

1 *Supplement of*

2 **Improved source apportionment of organic aerosols in complex**
3 **urban air pollution using the multilinear engine (ME-2)**

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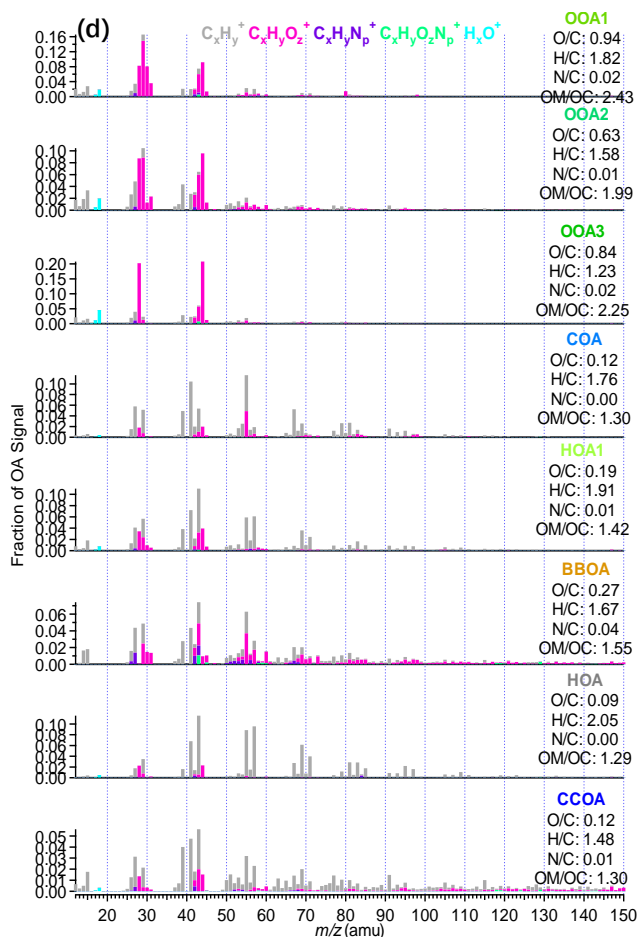
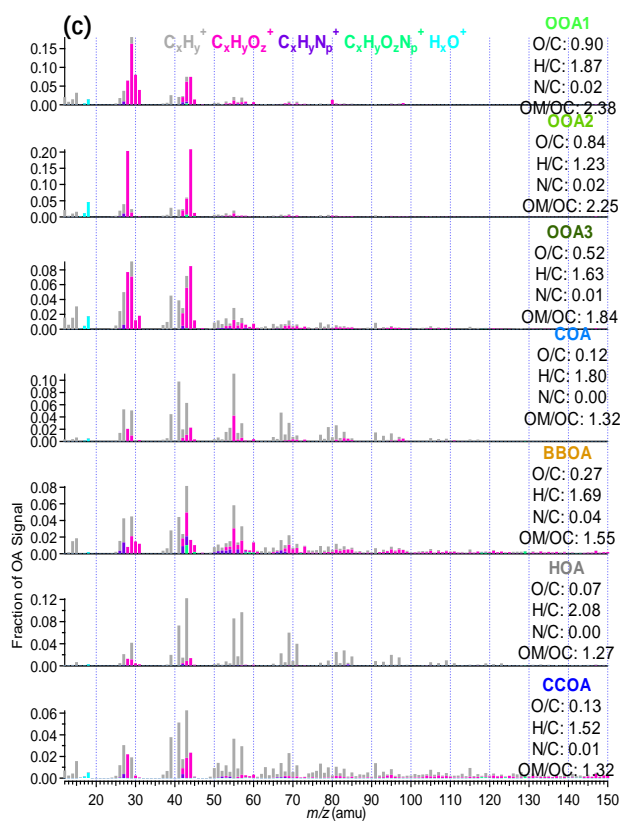
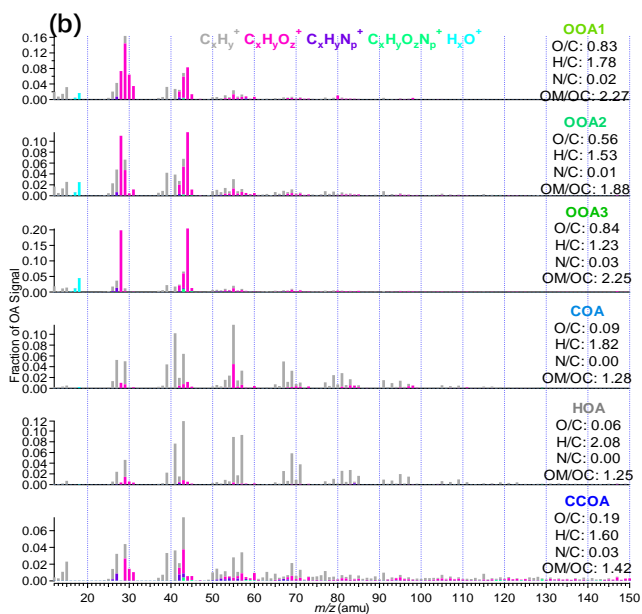
