



Supplement of

Meteorological-gaseous influences on seasonal PM_{2.5} variability in the Klang Valley urban-industrial environment

N. Amil et al.

Correspondence to: M. T. Latif (talib@ukm.my)

The copyright of individual parts of the supplement might differ from the CC-BY 3.0 licence.

1 **Experimental quality assurance and quality control (QA/QC)**

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

6

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

13

14 Trace element analysis: Using inductively coupled plasma mass spectrometry (ICPMS), it
15 was ensured that all element regression coefficients were better than 0.999 before the sample
16 analysis was begun and that every 15th sample, the QA/QC were samples run, i.e. the UPW,
17 1 ppm multi-element standard and standard reference material (SRM1648a). The reference
18 material was 10 mg of SRM1648a Urban Particulate Matter obtained from NIST (National
19 Institute of Standards and Technology, MD, USA) while 1 ppm Multi-Element Calibration
20 Standard 3 (Perkin Elmer Pure Plus, Perkin-Elmer; USA) both prepared the same manner as
21 the samples to test the recovery and validation of the method. All lab ware used for trace
22 element analyses was Teflon material, except for the syringes.

23

1 Table S1. Descriptive statistics of experimental data (meteorological, gaseous and ion parameters); unit: mean \pm std (min - max). Remarks:
 2 ud = undetected (below detection limit); SIA = secondary inorganic aerosol; NR = neutralisation ratio = $[\text{NH}_4^+] / ([\text{SO}_4^{2-}] + [\text{NO}_3^-])$ (Squizzato et
 3 al., 2013); SO_2 gas was converted from ppm to $\mu\text{g m}^{-3}$ assuming 1 ppm = 2619 $\mu\text{g m}^{-3}$ (25 °C, 1 atm).

4

Elements	Unit	ANNUAL	SW	INT.2
		5 Aug 2011 - 18 July 2012 n = 81	15 May - 14 Sept n = 29	15 Sept - 30 Oct n = 7
API	-	50 \pm 16 (29 - 127)	60 \pm 21 (36 - 127)	49 \pm 6 (40 - 59)
T	°C	28.49 \pm 1.19 (26.05 - 31.55)	28.88 \pm 1.36 (26.4 - 31.55)	28.53 \pm 1.2 (27.1 - 30.4)
RH	%	71.23 \pm 7.91 (50.42 - 86.67)	68.23 \pm 9.22 (50.42 - 86.67)	72.88 \pm 8.5 (59.67 - 82.67)
WS	ms^{-1}	1.29 \pm 0.19 (0.87 - 1.77)	1.39 \pm 0.19 (0.97 - 1.77)	1.25 \pm 0.2 (1.01 - 1.53)
WD	Degree	128.83 \pm 31.64 (23.13 - 208.21)	123.32 \pm 37.99 (23.13 - 205)	128.52 \pm 22.02 (100 - 167.42)
Rainfall	mm	10.37 \pm 17.51 (0 - 85.4)	6.27 \pm 10.63 (0 - 34.2)	8.46 \pm 16.88 (0 - 45.4)
CO	ppm	1.29 \pm 0.31 (0.61 - 2.16)	1.26 \pm 0.32 (0.61 - 1.99)	1.43 \pm 0.32 (1.1 - 1.93)
O ₃	ppm	0.01 \pm 0.01 (ud - 0.03)	0.01 \pm 0.01 (ud - 0.02)	0.02 \pm 0.01 (0.01 - 0.03)
SO ₂	ppm	0 \pm 0 (ud - 0.01)	0 \pm 0 (ud - 0.01)	0 \pm 0 (ud - 0.01)
NO _x	ppm	0.06 \pm 0.01 (0.03 - 0.11)	0.06 \pm 0.01 (0.03 - 0.08)	0.07 \pm 0.01 (0.06 - 0.09)
NO	ppm	0.03 \pm 0.01 (0.01 - 0.07)	0.03 \pm 0.01 (0.01 - 0.04)	0.03 \pm 0.01 (0.02 - 0.05)
NO ₂	ppm	0.03 \pm 0.01 (0.02 - 0.05)	0.03 \pm 0.01 (0.02 - 0.05)	0.04 \pm 0.01 (0.03 - 0.05)
SO ₄ ²⁻	$\mu\text{g m}^{-3}$	1.33 \pm 0.88	1.77 \pm 1.16	1.62 \pm 0.78
ss-SO ₄ ²⁻	$\mu\text{g m}^{-3}$	0.08 \pm 0.09	0.06 \pm 0.02	0.02 \pm 0.01
nss-SO ₄ ²⁻	$\mu\text{g m}^{-3}$	1.29 \pm 0.9	1.75 \pm 1.18	1.61 \pm 0.79
NO ₃ ⁻	$\mu\text{g m}^{-3}$	0.21 \pm 0.13	0.19 \pm 0.08	0.29 \pm 0.22
NH ₄ ⁺	$\mu\text{g m}^{-3}$	0.99 \pm 0.85	1.45 \pm 1.18	1 \pm 0.64
SIA	$\mu\text{g m}^{-3}$	2.4 \pm 1.68	3.28 \pm 2.25	2.78 \pm 1.58
SIA/PM _{2.5}	%	8.54 \pm 2.97	8.69 \pm 3.43	9.62 \pm 2.96
NR	-	0.65 \pm 0.31	0.72 \pm 0.19	0.55 \pm 0.07
SO ₄ ²⁻ -SO ₂	$\mu\text{g m}^{-3}$	1.33 - 8.19	1.77 - 9.5	1.62 - 10.84

