.985	0.288
	Factor 2	**1**	**0.864**
	Factor 3	0.367	-0.304
	Factor 4	-0.241	-0.563
	Factor 5	0.952	0.751
RH	$PM_{2.5}$ mass	-0.715	0.317
	SO_4^{2-}	-0.942	**0.733**
	NO_3^-		-0.540
	NH_4^+	-0.959	0.637
	Sea salt	-0.896	-0.490
	Dust	-0.758	**-0.779**
	BC	-0.675	-0.248
	Factor 1	-0.841	-0.214
	Factor 2	**-1**	**-0.786**
	Factor 3	-0.700	0.568
	Factor 4	-0.155	0.485
	Factor 5	-0.759	-0.567

	Variables	HAZE 2011	HAZE 2012
WS	$PM_{2.5}$ mass	-0.921	-0.432
	SO_4^{2-}	-0.673	-0.642
	NO_3^-		**0.759**
	NH_4^+	-0.630	-0.567
	Sea salt	-0.755	0.054
	Dust	-0.894	0.385
	BC	-0.941	-0.009
	Factor 1	0.176	0.170
	Factor 2	**-1**	0.413
	Factor 3	-0.929	-0.658
	Factor 4	-0.972	0.063
	Factor 5	0.309	0.234
WD	$PM_{2.5}$ mass	0.600	0.427
	SO_4^{2-}	0.880	0.316
	NO_3^-		0.405
	NH_4^+	0.905	0.334
	Sea salt	0.818	-0.110
	Dust	0.650	0.095
	BC	0.555	0.347
	Factor 1	0.913	0.713
	Factor 2	**1**	0.243
	Factor 3	0.583	0.284
	Factor 4	0.003	0.070
	Factor 5	0.849	-0.111
Rainfall	$PM_{2.5}$ mass	**-1**	-0.177
	SO_4^{2-}	**-1**	0.064
	NO_3^-		-0.244
	NH_4^+	**-1**	-0.013
	Sea salt	**-1**	-0.211
	Dust	**-1**	-0.389
	BC	**-1**	-0.512
	Factor 1	**1**	
	Factor 2		-0.440
	Factor 3	**-1**	0.022
	Factor 4	**-1**	0.295
	Factor 5	**1**	-0.152

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| b)	Variables	HAZE 2011	HAZE 2012		Variables	HAZE 2011	HAZE 2012
CO	$PM_{2.5}$ mass	0.541	**0.907**	NO_X	$PM_{2.5}$ mass	0.577	0.319
	SO_4^{2-}	0.843	**0.797**		SO_4^{2-}	0.866	-0.068
	NO_3^-		-0.180		NO_3^-		0.403
	NH_4^+	0.872	**0.819**		NH_4^+	0.892	0.028
	Sea salt	0.775	-0.028		Sea salt	0.802	0.227
	Dust	0.594	-0.014		Dust	0.629	0.552
	BC	0.494	0.691		BC	0.532	0.604
	Factor 1	0.940	0.461		Factor 1	0.924	0.797
	Factor 2	**1**	0.111		Factor 2	**1**	0.677
	Factor 3	0.523	0.772		Factor 3	0.560	0.299
	Factor 4	-0.069	-0.334		Factor 4	-0.025	-0.548
	Factor 5	0.885	-0.314		Factor 5	0.864	0.126
O_3	$PM_{2.5}$ mass	-0.169	0.706	NO	$PM_{2.5}$ mass	0.492	-0.493
	SO_4^{2-}	0.259	**0.917**		SO_4^{2-}	0.811	**-0.778**
	NO_3^-		-0.391		NO_3^-		0.336
	NH_4^+	0.313	**0.919**		NH_4^+	0.842	**-0.715**
	Sea salt	0.145	-0.326		Sea salt	0.737	0.394
	Dust	-0.105	-0.505		Dust	0.546	0.625
	BC	-0.222	0.271		BC	0.443	-0.034
	Factor 1	0.924	0.451		Factor 1	0.958	0.142
	Factor 2	**1**	-0.403		Factor 2	**1**	0.622
	Factor 3	-0.189	**0.856**		Factor 3	0.473	-0.710
	Factor 4	-0.724	0.621		Factor 4	-0.126	-0.443
	Factor 5	0.967	-0.500		Factor 5	0.910	0.520
SO_2	$PM_{2.5}$ mass	0.979	0.665	NO_2	$PM_{2.5}$ mass	0.663	**0.898**
	SO_4^{2-}	0.974	0.545		SO_4^{2-}	0.915	**0.798**
	NO_3^-		-0.104		NO_3^-		0.059
	NH_4^+	0.960	0.615		NH_4^+	0.937	**0.832**
	Sea salt	0.994	0.147		Sea salt	0.862	-0.193
	Dust	0.990	0.141		Dust	0.710	-0.101
	BC	0.966	0.652		BC	0.621	0.694
	Factor 1	0.416	0.741		Factor 1	0.877	0.437
	Factor 2	**1**	0.263		Factor 2	**1**	0.037
	Factor 3	0.975	0.600		Factor 3	0.647	0.623
	Factor 4	0.664	0.004		Factor 4	0.085	-0.117
	Factor 5	0.288	0.027		Factor 5	0.803	-0.463