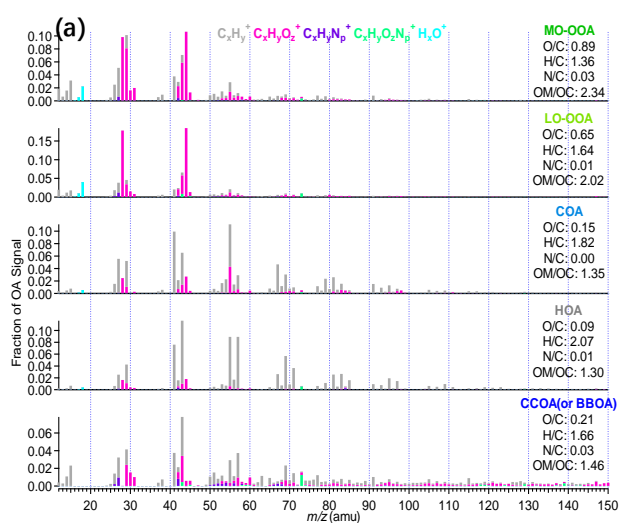
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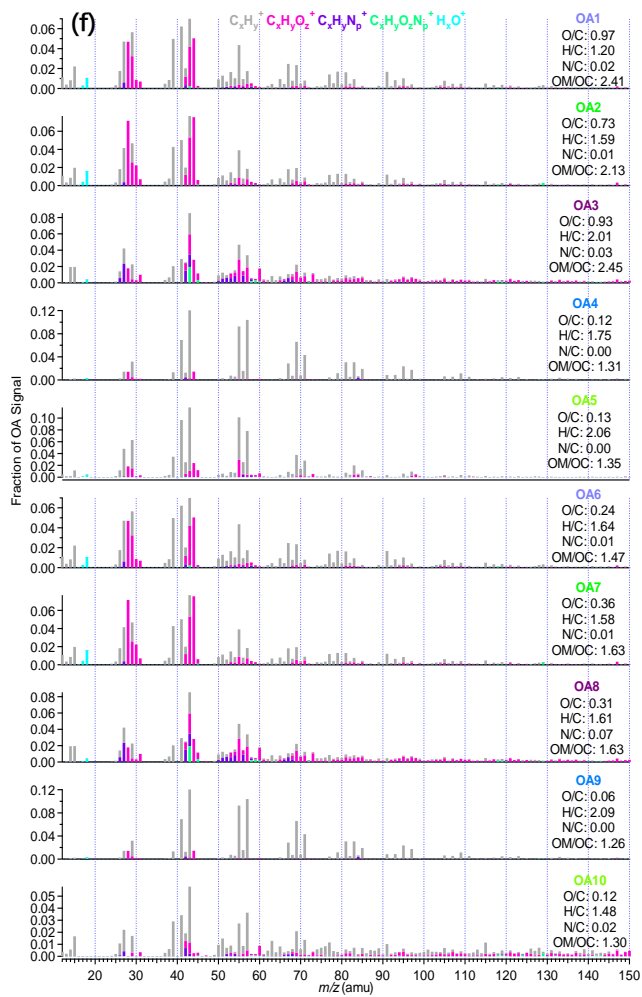
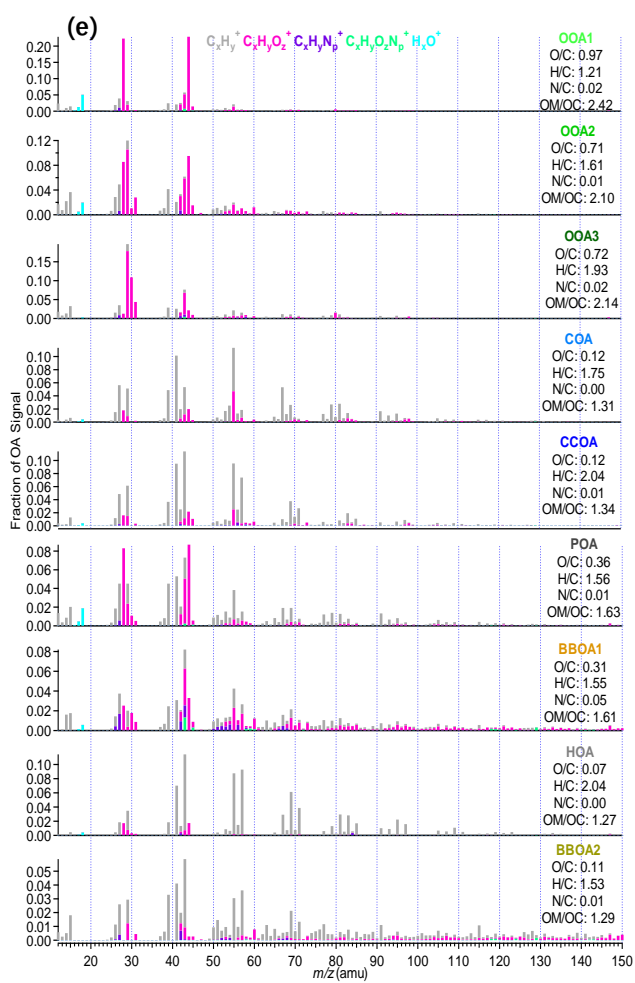
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26 **Figure S1.** Mass spectra of the (a) 5-factor, (b) 6-factor, (c) 7-factor, (d) 8-factor, (e) 9-factor, (f) 10-factor solution using

