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Chemical characterization and source identification of $PM_{2.5}$ at multiple sites in the Beijing–Tianjin–Hebei region, China

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Supplementary Material

Table S1 The distribution of the sample quantity during the entire study period at each site (sample quantity (proportion))

	Beijing			Tianjin			Shijiazhuang			Xinglong		
	C[a]	MP[b]	HP[c]	C	MP	HP	C	MP	HP	C	MP	HP
Total	100	70	54	80	93	41	41	87	93	149	41	21
Summer	33 (33%)	15(22%)	11(20%)	25(31%)	28(30%)	2(5%)	22(54%)	17(20%)	15(16%)	36(24%)	8(20%)	7(33%)
Autumn	25 (25%)	17(24%)	6(11%)	21(26%)	27(29%)	3(7%)	11(27%)	29(33%)	11(12%)	33(22%)	6(15%)	7(33%)
Winter	20 (20%)	16(23%)	22(41%)	19(24%)	12(13%)	21(51%)	4(10%)	13(15%)	40(43%)	47(32%)	8(20%)	4(19%)
Spring	22 (22%)	22(31%)	15(28%)	15(19%)	26(28%)	15(37%)	4(10%)	28(32%)	27(29%)	33(22%)	19(46%)	3(14%)

[a] Clean days ($PM_{2.5}$＜75 μg/m^3); [b] Moderate pollution days (75≤$PM_{2.5}$＜150 μg/m^3); [c] Heavy pollution days ($PM_{2.5}$≥150 μg/m^3)

Table S2 The meteorological parameters under specific conditions during the sampling periods

		Temperature (℃)				Relative humidity (%)				Wind speed (m/s)			
		Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn	Winter	Spring
Beijing	Annual	28.0	18.0	1.5	14.0	51	59	32	35	1.0	0.8	1.5	1.7
	C[a]	28.3	18.6	1.5	12.1	44	46	21	32	1.2	1.2	2.3	2.3
	MP[b]	27.4	18.7	2.3	14.2	57	69	34	33	0.9	0.6	1.4	1.6
	HP[c]	27.7	17.5	1.1	16.4	67	69	41	42	0.7	0.6	0.9	1.2
	D[d]	30.3	19.8	2.8	16.0	43	51	28	27	1.4	1.0	1.7	2.1
	N[e]	25.8	16.2	0.1	11.2	60	67	37	43	0.7	0.6	1.3	1.3
Tianjin	Annual	27.7	18.4	1.1	13.0	54	60	41	41	1.5	1.3	1.4	1.8
	C	28.6	18.9	1.5	11.2	47	61	31	39	1.6	1.4	1.8	2.1
	MP	27.2	19.1	1.1	13.3	59	61	40	44	1.4	1.1	1.6	1.8
	HP	28.4	16.9	1.7	13.4	66	53	46	40	1.2	1.2	1.1	1.5
	D	29.7	20.1	2.6	15.2	47	53	35	34	1.8	1.5	1.7	2.1
	N	25.7	16.6	-0.4	10.9	62	69	45	48	1.2	1.0	1.2	1.5
Shijiazhuang	Annual	26.9	17.9	1.0	13.8	63	75	41	46	1.2	0.9	1.0	1.5
	C	28.6	16.3	1.9	9.1	50	64	21	64	1.3	1.1	1.3	1.2
	MP	26.6	18.9	1.6	13.0	66	78	35	47	1.2	0.9	1.4	1.5
	HP	26.2	17.7	0.6	15.5	71	78	46	40	1.1	0.7	0.9	1.6
	D	29.2	19.6	3.0	16.0	54	68	36	39	1.4	1.1	1.2	1.9
	N	24.8	16.2	-1.0	11.5	70	83	47	53	1.0	0.7	0.8	1.2
Xinglong	Annual	20.6	12.1	-4.8	7.0	67	66	40	44	2.3	2.5	2.7	3.3
	C	20.6	12.1	-5.0	6.5	61	58	35	30	2.1	2.6	2.9	3.4
	MP	19.7	14.0	-3.1	8.9	80	68	42	52	2.5	3.3	2.6	3.4
	HP	21.5	13.8	-4.9	8.9	85	74	79	66	2.7	2.4	1.4	3.6
	D	21.7	13.2	-3.7	8.3	64	62	36	38	2.4	2.7	2.7	3.6
	N	19.4	10.9	-5.8	5.5	70	71	43	50	2.1	2.3	2.7	3.1

