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Supplement of

Direct observations of organic aerosols in common wintertime hazes in North China: insights into direct emissions from Chinese residential stoves

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1 The statistics about air quality during the whole winter of 2014 in NCP

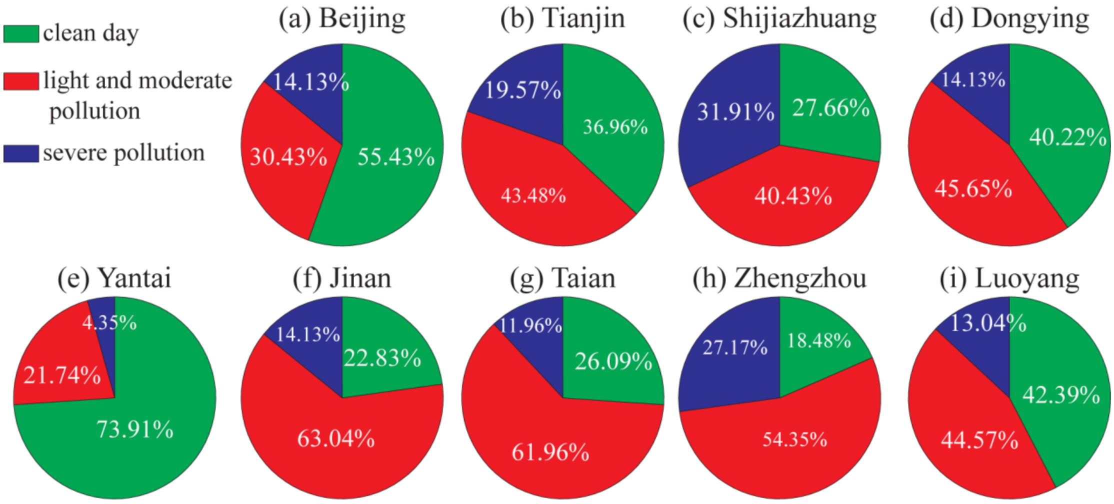

Figure S1. The statistics about air quality during the whole winter (~ 92 days) of 2014-2015 in nine representative cities in the NCP. Green color represents clean day ($PM_{2.5}$ < 75 μg m^{-3}), red color represents light and moderate pollution (75~250 μg m^{-3}) and blue color represents severe pollution ($PM_{2.5}$ > 250 μg m^{-3}). The light and moderate haze day account for 30-63% but the severe haze days is only about 4-32% in NCP.

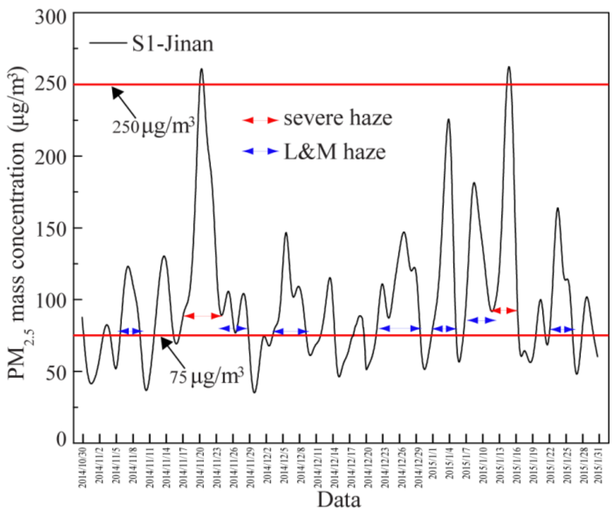

Figure S2. The timescale of haze events in Jinan city based on the statistic data.

2 The relationship between EVD and ECD obtained by AFM

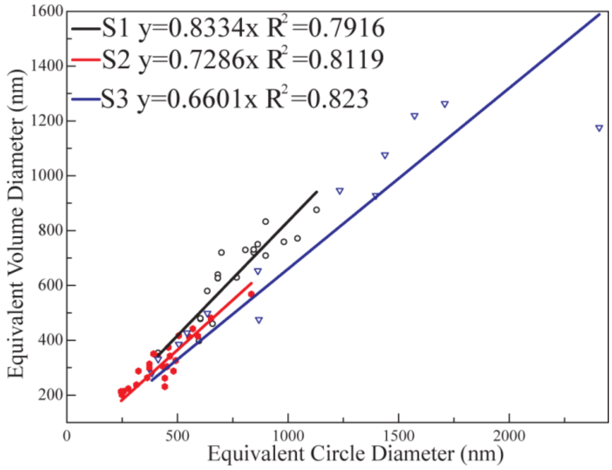

Figure S3. The correlation of equivalent circle diameter (ECD, x) and equivalent volume diameter (EVD, y) at three sampling sites obtained by AFM.

$$A = \pi r^2 = \pi \left(\frac{x}{2}\right)^2 = \frac{\pi x^2}{4} \rightarrow x = \sqrt{\frac{4A}{\pi}} \tag{1}$$

$$V = \frac{4}{3}\pi r^3 = \frac{4}{3} \times \frac{\pi y^3}{8} = \frac{\pi y^3}{6} \rightarrow y = \sqrt[3]{\frac{6V}{\pi}} \tag{2}$$

Where x is the equivalent circle diameter (ECD) and y is the equivalent volume diameter (EVD).