27 unconstrained PMF method in Qingdao

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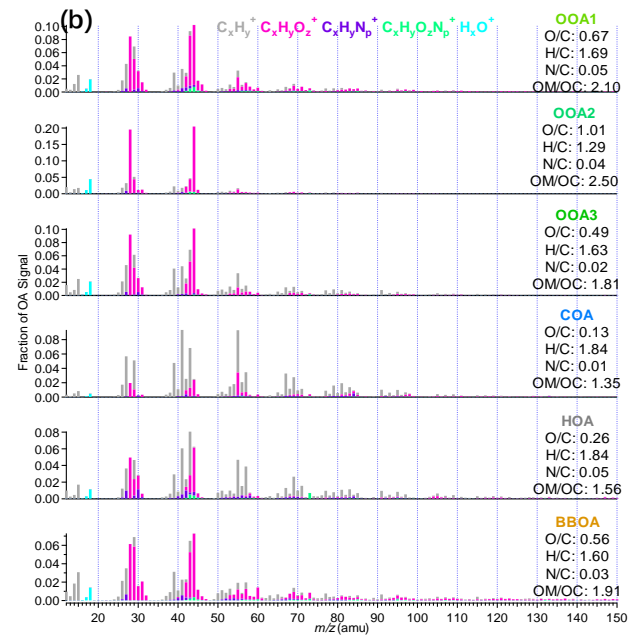
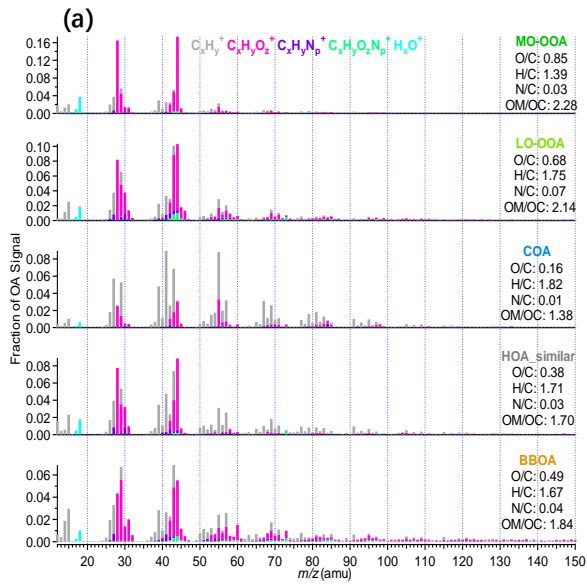
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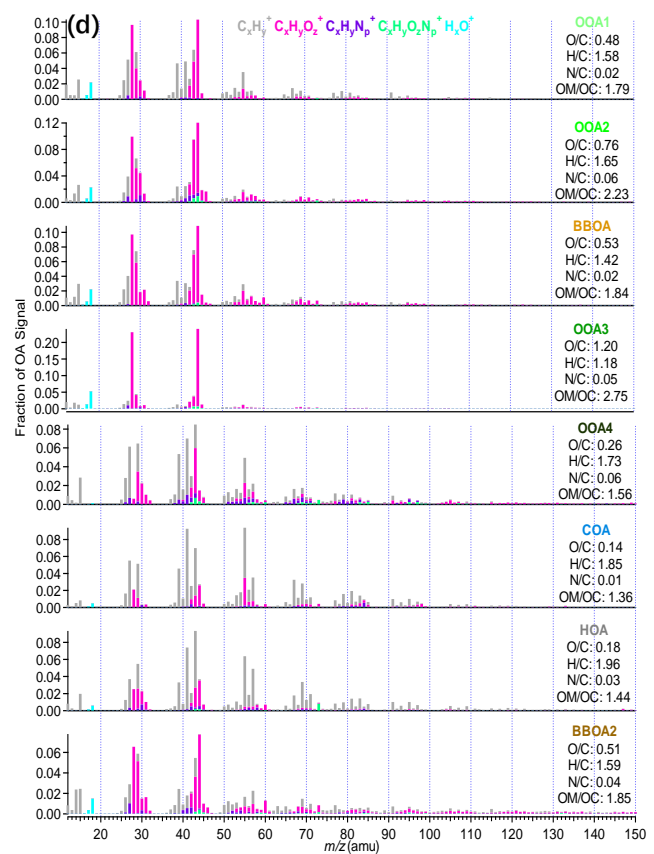
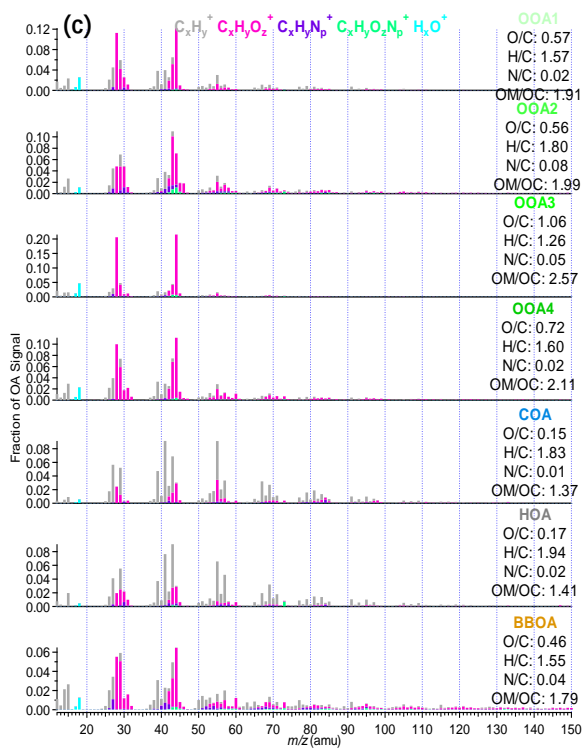
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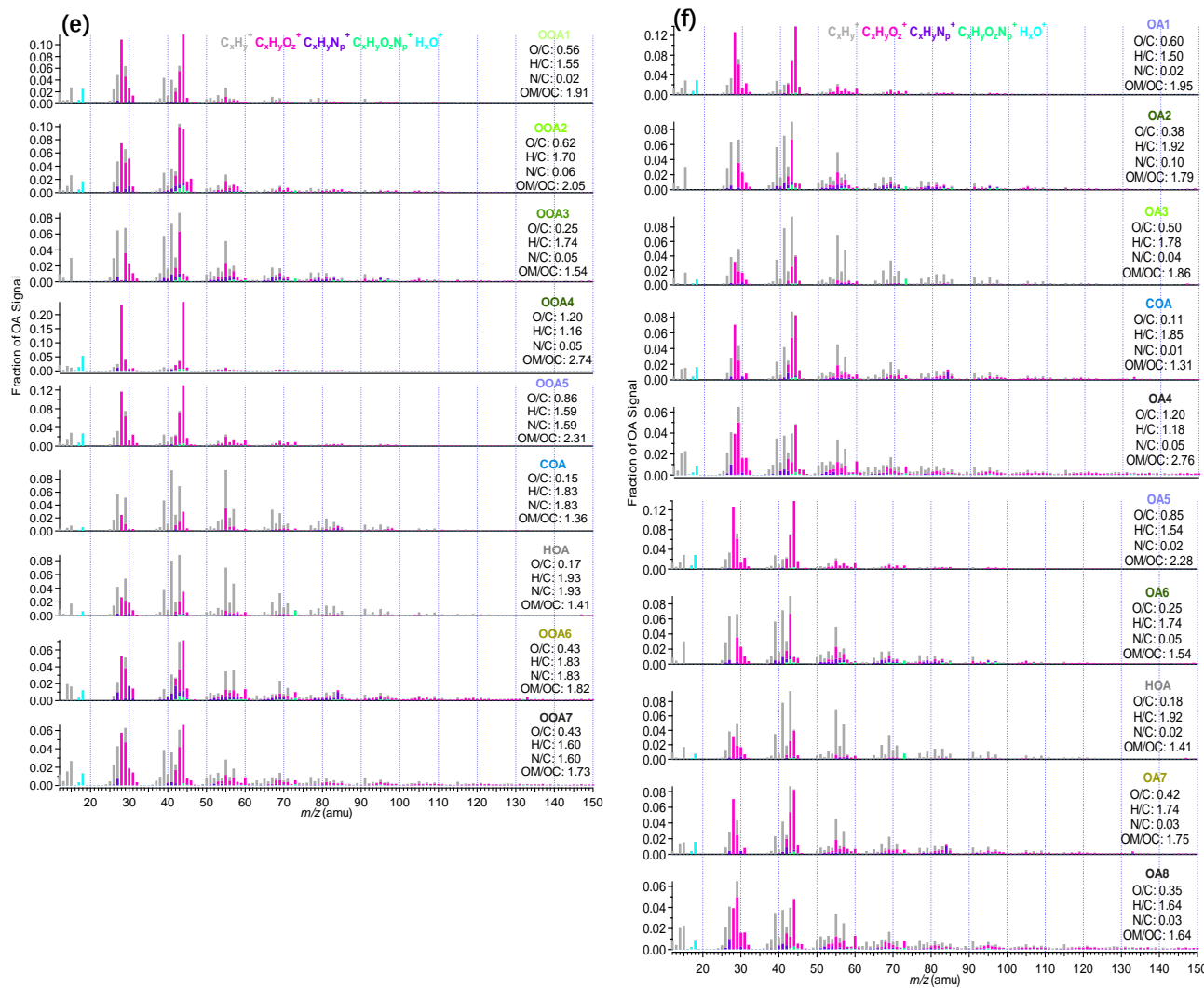
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42 **Figure S2.** Mass spectra of the (a) 5-factor, (b) 6-factor, (c) 7-factor, (d) 8-factor, (e) 9-factor, (f) 10-factor solution