[a] Clean days ($PM_{2.5}$＜75 μg/m^3); [b] Moderate pollution days (75≤$PM_{2.5}$＜150 μg/m^3); [c] Heavy pollution days ($PM_{2.5}$≥150 μg/m^3);

[d] Daytime; [e] Nighttime

Table S3 Comparisons of the $PM_{2.5}$ source apportionment results from this study and other recent studies

Reference	Wu et al. (2014)		Liu et al. (2017)		EPB[a]				This study			
City	BJ[b]		TJ[c]			BJ	TJ	SJZ[d]		BJ	TJ	SJZ
Time	2010		06-08/2015			2013-2014				2014-2015		
Method	PMF		PMF			multiple models				PMF		
Sources and contributions (%)	traffic emissions	12.0	vehicle exhaust	25.4	vehicle exhaust	19.9–22.4	13.2–15.6	10.5–11.6	motor vehicle exhaust	24.9	15.2	17.3
	coal combustion	22.0	coal combustion	16.5	coal combustion	14.3–16.1	17.8–21.1	20.0–21.9	coal combustion	5.6	12.4	15.5
	secondary sulfate/nitrate	30.2	secondary sources	26.1					secondary inorganic	40.5	29.2	36.4
	dust/soil	12.4	crustal dust	13.2	soil dust	9.2–10.3	19.8–23.4	15.8–17.3	mineral dust	8.6	11.7	8.5
	metallurgical emission	0.4							oil refining /metal smelting source		2.8	0.7
	industry	6.9			industry	11.6–13.0	11.2–13.3	17.6–19.4	industrial process	3.2	8.9	6.3
	secondary organic aerosol	9.9			regional transport	28–36	22–34	23–30				
			biomass burning	10.2	other sources [e]	9.0–10.2	4.0–4.7	6.2–6.8	biomass burning	4.5	5.3	2.8

[a] Beijing Municipal Environmental Protection Bureau (http://www.bjepb.gov.cn/), Tianjin Municipal Environmental Protection Bureau (http://www.tjhb.gov.cn/) and Shijiazhuang Municipal Environmental Protection Bureau (http://www.sjzhb.gov.cn/)

[b] Beijing; c Tianjin; d Shijiazhuang

[e] Including emissions from biomass burning, cooking, and agricultural, etc.