3 The daily average $PM_{2.5}$ mass concentration during sampling period

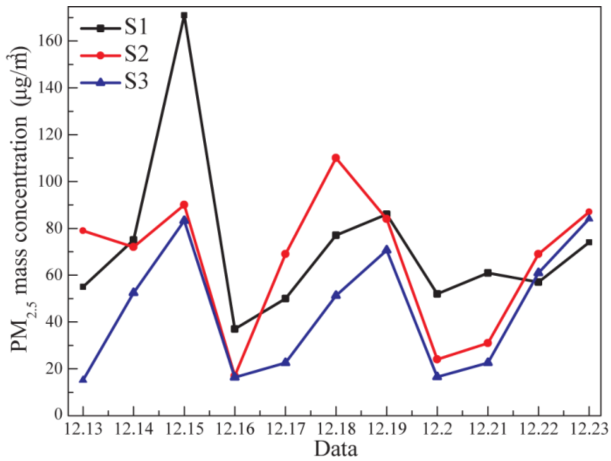

Figure S4. The series daily average $PM_{2.5}$ mass concentrations at three sampling sites in 13-23 December, 2014. We can see three haze events occurred on 14-15, 18-19, 23 December.

4 RH at three sampling sites in winter, 2014

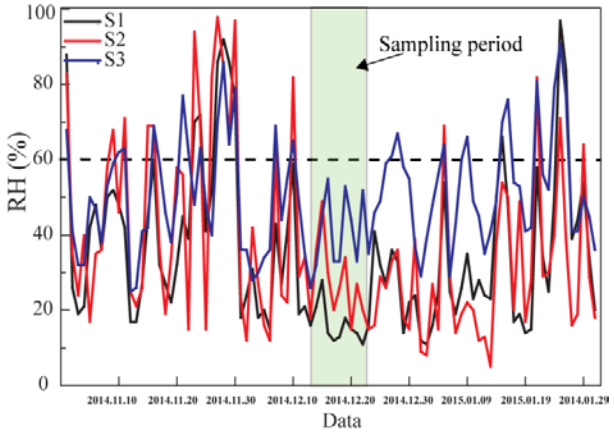

Figure S5. RH at three sampling sites in winter, 2014. We can see that the daily average RH during whole wintertime in NCP were at a low-level. Especially during the sampling period, lower than 60%.

5 Information about samples

Table S1. Average concentrations of aerosol particles and various trace gases during clean and haze periods at three sampling sites.

site	Air Quality	OC ($\mu g/m^3$)	EC ($\mu g/m^3$)	$PM_{2.5}$ ($\mu g/m^3$)	Water Soluble Ions ($\mu g/m^3$)	OC/EC	$TC/PM_{2.5}$	Water Soluble Ions $/PM_{2.5}$	SO_2 (ppb)	CO (ppm)	NO_2 (ppb)	O_3 (ppb)
S1	Clean day	12.2	2.5	60	28.1	4.9	0.24	0.47	32.6	0.8	26.1	23.5
	Haze day	22.1	4.1	97	50.2	5.5	0.27	0.52	33.7	0.96	29.9	22.8
S2	Clean day	4.4	0.8	48	14.9	5.1	0.14	0.31	21.8	1.1	24.2	32.1
	Haze day	9.8	1.5	89	34.5	6.8	0.15	0.39	28.8	1.5	33.9	25.8
S3	Clean day			32					14.5	0.7	17.0	24.7
	Haze day			80					21.8	1.3	29.1	20.7

From Table S1, we found that the average concentration of OC, EC, OC/EC and water-soluble ions were much higher on haze days than those on clear days. For example, at S2, total carbon (TC) and water-soluble ions accounting for an average of 15% and 39% of $PM_{2.5}$ mass concentration on haze days and 14% and 31% on clear days, respectively; OC was 9.8 μg m^{-3}, more than 2 times higher than on clean days (4.0 μg m^{-3}); EC was 1.5 μg m^{-3}, about 2 times higher than that on clean days (0.8 μg m^{-3}); The OC/EC ratio was elevated (6.75) during haze periods, lower value (5.07) were found during clean periods. At S1, TC and water-soluble ions accounting for an average of 27% and 52% of $PM_{2.5}$ mass concentration on haze days and 24% and 47% on clear days; OC was 22.1 μg m^{-3}, 1.7 times higher than that on clean days (12.2 μg m^{-3}); EC was 4.1μg m^{-3}, 1.6 times higher than that on clean days (2.5 μg m^{-3}). The OC/EC ratio was 5.52 on haze days, higher than 4.87 on clean days. Guinot et al (2007) and Yao et al. (2016) reported that the ratios of OM/OC for urban and non-urban areas in NCP were 1.4-1.8 (1.7 in Beijing) and 2.07. As a result, OM was calculated using 1.5 times of OC at S1 and 2.07 times of OC at S2.