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2   Values in bold are different from 0 with a significance level alpha=0.05

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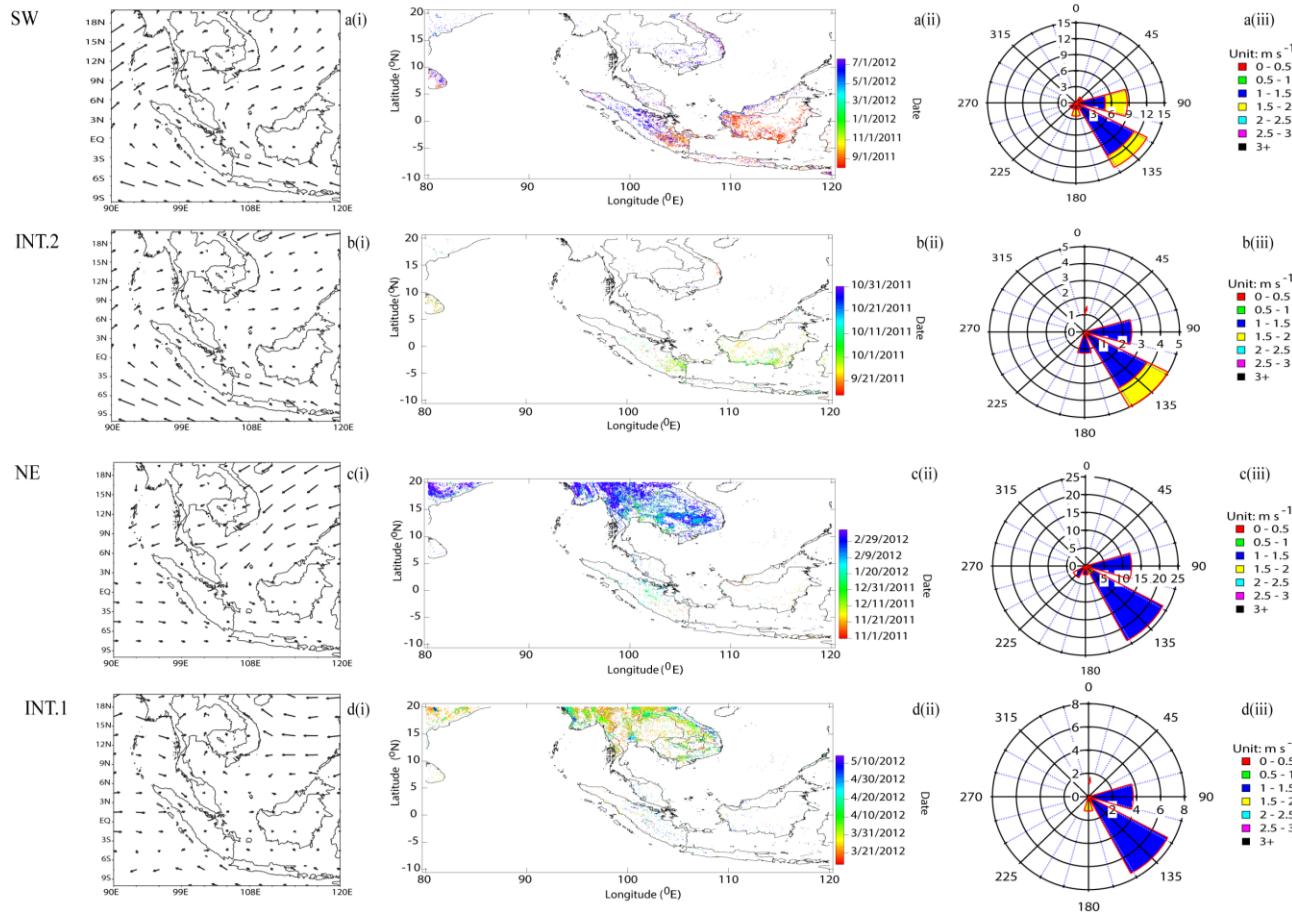
Figure S1. Seasonal plot for a) SW, b) INT.2, c) NE, and d) INT.1 pertaining to: i) regional