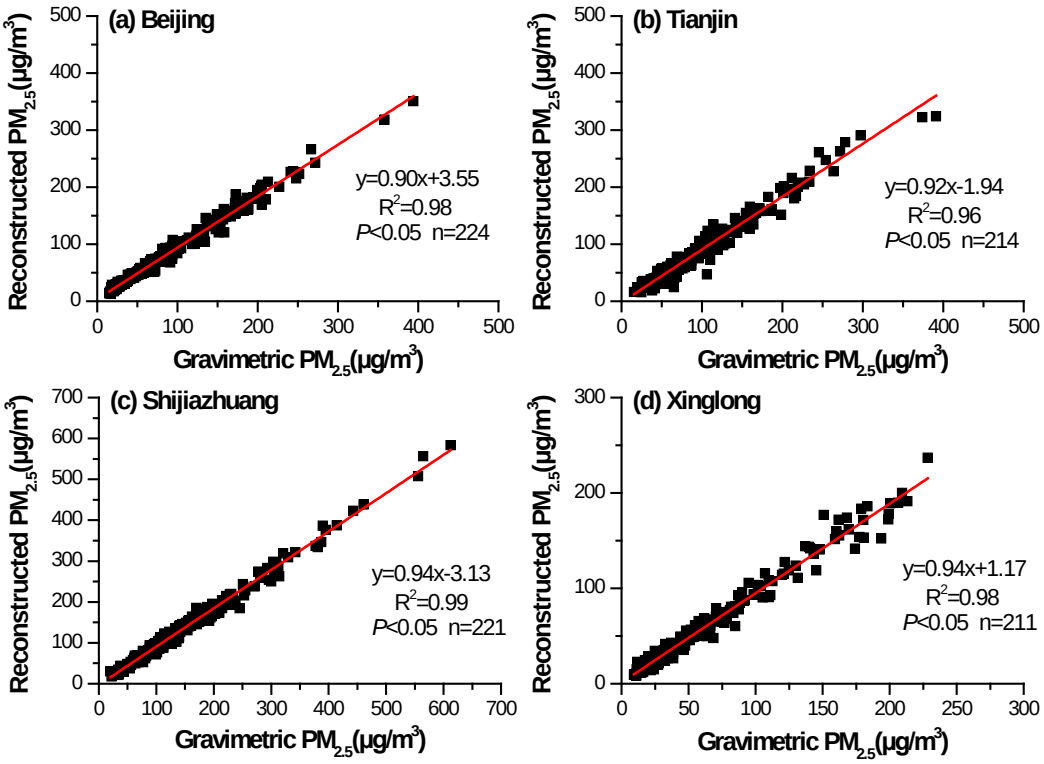

Figure S1. Gravimetric $PM_{2.5}$ versus reconstructed $PM_{2.5}$ mass concentrations from Beijing (a), Tianjin (b), Shijiazhuang (c) and Xinglong (d). "n" represents the sample quantity at each site.