Compared with S2, there were higher OC, EC and water-soluble ions concentration and lower OC/EC ratio at S1. The higher OC/EC ratio at S2 was correlated with the formation of secondary organic carbon at remote region. In addition, the mass concentration of SO_2, CO, NO_2 were little higher and O3 was little lower on haze days at all sampling sites (Table S1).

Table S2. Sampling dates, sampling times, and meteorological conditions for 33 individual particle samples.

Sampling site	No.	Sampling date	Sampling time	T (℃)	RH (%)	Wind speed (m/s)	Wind direction
S1	1	2014.12.13	13:00-13:02	6.5	15.2	2.2	265
	2	2014.12.14	7:40-7:42	3.4	30.9	0.1	322
	3	2014.12.15	7:40-7:42	3.8	54.7	0.7	90
	4	2014.12.16	19:40-19:45	0.5	22.0	2.7	269
	5	2014.12.17	19:00-19:04	4.5	22.3	0.6	222
	6	2014.12.18	12:30-12:35	10.3	16.9	1.2	191

	7	2014.12.19	7:40-7:45	6.5	30.8	3.3	235
	8	2014.12.20	20:00-20;05	-0.7	27.9	2.2	250
	9	2014.12.21	7:30-7:35	-4.2	36.9	0.8	112
	10	2014.12.22	18:30-18:40	1	30	0.3	320
	11	2014.12.23	7:30-7:40	6.5	28.9	1.3	202
S2	1	2014.12.13	16:00-16:20	-2.9	48.3	0	264
	2	2014.12.14	8:55-9:15	-4.6	78	2.4	206
	3	2014.12.15	9:10-9:25	-12.8	70	2.3	250
	4	2014.12.16	16:10-16:30	-0.4	45.9	0.9	320
	5	2014.12.17	15:34-16:04	-0.3	57.1	1.8	253
	6	2014.12.18	15:40-16:10	0.3	58.1	2.8	231
	7	2014.12.19	8:10-8:30	-12.3	46.9	1.8	53
	8	2014.12.20	9:00-9:20	-6.7	52.6	1.1	210
	9	2014.12.21	8:42-9:12	-0.8	51.2	0.6	323
	10	2014.12.22	15:36-16:06	-1.4	47.1	1.3	209
	11	2014.12.23	8:36-9:06	-3.7	48.7	0	170
S3	1	2014.12.13	15:57-16:09	0.2	43.7	0.5	335
	2	2014.12.14	7:28-7:37	1.8	61.5	2.6	242
	3	2014.12.15	15:58-16:07	2.1	62.2	3	9
	4	2014.12.16	15:56-16:07	-3.4	52.8	0.4	210
	5	2014.12.17	16:04-16:13	1	49	4.5	278
	6	2014.12.18	7:08-7:17	1.5	49.4	3.4	136
	7	2014.12.19	7:06-7:17	3.3	57.6	1.4	356
	8	2014.12.20	7:08-7:14	-1.3	55.4	1.7	203
	9	2014.12.21	15:56-16:02	-2.4	48.5	1.1	112
	10	2014.12.22	16:04-16:12	3.6	70.1	0.7	178
	11	2014.12.23	7:05-7:13	2.6	61.4	1.4	159