5

6

1 Continuation of Table S1 (2)

2

Elements	Unit	NE	INT.1	HAZE
		1 Nov - 14 Mar n = 35	15 Mar - 14 May n = 10	n = 11
API	-	44 ± 8 (29 - 58)	45 ± 9 (33 - 58)	78 ± 22 (49 - 127)
T	°C	28.07 ± 1.02 (26.05 - 30.37)	28.76 ± 0.78 (27.47 - 30.18)	29.5 ± 1.33 (26.66 - 31.55)
RH	%	73.58 ± 6.79 (56.54 - 85.54)	70.51 ± 4.01 (65.08 - 76.96)	63 ± 9.91 (50.42 - 81.63)
WS	ms ⁻¹	1.2 ± 0.17 (0.87 - 1.46)	1.32 ± 0.18 (1.08 - 1.71)	1.49 ± 0.14 (1.27 - 1.7)
WD	Degree	132.47 ± 31.19 (83.17 - 208.21)	128.43 ± 25.06 (103.33 - 177.83)	102.84 ± 33.19 (23.13 - 136.88)
Rainfall	mm	15.13 ± 22.69 (0 - 85.4)	7.04 ± 9.69 (0 - 24)	2.28 ± 5.18 (0 - 15.8)
CO	ppm	1.29 ± 0.3 (0.92 - 2.16)	1.31 ± 0.28 (0.84 - 1.75)	1.45 ± 0.31 (0.89 - 1.99)
O ₃	ppm	0.01 ± 0.01 (ud - 0.03)	0.01 ± 0 (ud - 0.02)	0.02 ± 0 (0.01 - 0.02)
SO ₂	ppm	0 ± 0 (ud - 0)	0 ± 0 (ud - 0)	0 ± 0 (ud - 0.01)
NO _x	ppm	0.07 ± 0.01 (0.04 - 0.11)	0.06 ± 0.01 (0.04 - 0.07)	0.06 ± 0.01 (0.03 - 0.07)
NO	ppm	0.03 ± 0.01 (0.02 - 0.07)	0.03 ± 0.01 (0.01 - 0.04)	0.02 ± 0.01 (0.01 - 0.04)
NO ₂	ppm	0.03 ± 0.01 (0.02 - 0.05)	0.03 ± 0.01 (0.02 - 0.04)	0.03 ± 0.01 (0.02 - 0.05)
SO ₄ ²⁻	µg m ⁻³	0.98 ± 0.41	1.06 ± 0.7	2.4 ± 1.24
ss-SO ₄ ²⁻	µg m ⁻³	0.05 ± 0.03	0.16 ± 0.15	0.06 ± 0
nss-SO ₄ ²⁻	µg m ⁻³	0.95 ± 0.42	0.9 ± 0.6	2.4 ± 1.24
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	2.21 ± 1.47
SIA	µg m ⁻³	1.72 ± 0.81	1.98 ± 0.87	4.67 ± 2.59
SIA/PM _{2.5}	%	8.23 ± 2.77	8.44 ± 2.31	7.5 ± 2.66
NR	-	0.56 ± 0.11	0.83 ± 0.76	0.85 ± 0.22
SO ₄ ²⁻ -SO ₂	µg m ⁻³	0.98 - 6.64	1.06 - 7.96	2.4 - 9.09