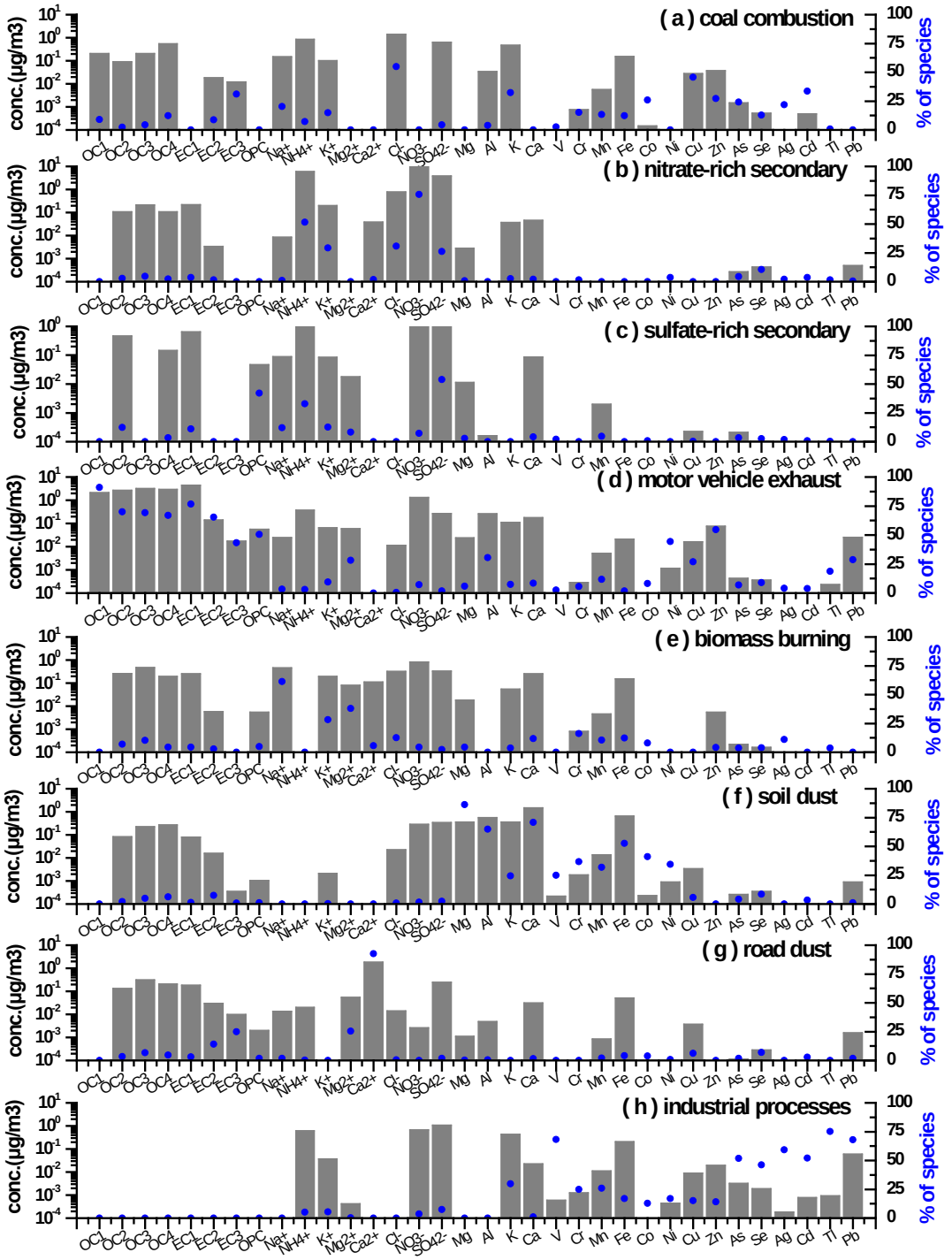

Figure S2. PMF factor/source profiles for the $PM_{2.5}$ samples throughout the entire study period in Beijing in terms of concentrations (μg/m^3) and percentages (%)

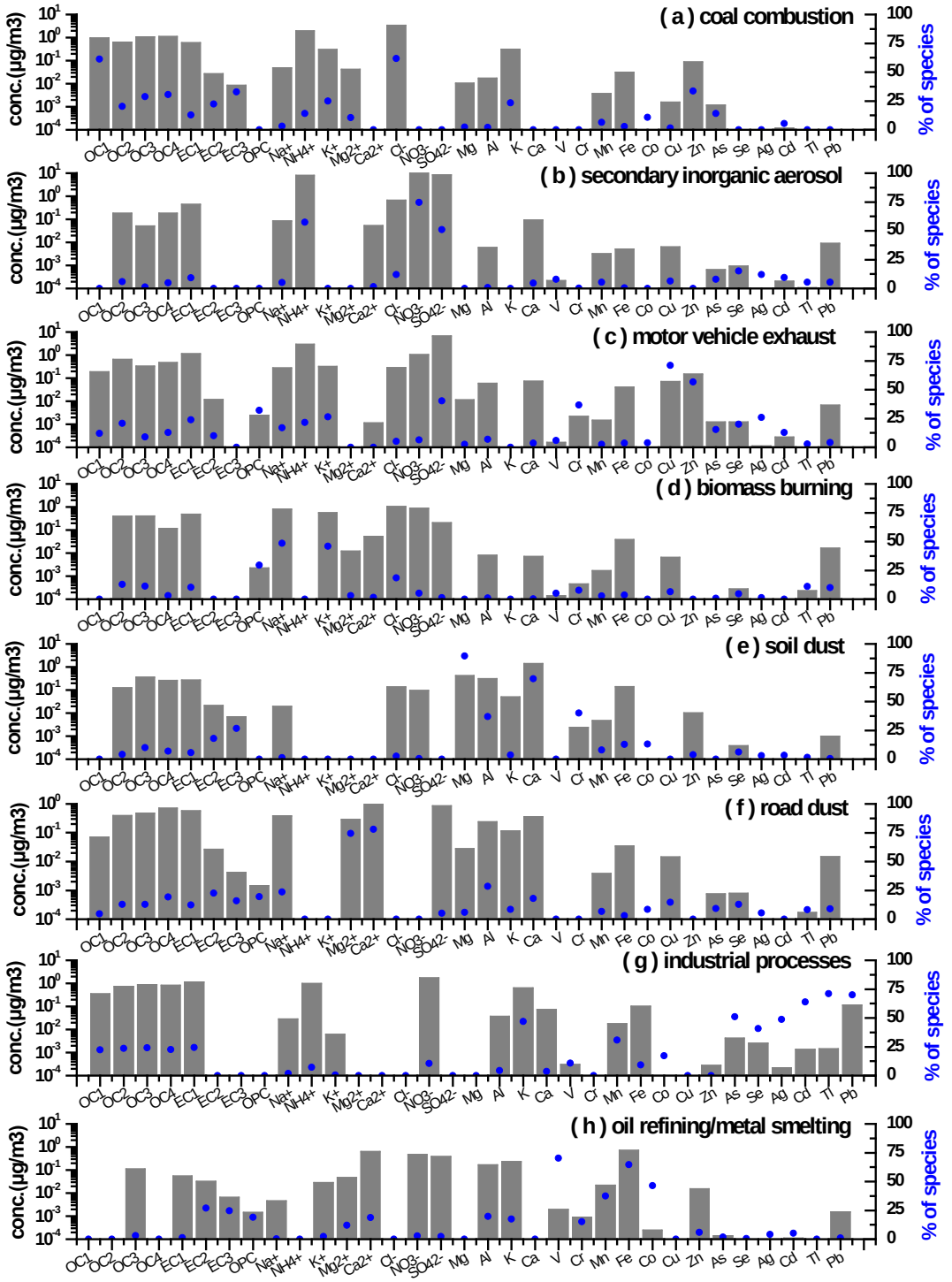

Figure S3. PMF factor/source profiles for the $PM_{2.5}$ samples throughout the entire study period in Tianjin in terms of concentrations ($\mu g/m^3$) and percentages (%)