43 using unconstrained PMF method in Dongguan

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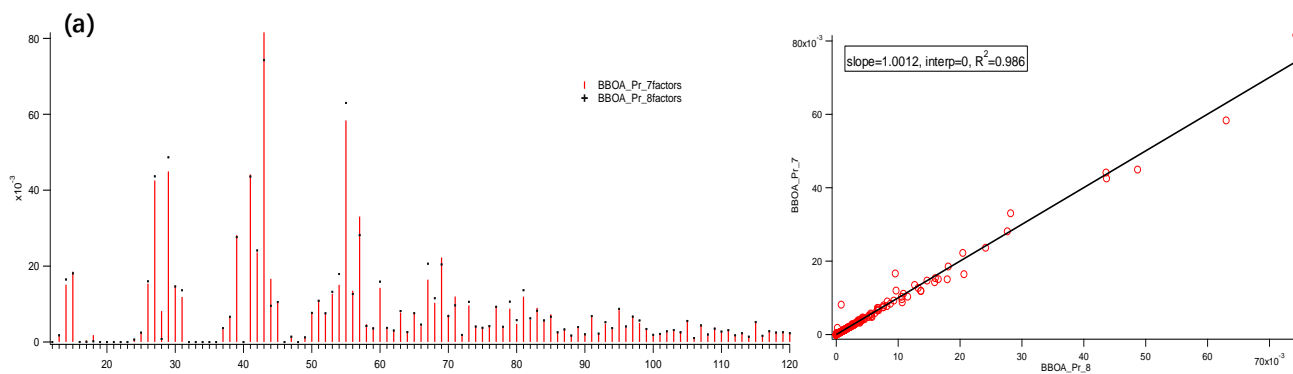
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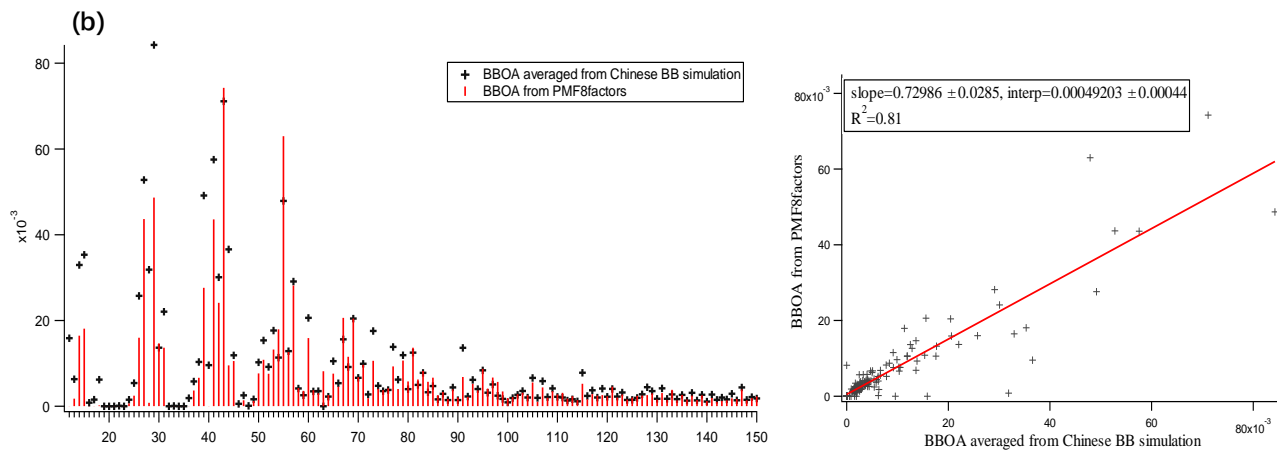
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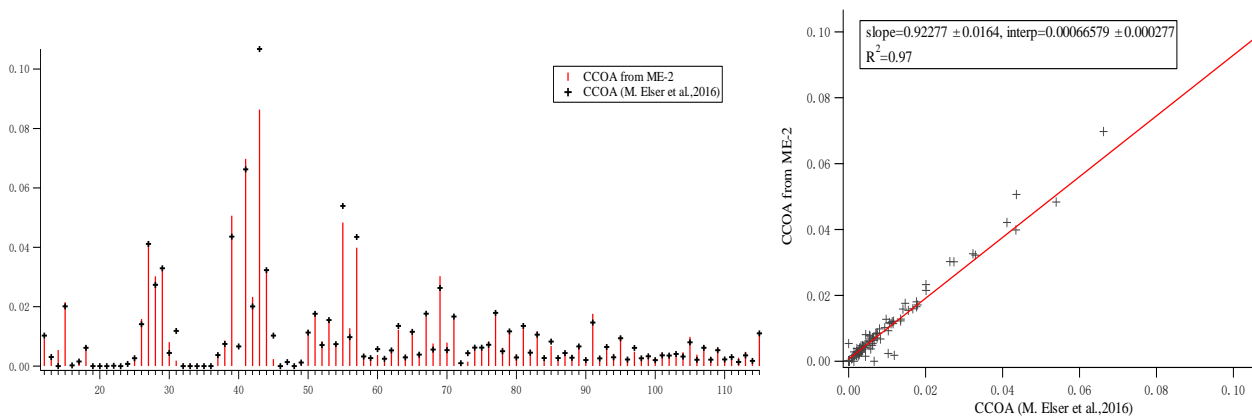