synoptic wind field plotted for 925 hPa (500 m); ii) biomass fire hotspot; and iii) wind rose at the sampling site.
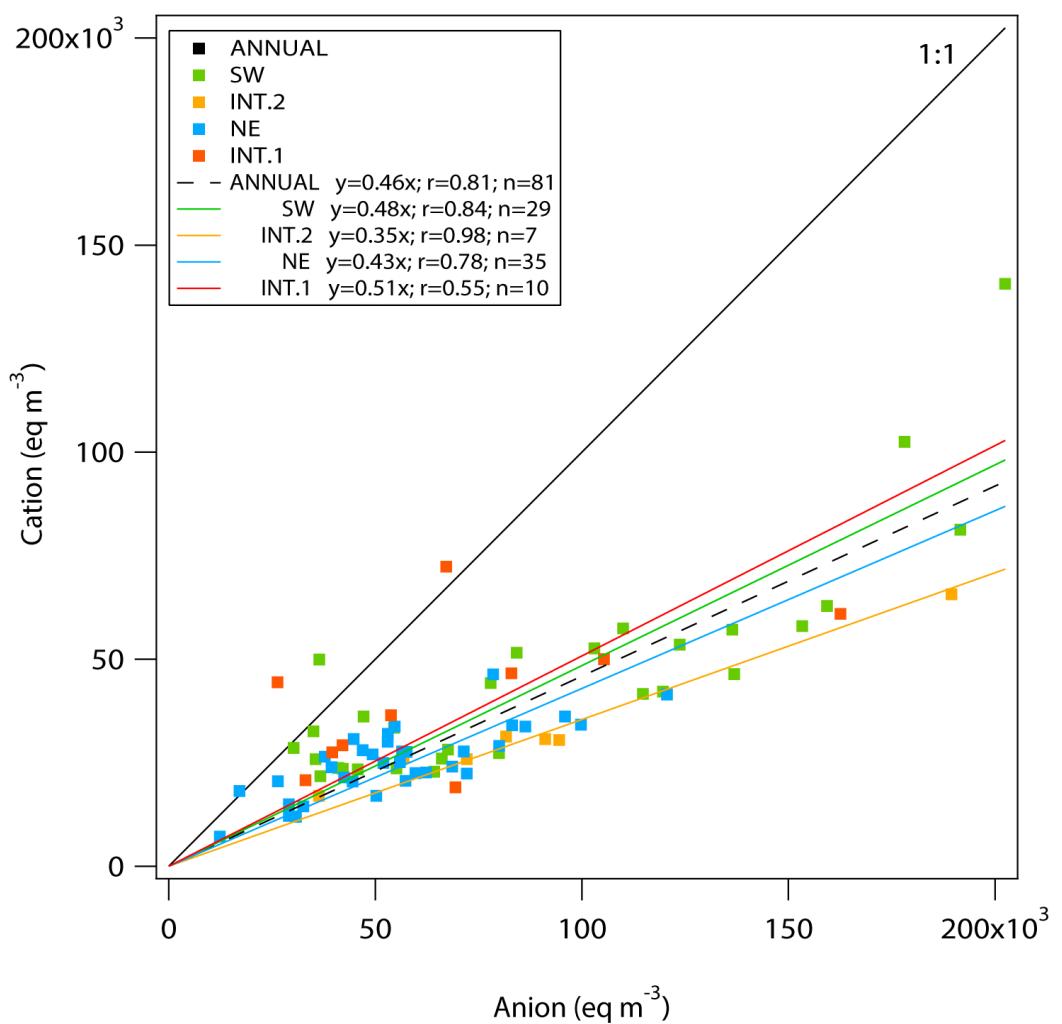
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3 Figure S2. Correlations between cations and anions of $PM_{2.5}$ on a seasonal and annual basis,