3

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
 5

	Elements	Unit	ANNUAL					SW				
			n = 81					n = 29				
			min	max	med	avg	std	min	max	med	avg	std
1	PM _{2.5}	µg/m ³	6	118	23	28	17	14	118	29	38	24
2	F ⁻	µg/m ³	< MDL	0.018	0.002	0.003	0.004	< MDL	0.018	0.003	0.006	0.005
3	Cl ⁻	µg/m ³	< MDL	0.141	0.016	0.026	0.026	0.007	0.085	0.018	0.024	0.017
4	NO ₂ ⁻	µg/m ³	< MDL	0.009	0.004	0.004	0.002	< MDL	0.005	0.003	0.003	0.001
5	Br ⁻	µg/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 ⁻	µg/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⁻	µg/m ³	< MDL	0.205	0.072	0.079	0.063	< MDL	0.074	0.048	0.054	0.017
8	SO ₄ ²⁻	µg/m ³	0.139	4.155	1.093	1.328	0.885	0.368	4.155	1.385	1.769	1.158
9	Na ⁺	µg/m ³	< MDL	2.113	0.218	0.300	0.357	0.113	0.385	0.201	0.236	0.093
10	NH ₄ ⁺	µg/m ³	0.085	5.440	0.691	0.987	0.852	0.193	5.440	1.054	1.453	1.180
11	K ⁺	µg/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⁺	µg/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⁺	µg/m ³	< MDL	0.391	0.024	0.033	0.044	0.015	0.115	0.029	0.035	0.021
14	Al	µg/m ³	< MDL	7.740	0.712	1.063	1.226	< MDL	7.740	0.974	1.723	1.973
15	Ba	µg/m ³	0.011	0.115	0.030	0.036	0.019	0.014	0.115	0.027	0.040	0.025
16	Fe	µg/m ³	< MDL	5.245	0.879	1.000	0.706	< MDL	5.245	0.940	1.236	1.108
17	Pb	µg/m ³	0.015	0.787	0.077	0.108	0.116	0.015	0.146	0.060	0.069	0.033
18	Zn	µg/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	µg/m ³	2.548	5.548	4.248	4.155	0.642	2.974	5.167	4.248	4.155	0.581

1
2
3
4
5

Continuation of Table S2 (2)

	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

6

1
2
3
4
5

Continuation of Table S2 (3)

	Elements	Unit	INT.1					HAZE					% Recovery
			n = 10					n = 11					
			min	max	med	avg	std	min	max	med	avg	std	
1	PM _{2.5}	µg/m ³	14	39	22	23	8	40	118	54	61	24	
2	F ⁻	µg/m ³	< MDL	0.011	0.004	0.004	0.004	0.003	0.018	0.011	0.011	0.006	105
3	Cl ⁻	µg/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 ⁻	µg/m ³	< MDL	0.003	0.003	0.003	0.000	0.003	0.004	0.004	0.004	0.001	107
5	Br ⁻	µg/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 ⁻	µg/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⁻	µg/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⁻	µg/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 ⁺	µg/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 ⁺	µg/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 ⁺	µg/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⁺	µg/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⁺	µg/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	µg/m ³	0.025	0.990	0.387	0.408	0.292	< MDL	7.740	2.050	2.788	2.564	36
15	Ba	µg/m ³	0.018	0.052	0.032	0.033	0.011	0.022	0.115	0.043	0.046	0.026	
16	Fe	µg/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	µg/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	µg/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	µg/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. Seasonal results of Pearson correlation matrices between $PM_{2.5}$ mass, chemical mass closure (CMC) components and positive matrix
2 factorisation (PMF) factors identified towards: a) meteorological parameters, and b) gaseous parameters. Remarks: BC = black carbon while
3 PMF factors were: factor 1= combustion of engine oil; factor 2= mineral dust; factor 3= mixed secondary inorganic aerosol (SIA) and biomass
4 burning; factor 4= mixed traffic and industrial; and factor 5= sea salt. For meteorological parameters, API is air pollution index; T =
5 temperature; RH = relative humidity; WS = wind speed; and WD = wind direction.