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55 **Figure S3.** (a) Correlation between BBOA in 7- and 8-factor solution resolved from PMF in Qingdao; (b) Correlation
 56 between BBOA in 8-factor solution resolved from PMF in Qingdao and BBOA averaged from BB simulation.

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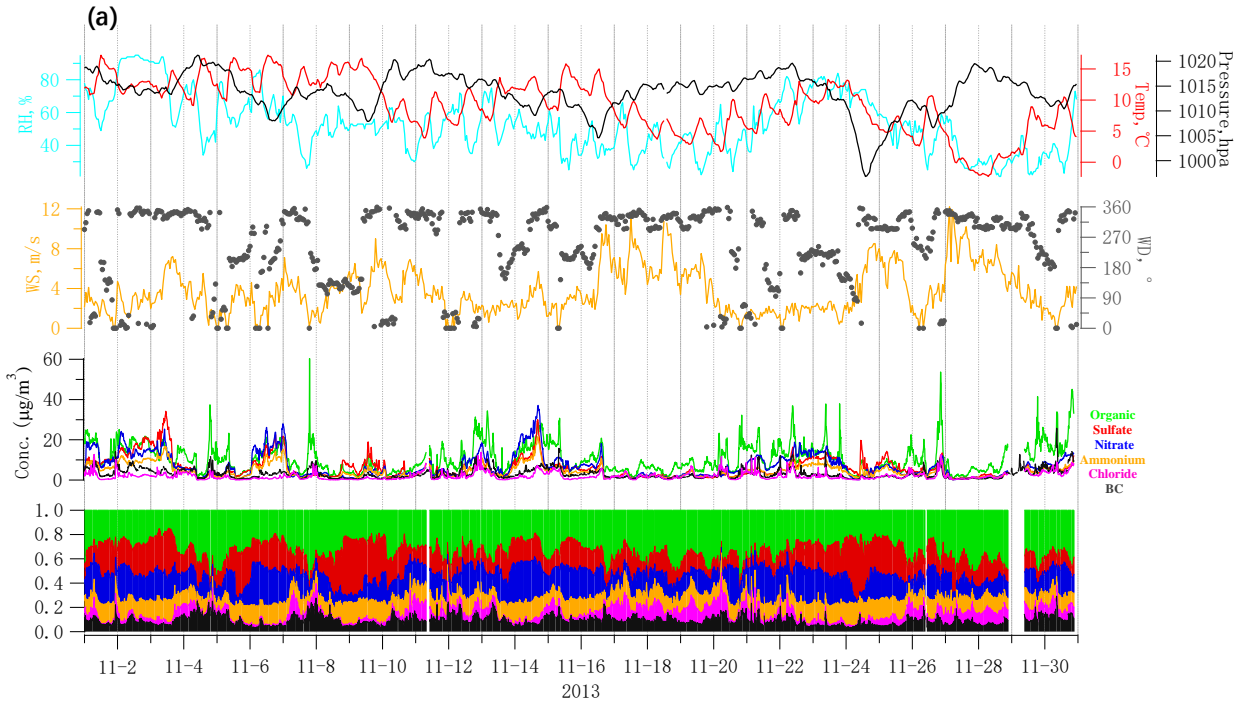
58 **Figure S4.** Correlation between CCOA resolved from ME-2 in Qingdao and CCOA reported in Elser et al., 2016