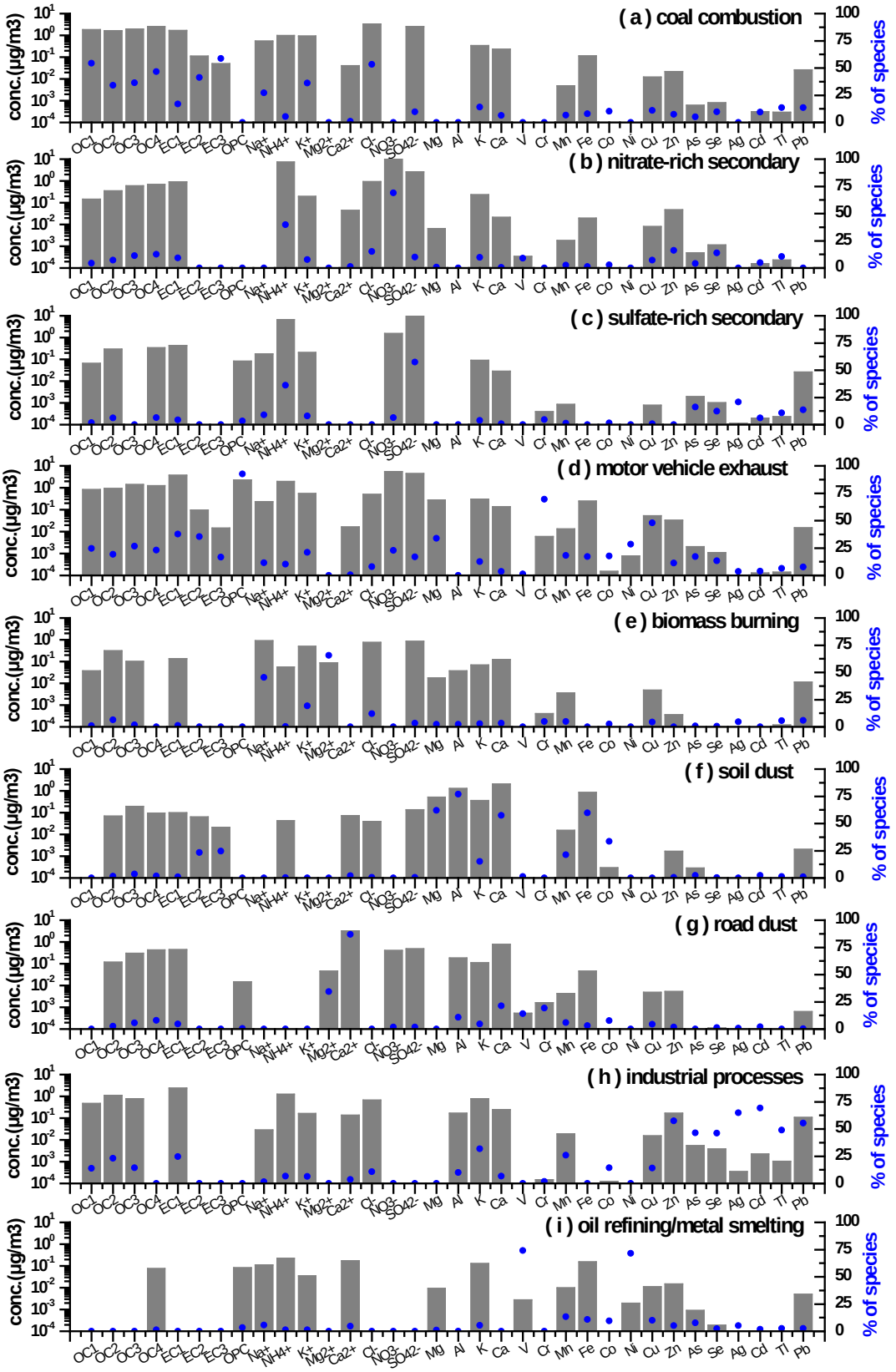

Figure S4. PMF factor/source profiles for $PM_{2.5}$ samples throughout the entire study period in Shijiazhuang in terms of concentrations ($\mu g/m^3$) and percentages (%)