4 each with respective linear regression equations. The 1:1 line is given for comparison.
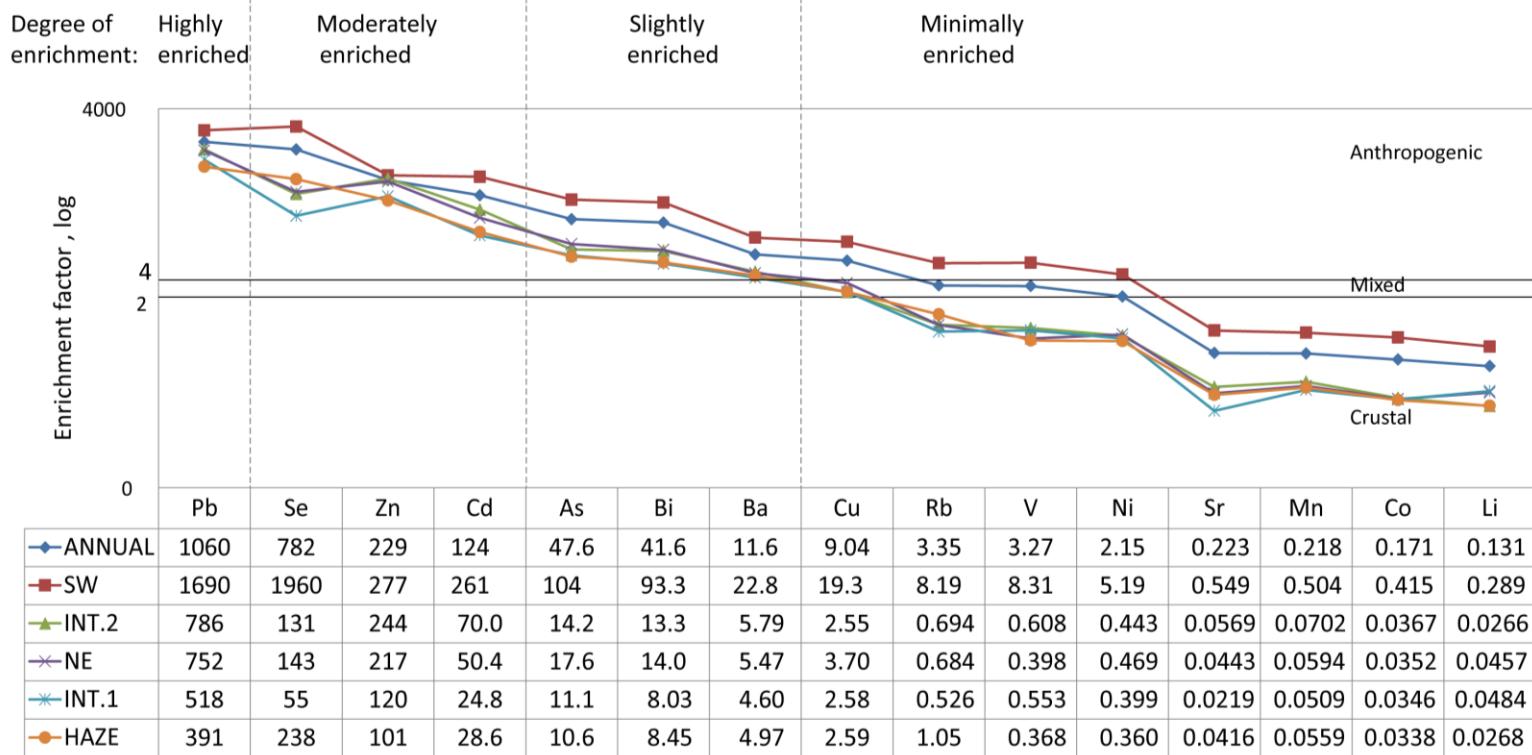
Figure S3. Seasonal variation of enrichment factor (EF) in the trace element of $PM_{2.5.}$

Remarks:

EF category = Anthropogenic: EF > 4; Mixed: 2 < EF < 4; Crustal: EF < 2.