6

1
2

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	0.48
	CMC NO ₃ ⁻	-0.172	-0.35	0.37	0.079	0.623	-0.175
	CMC NH ₄ ⁺	0.712	0.667	0.75	0.561	0.46	0.606
	CMC Sea salt	-0.087	-0.294	0.214	-0.088	0.547	-0.102
	CMC Dust	0.227	0.22	-0.033	-0.101	0.302	-0.114
	CMC BC	0.353	0.58	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	0.31	0.236	0.572	0.201	0.03	-0.05
	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	-0.11	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	-0.28	0.403	0.089	
PMF Factor 5	0.213	0.313	0.14	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	-0.01	-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	-0.64	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	-0.6	-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	-0.22	-0.353	-0.131	-0.219	0.6	-0.57	

	Variables	ANNUAL	SW	INT.2	NE	INT.1	HAZE
WS	PM _{2.5} mass	0.274	0.164	0.245	-0.03	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 ⁺	0.14	0.015	-0.103	-0.034	-0.421	-0.53
	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	0.43
	CMC BC	-0.07	0.162	0.445	-0.403	-0.034	-0.06
	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	0.17	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	-0.19	0.309	0.145	-0.039
	CMC Dust	0.014	-0.085	0.625	0.406	-0.699	0.197
	CMC BC	-0.063	-0.13	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 ⁻	-0.03	-0.03	-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	0.53	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	-0.11	-0.102	-0.743	-0.296	-0.524	-0.343	
PMF Factor 5	-0.05	-0.128	-0.816	0.144	0.338	-0.101	

3

1 Continuation of Table S3 (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	0.8	0.38	0.588	0.192
	CMC SO ₄ ²⁻	0.455	0.579	0.793	0.428	0.582	0.412		CMC SO ₄ ²⁻	0.05	0.038	0.709	0.18	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 ⁺	0.05	0.127	0.663	0.351	0.226	0.03
	CMC Sea salt	0.02	-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	0.06	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	0.17	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	-0.28	0.081	0.65
	PMF Factor 3	0.302	0.612	0.42	-0.083	0.072	0.6		PMF Factor 3	-0.139	0.014	0.504	-0.22	0	0.12
PMF Factor 4	0.199	0.157	0.577	0.04	-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	0.01	PMF Factor 5	-0.122	0.029	0.18	-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	-0.27	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	-0.21	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	0.04	0.358		PMF Factor 1	0.083	0.247	0.651	0.326	-0.246	0.05
	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	-0.36	0.698
	PMF Factor 3	0.304	0.597	0.204	0.383	0.703	0.538		PMF Factor 3	-0.39	-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	-0.37	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	-0.25	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	0.67	-0.017		CMC SO ₄ ²⁻	0.444	0.359	0.819	0.545	0.846	0.155
	CMC NO ₃ ⁻	0.03	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 ⁺	0.31	0.267	-0.442	0.46	-0.404	0.353		CMC NH ₄ ⁺	0.433	0.528	0.828	0.7	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	0.01	-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	0.38	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	0.06	0.583	0.471		PMF Factor 2	0.26	0.422	-0.145	-0.201	0.388	0.345
	PMF Factor 3	0.181	0.108	-0.26	-0.008	-0.168	0.375		PMF Factor 3	0.291	0.422	0.783	0.057	0.736	0.34
PMF Factor 4	-0.008	0.2	-0.032	0.114	0.33	0.465	PMF Factor 4	0.311	0.29	0.766	0.172	0.337	0.239		
PMF Factor 5	-0.026	-0.082	-0.581	-0.211	0.32	0.267	PMF Factor 5	-0.073	-0.152	-0.017	-0.118	0.641	0.007		

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

1 Table S4. Relative contribution from five identified sources for each HAZE episode.

Source contribution, $\mu\text{g m}^{-3}$ (%)	HAZE 2011	HAZE 2012
	n = 3	n = 8
Factor 1: Combustion of engine oil	5.66 (10%)	3.71 (6%)
Factor 2: Mineral dust	1.17 (2%)	15.07 (25%)
Factor 3: Mixed SIA and biomass burning	46.97 (81%)	33.15 (56%)
Factor 4: Mixed traffic and industrial	1.12 (2%)	2.12 (4%)
Factor 5: Sea salt	2.87 (5%)	5.28 (9%)

2

1 Table S5. Pearson correlation matrix results between chemical mass closure (CMC)
 2 components and positive matrix factorisation (PMF) factor for: a) HAZE 2011, and b) HAZE
 3 2012 episodes. Remarks: SIA = secondary inorganic aerosol.