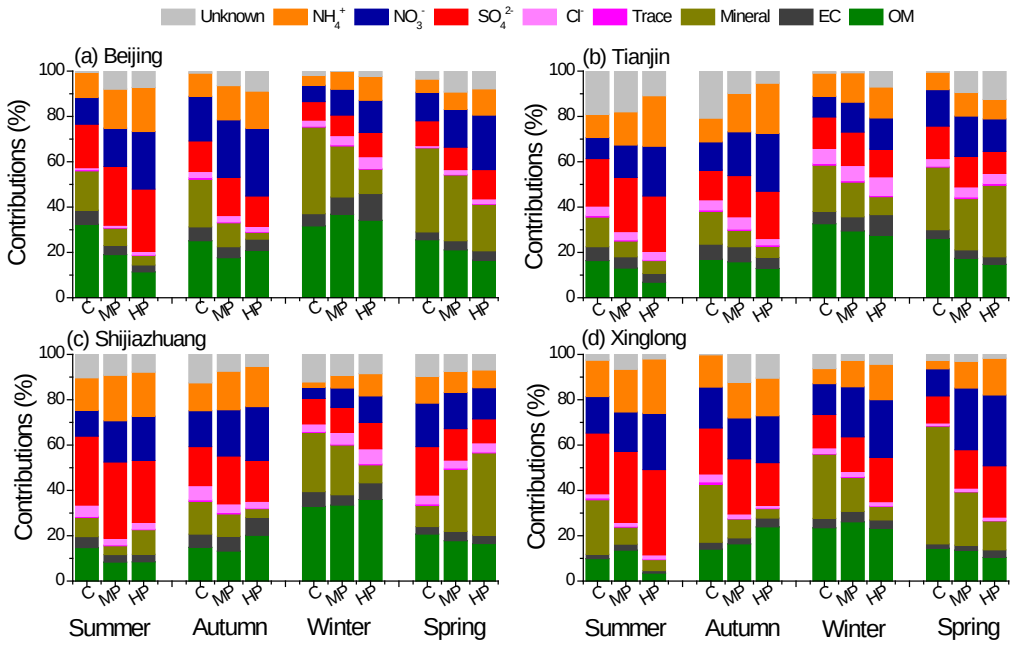

Figure S5. The mass fraction of each aerosol chemical species at different pollution levels throughout the entire study period. C, MP, and HP represent clean days ($PM_{2.5}$＜75 μg/m^3), moderate pollution days (75≤$PM_{2.5}$＜150 μg/m^3) and heavy pollution days ($PM_{2.5}$≥150 μg/m^3), respectively. "%" represents the proportion of filter sample quantity at each pollution level to the total samples.