Degree of enrichment = Highly enriched: EF ≥ 1000; Moderately enriched: 100 < EF < 1000; Slightly enriched:10 < EF < 100; and Minimally enriched: EF < 10.
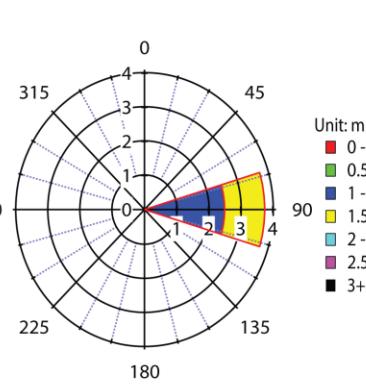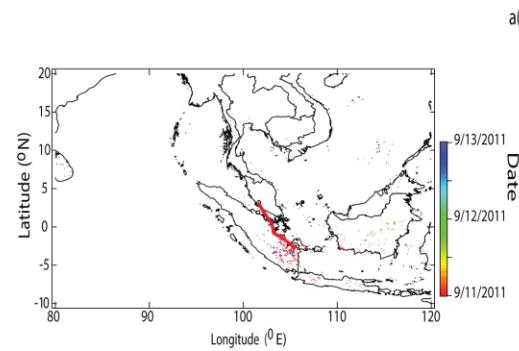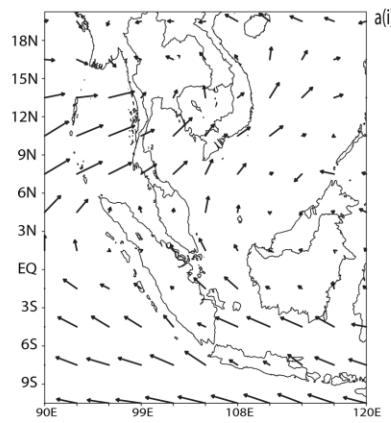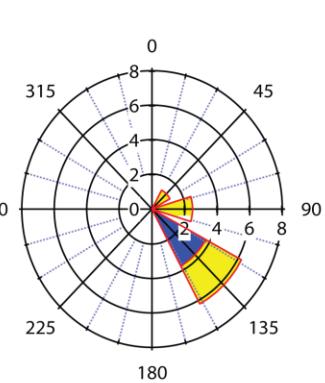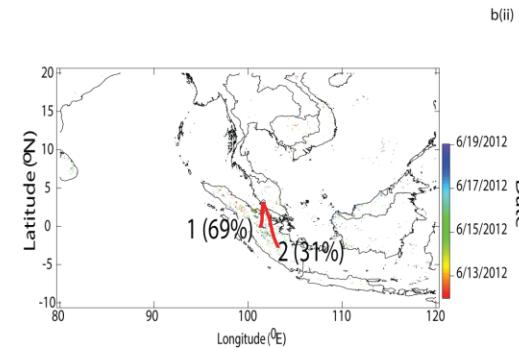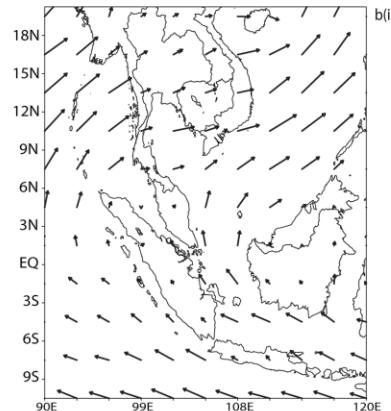
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3   Figure S4. Haze episodes plot a) 2011 HAZE, and b) 2012 HAZE of i) synoptic wind field plotted on 925 hPa (500 m); ii) biomass fire hotspot