6 Low view of TEM images in haze days

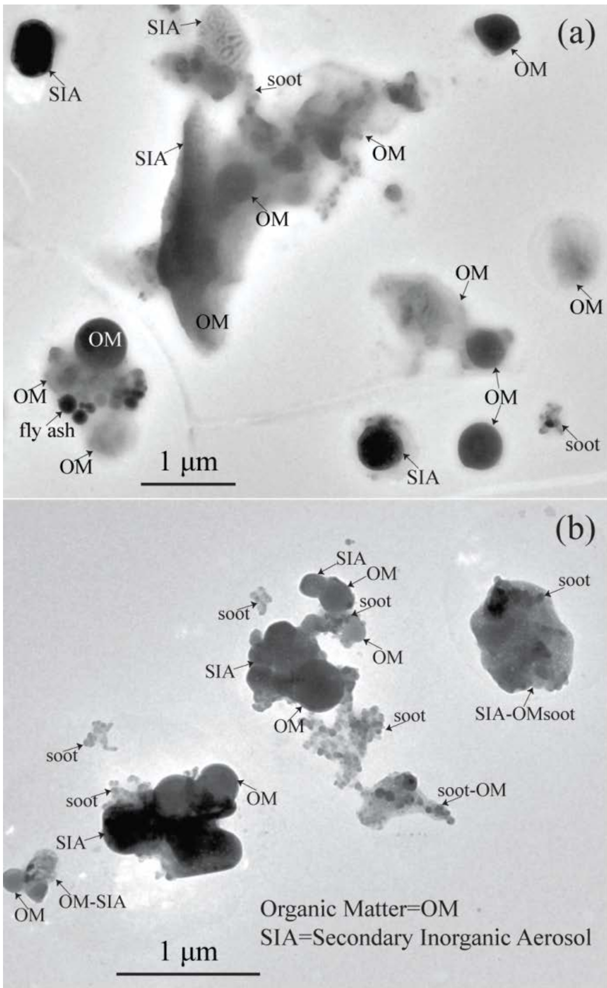

Figure S6. Individual particles during haze periods with $PM_{2.5}$ mass concentration larger than 75 μg m^{-3}. (a) low view of many particles containing elemental C collected on 15 December at S1 site. (b) low view of many particles containing elemental C collected on 18 December at S2 site.

7 NanoSIMS analysis

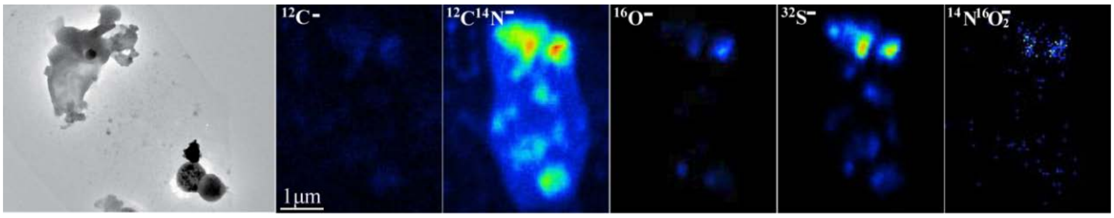

Figure S7. NanoSIMS-based ion intensity mappings of $^{12}C^-$, $^{12}C^{14}N^-$, $^{16}O^-$, $^{32}S^-$, and $^{14}N^{16}O_2^-$ from one individual OM mixture particle.

8 Comparison of OM-soot particles in three sampling sites

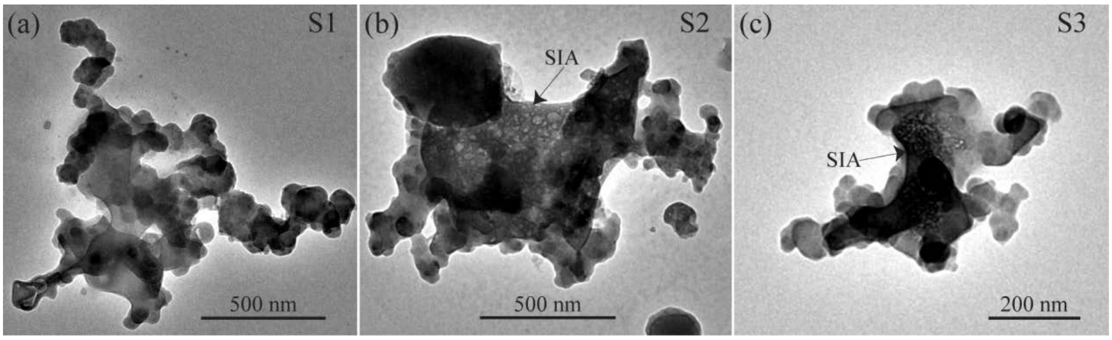

Figure S8. OM-soot particles in three sampling sites are different. (a) Exist alone and no SIA at S1 site; (b) and (c) OM-soot mixed with SIA. This is a common phenomenon during this field campaign.

9 Comparison of C, O, and Si of OM particles from different sources

OM aerosol particles produced by coal combustion and corn stalks were collected in laboratory. The elemental composition of 39 OM particles from coal combustion and 28 OM particles from corn stalks combustion were analyzed using the EDX. The ratios of Si/C and Si/O in them were calculated. The same information of 281 individual OM particles in light and moderately haze samples were also obtained. The three lines represent the links between average ratios of Si/O and Si/C in OM particles. We calculated the fraction of haze OM particles with lower ratios of Si/O and Si/C than coal combustion, and found that 71% associated with coal line.

References

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