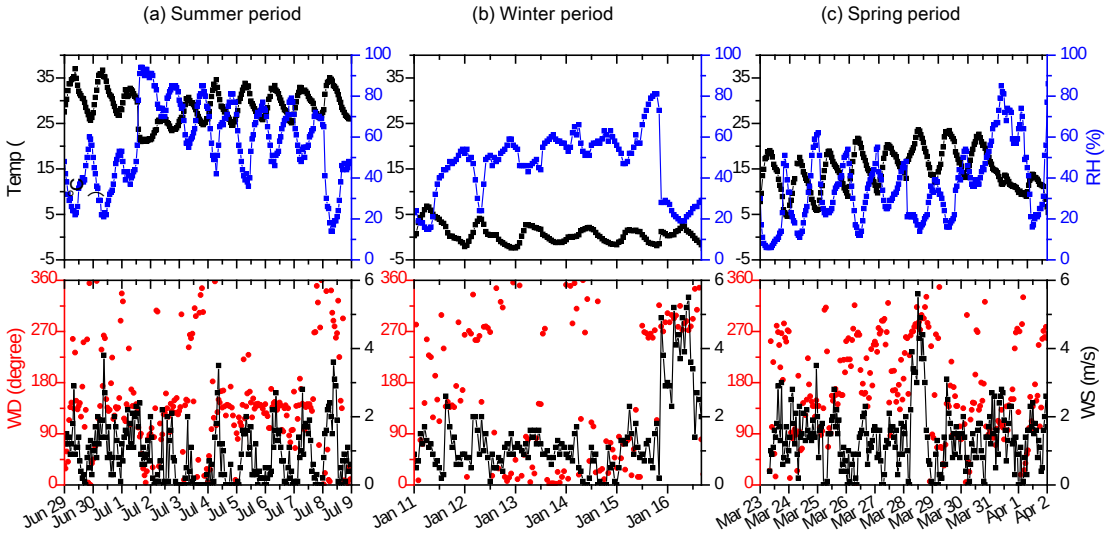

Figure S6. Time series variations of the meteorological parameters from Beijing during the specific pollution periods in summer (a), winter (b) and spring (c).

Supplementary text for describing the day-night variations of $PM_{2.5}$ and major chemical components

The analysis of the day-night variations indicates that the differences in the $PM_{2.5}$ annual average concentrations during the day and those during the night were significant at the urban sites, where the values were 8–19% higher at night than those during the day, while negligible differences were found on the annual scale in Xinglong (Fig. S7). This obvious day-night variation of the $PM_{2.5}$ concentrations in urban areas was probably due to the apparent changes in the height of the mixing layer between day and night (Zhao et al., 2009). However, in Xinglong, the dominant source was from the regional or long-range transport, with fewer contributions from local emissions; thus, the nocturnal stable boundary layer could have reduced the quantity of transmissions from the outside. The chemical compositions also recorded obvious day-night variations, as the mass ratio of NO_3^-/SO_4^{2-}recorded higher values at night (0.99–1.39) than during the day (0.81–1.13), which is consistent with the similar results obtained by Sun et al. (2016) in Xianghe, which is located approximately 50 km southeast of Beijing. Such day-night variations indicate the important role of the gas-phase photochemical production of sulfate during the day while the facilitated gas-to-particle partitioning of semi-volatile nitrate is associated with the low temperatures (Sun et al., 2016) and effective hydrolysis of N_2O_5 at night, which is a major source of nitric acid in the urban atmosphere during the night and is more efficient on wet surfaces (Zhang et al., 2015). In addition, the relatively static and stable meteorological conditions at night resulted in obviously lower fractions of mineral dust (11.3–17.0%, except for in Tianjin), than those recorded during the day (18.3–24.3%).

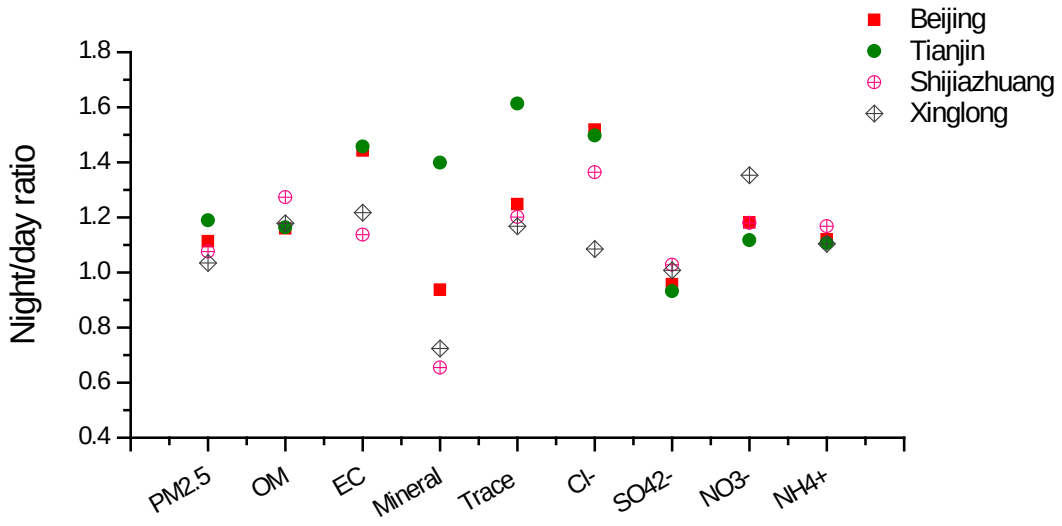

Figure S7. Day-night variations of $PM_{2.5}$ and major chemical components, based on annual data

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