4   with 48 h backwards trajectories at the releasing 925 hPa (500 m) with 6 h trajectory intervals; and iii) wind rose at the sampling site.
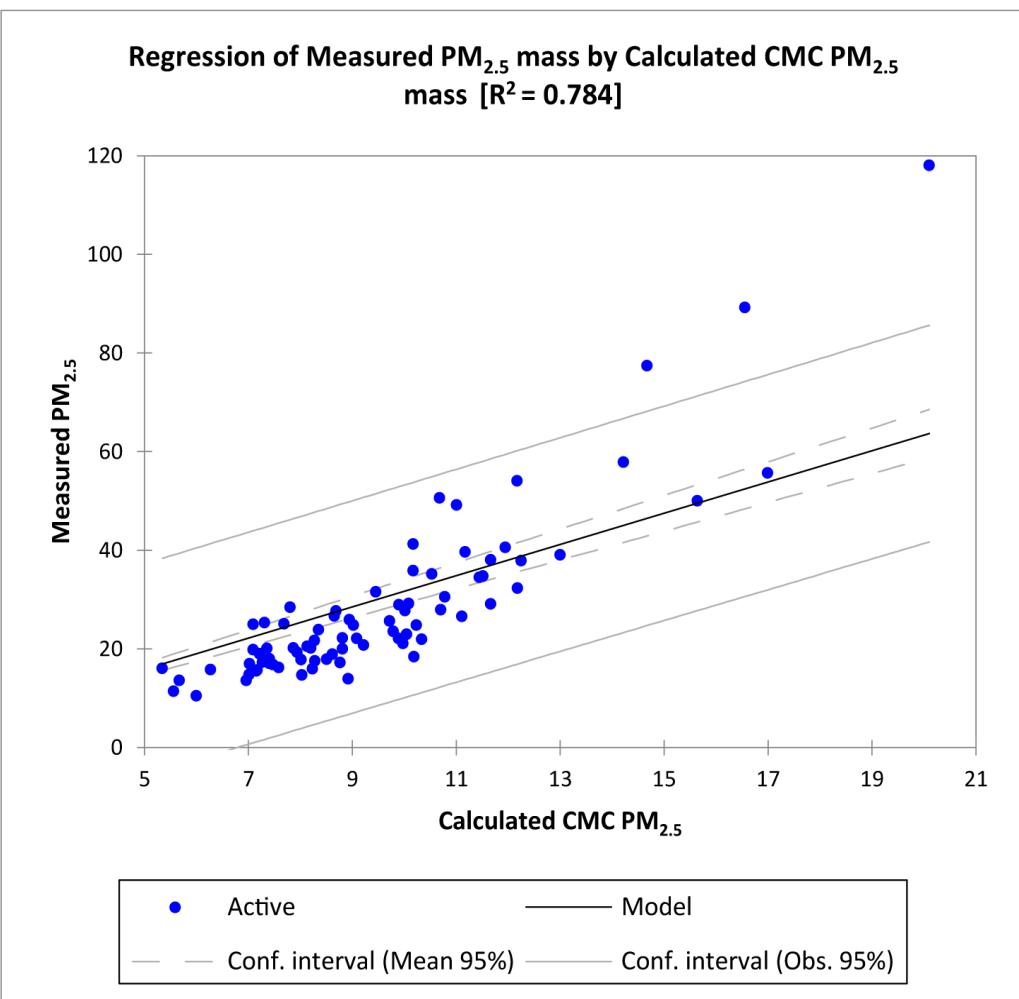
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2   Figure S5. Regression plot between calculated chemical mass closure (CMC; only identified