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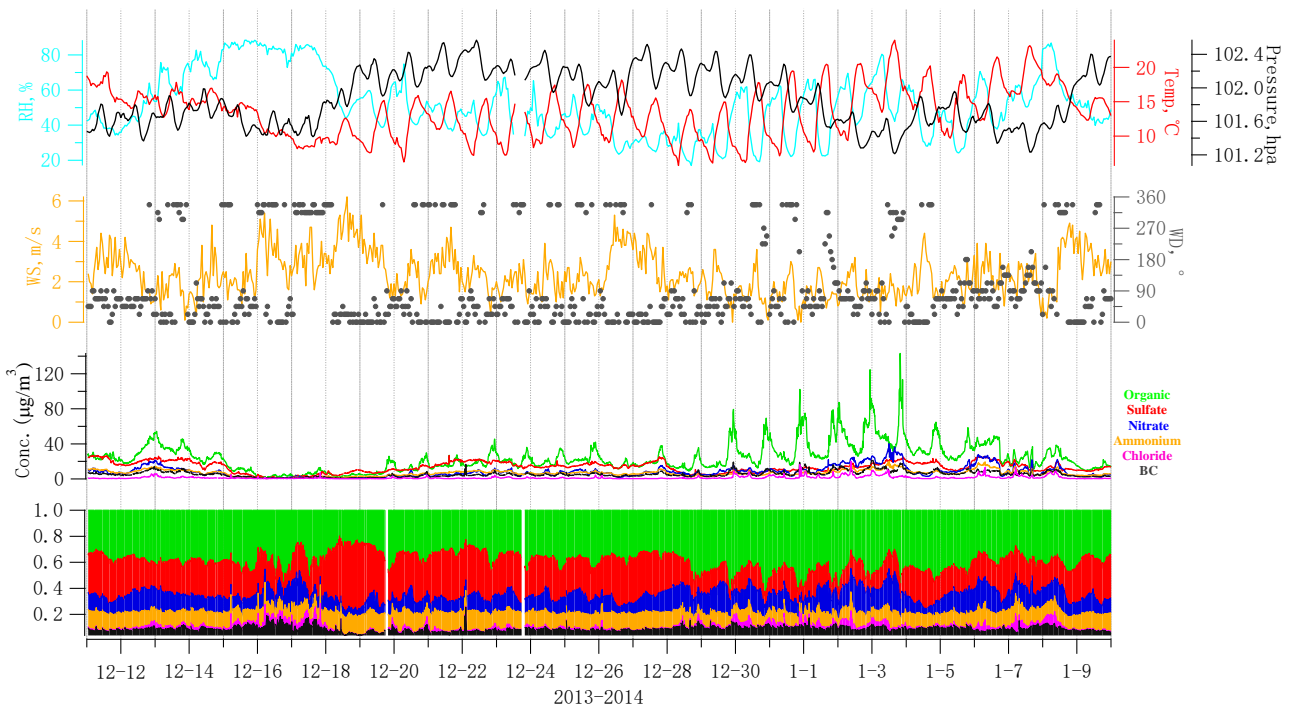
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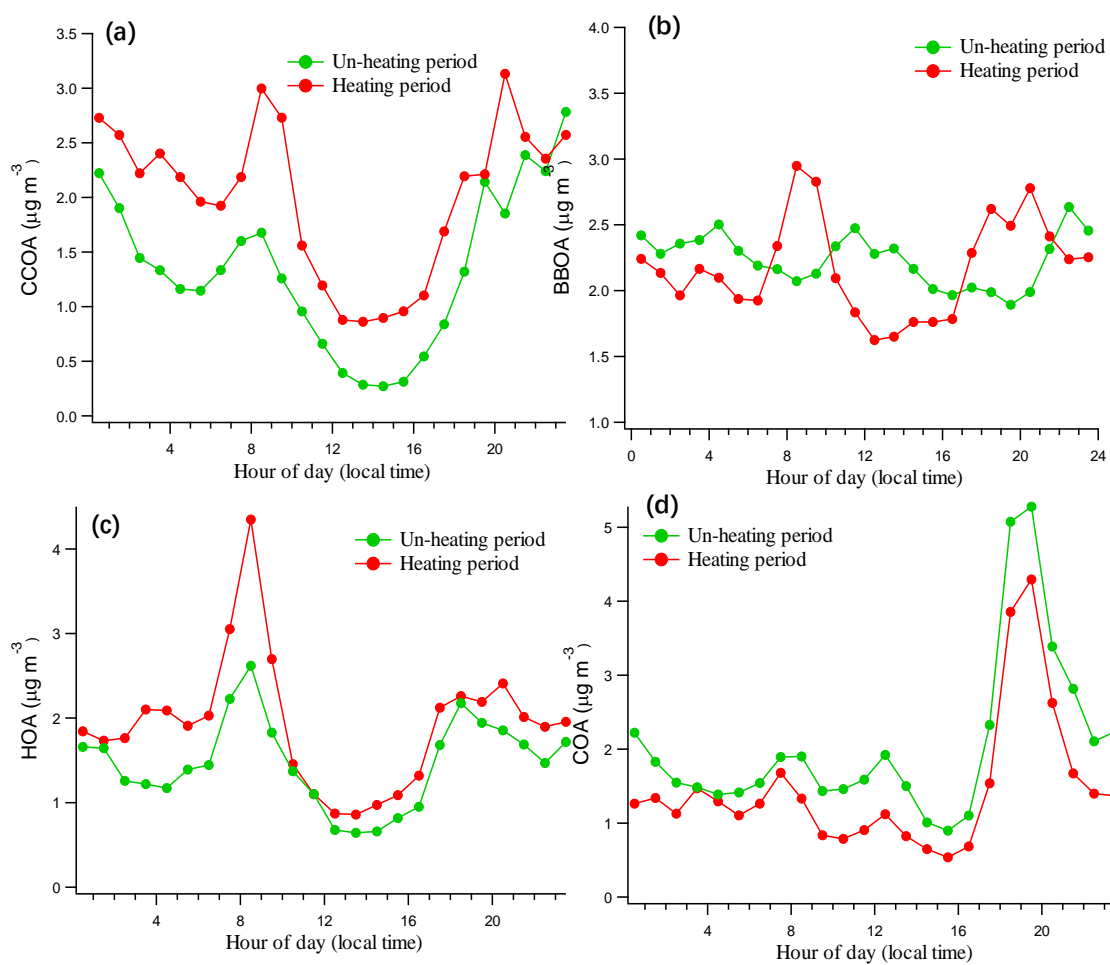
(b)



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65 **Figure S5.** Time series of meteorological parameters (relative humidity, temperature, pressure, wind speed and wind
 66 direction), NR-PM₁ compounds and relative contribution of the different components in Qingdao (a) and Dongguan (b)

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Figure S6. Diurnal variation of the (a) CCOA; (b) BBOA; (c) HOA and (d) COA before and during the central-heating period in Qingdao.