4 a) HAZE 2011

Variables	PMF Factor 3	CMC				
		CMC SO ₄ ²⁻	CMC NH ₄ ⁺	CMC K ⁺	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

5 b) HAZE 2012

Variables	PMF Factor 3	CMC				
		CMC SO ₄ ²⁻	CMC NH ₄ ⁺	CMC K ⁺	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

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

1 Table S6. 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

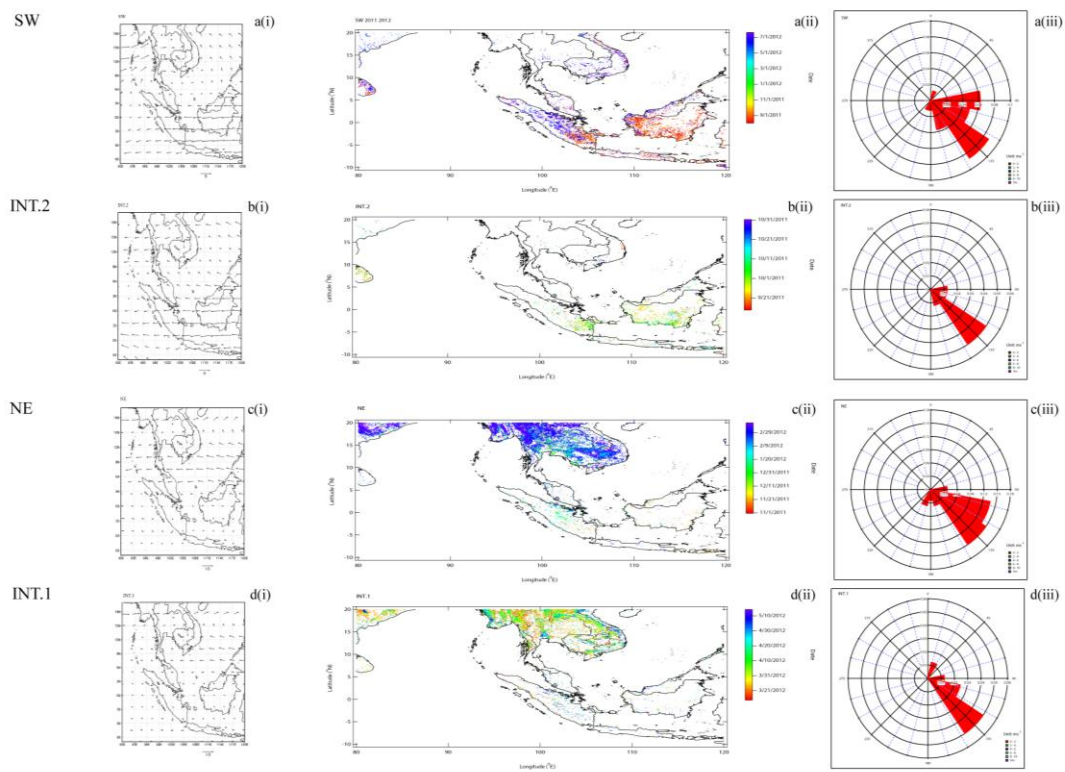
a)	Variables	HAZE 2011	HAZE 2012		Variables	HAZE 2011	HAZE 2012
API	PM _{2.5} mass	0.082	0.613	WS	PM _{2.5} mass	-0.921	-0.432
	SO ₄ ²⁻	0.491	0.736		SO ₄ ²⁻	-0.673	-0.642
	NO ₃ ⁻		-0.313		NO ₃ ⁻		0.759
	NH ₄ ⁺	0.539	0.67		NH ₄ ⁺	-0.63	-0.567
	Sea salt	0.386	-0.204		Sea salt	-0.755	0.054
	Dust	0.145	-0.298		Dust	-0.894	0.385
	BC	0.027	0.245		BC	-0.941	-0.009
	Factor 1	0.99	0.093		Factor 1	0.176	0.17
	Factor 2	1	-0.231		Factor 2	-1	0.413
	Factor 3	0.061	0.611		Factor 3	-0.929	-0.658
	Factor 4	-0.53	-0.075		Factor 4	-0.972	0.063
Factor 5	1	-0.37	Factor 5	0.309	0.234		
T	PM _{2.5} mass	0.387	-0.188	WD	PM _{2.5} mass	0.6	0.427
	SO ₄ ²⁻	0.737	-0.612		SO ₄ ²⁻	0.88	0.316
	NO ₃ ⁻		0.287		NO ₃ ⁻		0.405
	NH ₄ ⁺	0.774	-0.517		NH ₄ ⁺	0.905	0.334
	Sea salt	0.653	0.693		Sea salt	0.818	-0.11
	Dust	0.445	0.863		Dust	0.65	0.095
	BC	0.336	0.371		BC	0.555	0.347
	Factor 1	0.985	0.288		Factor 1	0.913	0.713
	Factor 2	1	0.864		Factor 2	1	0.243
	Factor 3	0.367	-0.304		Factor 3	0.583	0.284
	Factor 4	-0.241	-0.563		Factor 4	0.003	0.07
Factor 5	0.952	0.751	Factor 5	0.849	-0.111		
RH	PM _{2.5} mass	-0.715	0.317	Rainfall	PM _{2.5} mass	-1	-0.177
	SO ₄ ²⁻	-0.942	0.733		SO ₄ ²⁻	-1	0.064
	NO ₃ ⁻		-0.54		NO ₃ ⁻		-0.244
	NH ₄ ⁺	-0.959	0.637		NH ₄ ⁺	-1	-0.013
	Sea salt	-0.896	-0.49		Sea salt	-1	-0.211
	Dust	-0.758	-0.779		Dust	-1	-0.389
	BC	-0.675	-0.248		BC	-1	-0.512
	Factor 1	-0.841	-0.214		Factor 1	1	
	Factor 2	-1	-0.786		Factor 2		-0.44
	Factor 3	-0.7	0.568		Factor 3	-1	0.022
	Factor 4	-0.155	0.485		Factor 4	-1	0.295
Factor 5	-0.759	-0.567	Factor 5	1	-0.152		