3   components) and measured PM$_{2.5}$ mass

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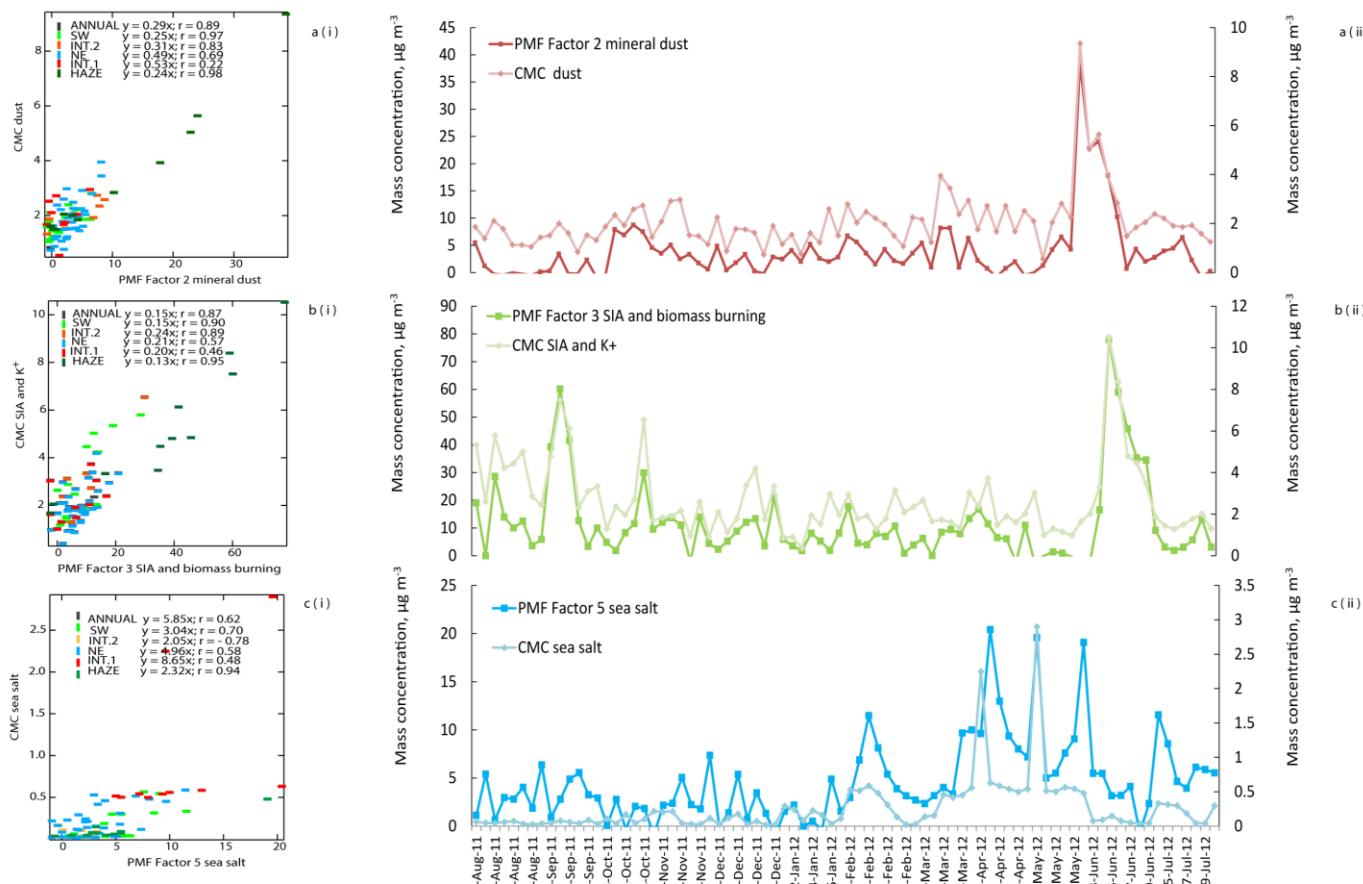
Figure S6. Comparison results between chemical mass closure (CMC) and positive matrix factorisation (PMF) results on three selected factors:

a) dust; b) secondary inorganic aerosol (SIA) and biomass burning; and c) sea salt; where i) Correlations between CMC and PMF of $PM_{2.5}$ are on

a seasonal and annual basis, each with respective linear regression equations. The 1:1 line is given for comparison; and ii) time series of the mass

concentration predicted by both CMC and PMF approach.

Reference

Squizzato, S., Masiol, M., Brunelli, A., Pistollato, S., Tarabotti, E., Rampazzo, G., and Pavoni, B.: Factors determining the formation of secondary inorganic aerosol: a case study in the Po Valley (Italy), Atmos. Chem. Phys., 13, 1927-1939, 10.5194/acp-13-1927-2013, 2013.