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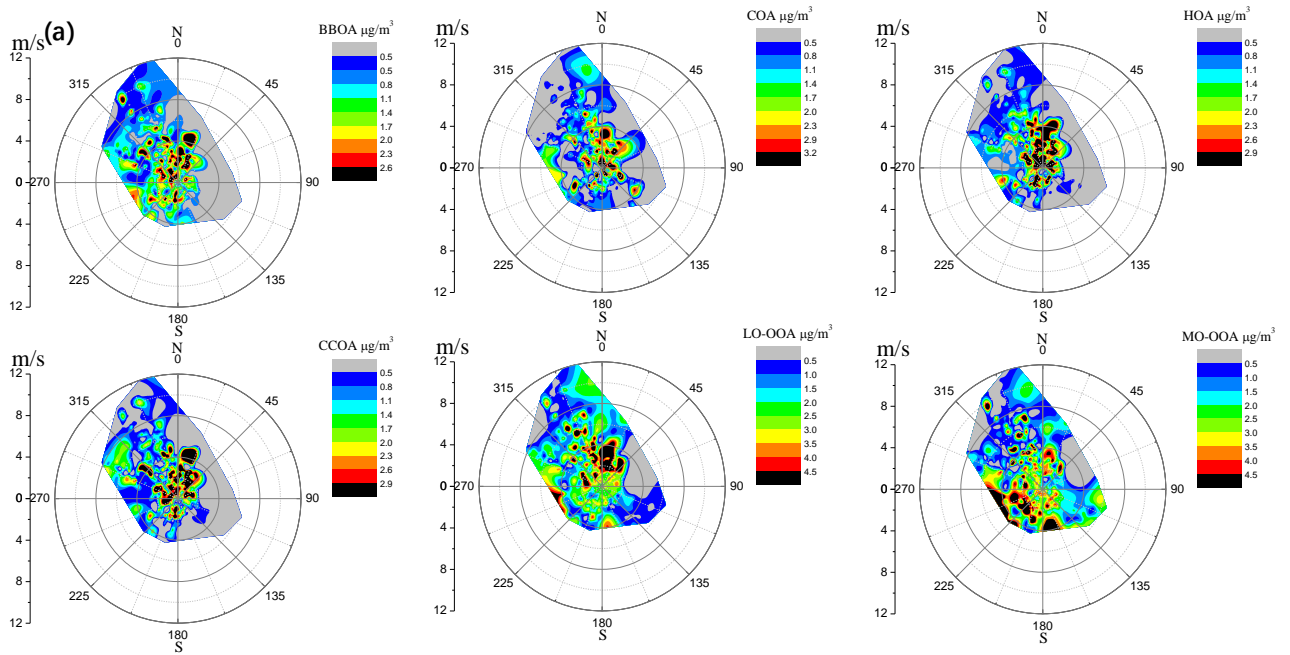
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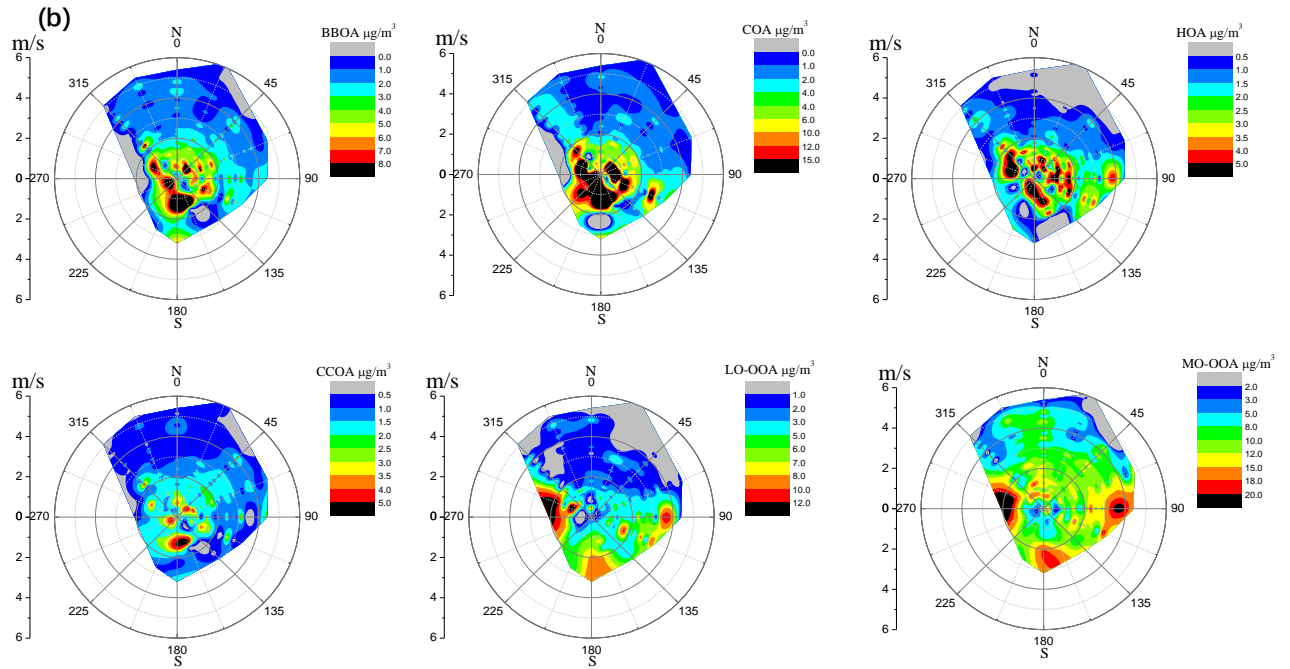
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84 **Figure S7.** The variation of OA sources concentration with wind direction and speed in Qingdao (a) and Dongguan (b).