b)	Variables	HAZE			Variables	HAZE	
		2011	2012			2011	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 ⁻		-0.18		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	0.94	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	0.56	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	-0.71
Factor 4	-0.724	0.621	Factor 4	-0.126	-0.443		
Factor 5	0.967	-0.5	Factor 5	0.91	0.52		
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 ⁺	0.96	0.615		NH ₄ ⁺	0.937	0.832
	Sea salt	0.994	0.147		Sea salt	0.862	-0.193
	Dust	0.99	0.141		Dust	0.71	-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	0.6		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		

1

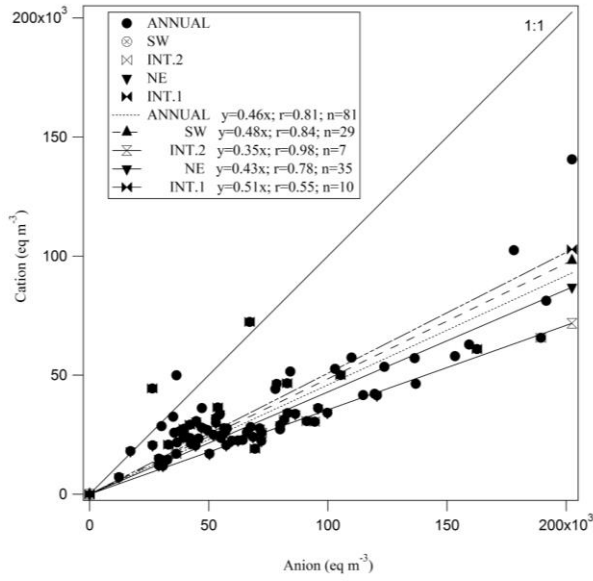
2 Values in bold are different from 0 with a significance level alpha=0.05

3



1
2
3
4
5
6

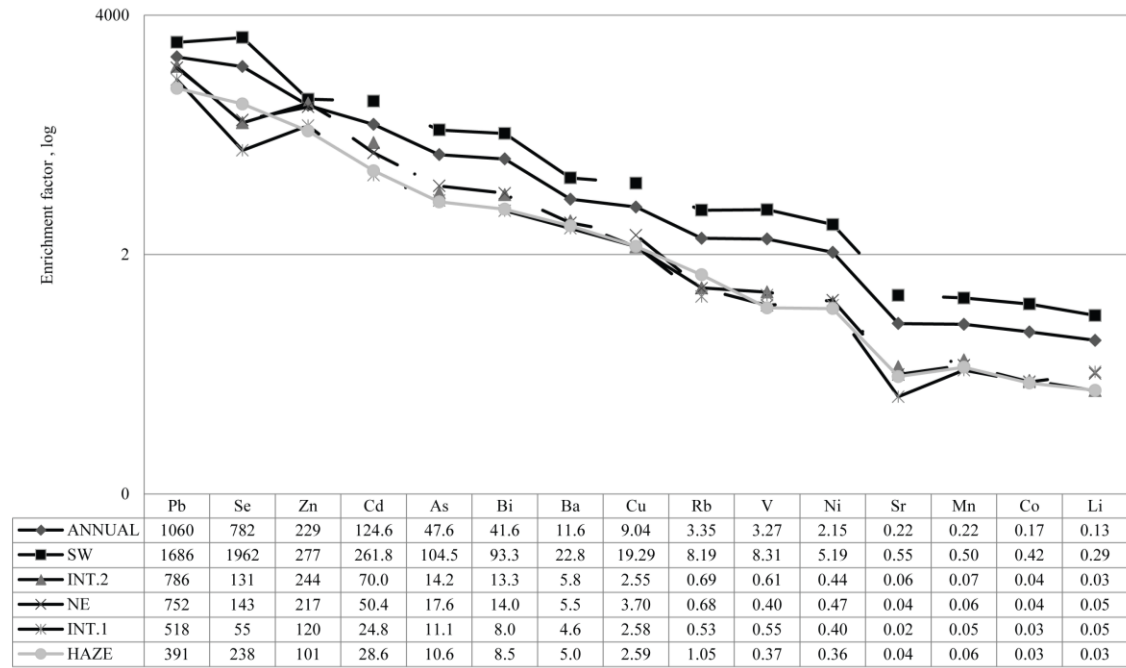
Figure S1. Seasonal plot for a) SW, b) INT.2, c) NE, and d) INT.1 pertaining to: i) regional synoptic wind field plotted for 925 hPa (500 m); ii) biomass fire hotspot; and iii) wind rose at the sampling site.



1

2 Figure S2. Correlations between cations and anions of PM_{2.5} on a seasonal and annual basis,
 3 each with respective linear regression equations. The 1:1 line is given for comparison.

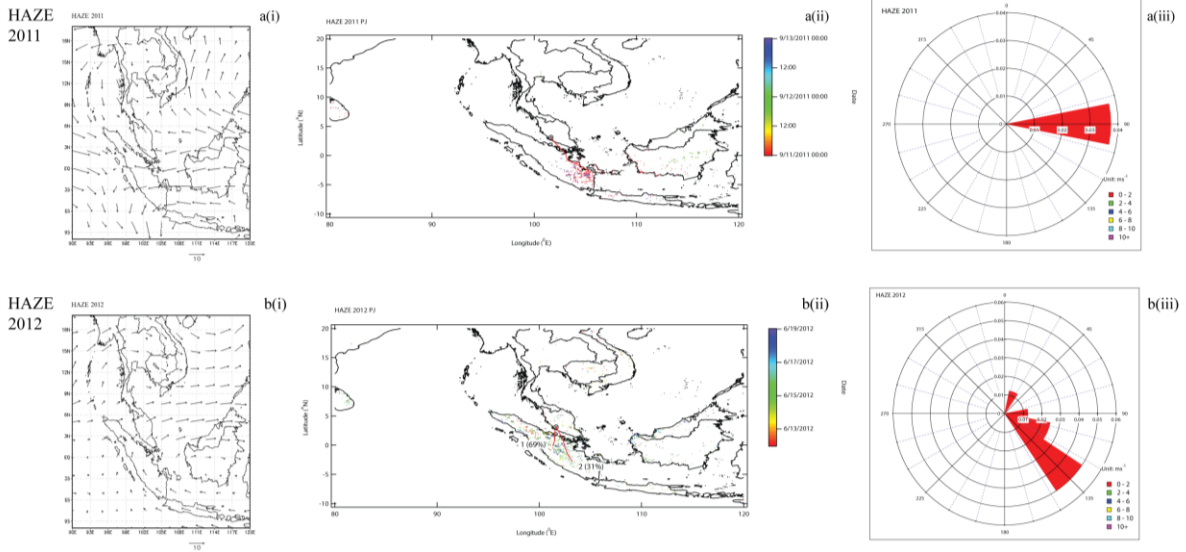
4



1

2 Figure S3. Seasonal variation of enrichment factor (EF) in the trace element of PM_{2.5}.

3



1
2
3
4
5
6
7

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 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.

1 **Reference**

2

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

6