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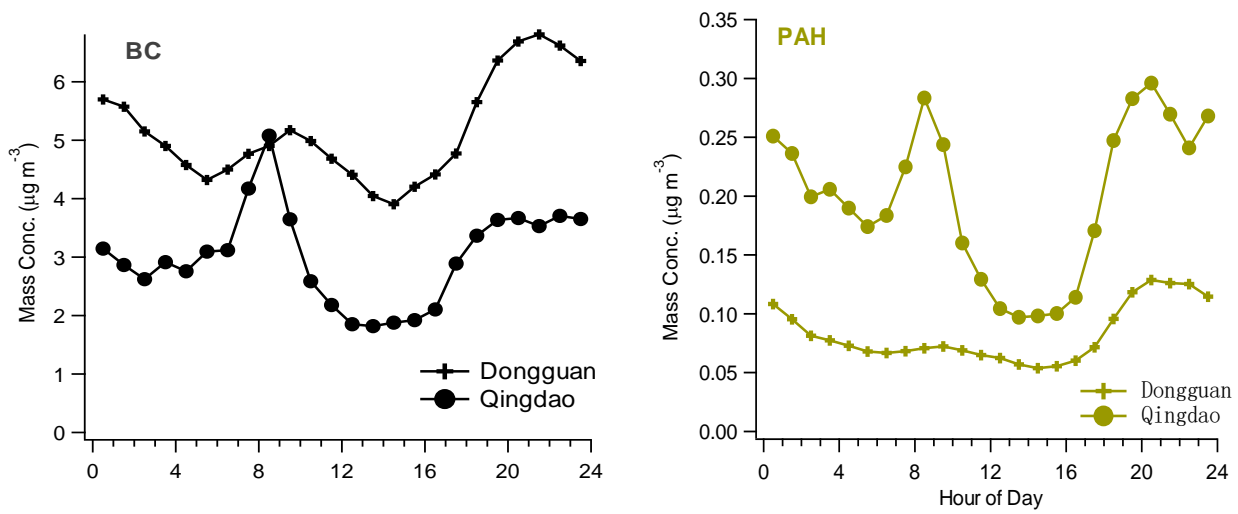


Figure S8. The diurnal trends of BC and PAHs in Qingdao and Dongguan.

Table S1. The O/C ratios for each resolved factor with an a value from 0 to 1 for CCOA and from 0 to 0.1 for HOA using the ME-2 method for Dongguan.

a value for CCOA \ a value for HOA	0	0.1	0	0.1	0	0.1	0	0.1	0	0.1	0	0.1
	HOA	CCOA	COA	BBOA	LO-OOA	MO-OOA						
0	0.17	0.17	0.12	0.12	0.17	0.16	0.59	0.59	0.80	0.80	0.81	0.82
0.1	0.17	0.17	0.13	0.13	0.17	0.16	0.59	0.61	0.78	0.79	0.82	0.82
0.2	0.17	0.17	0.13	0.12	0.17	0.17	0.61	0.61	0.80	0.78	0.82	0.83
0.3	0.17	0.17	0.13	0.12	0.18	0.18	0.61	0.61	0.78	0.78	0.83	0.84
0.4	0.17	0.18	0.13	0.12	0.18	0.17	0.62	0.62	0.76	0.69	0.85	0.86
0.5	0.17	0.18	0.12	0.12	0.18	0.18	0.62	0.61	0.77	0.76	0.84	0.85
0.6	0.17	0.18	0.12	0.12	0.18	0.19	0.62	0.61	0.75	0.72	0.85	0.86
0.7	0.17	0.18	0.13	0.12	0.18	0.17	0.62	0.62	0.70	0.70	0.86	0.86
0.8	0.17	0.18	0.14	0.13	0.19	0.18	0.61	0.60	0.70	0.66	0.86	0.87
0.9	0.17	0.18	0.18	0.18	0.18	0.18	0.61	0.59	0.63	0.65	0.90	0.87
1	0.17	0.18	1.25	1.25	0.11	0.12	0.38	0.39	0.63	0.64	0.86	0.86

97 **Table S2.** The O/C ratio for each resolved factor and correlation parameters between POAs and their tracers with *a* value
 98 of 0 to 1 for BBOA using ME-2 method in Qingdao.

<i>a</i> value for BBOA	BBOA	CCOA	HOA	COA	LO-OOA	MO-OOA	CCOA vs PAHs (R ²)	HOA vs BC (R ²)
0	0.27	0.14	0.06	0.14	0.59	0.89	0.94	0.65
0.1	0.27	0.15	0.06	0.14	0.60	0.89	0.95	0.66
0.2	0.27	0.15	0.06	0.14	0.60	0.87	0.95	0.66
0.3	0.27	0.15	0.06	0.14	0.61	0.89	0.95	0.65
0.4	0.27	0.15	0.06	0.14	0.61	0.87	0.95	0.65
0.5	0.27	0.16	0.07	0.14	0.61	0.87	0.96	0.65
0.6	0.27	0.19	0.06	0.14	0.60	0.86	0.96	0.65
0.7	0.27	0.20	0.06	0.14	0.61	0.86	0.96	0.65
0.8	0.27	0.19	0.07	0.14	0.61	0.87	0.96	0.65
0.9	0.27	0.19	0.07	0.14	0.61	0.87	0.96	0.66
1	0.27	0.19	0.07	0.14	0.61	0.87	0.96	0.65

99 **Table S3(a).** The O/C ratio for HOA with different *a* value sets using ME-2 method in Dongguan.

<i>a</i> value for HOA	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1
0	0.17	0.17	0.19	0.19	0.24	0.22	0.25	0.32	0.28	0.36	0.42
0.1	0.17	0.17	0.19	0.22	0.23	0.25	0.28	0.32	0.36	0.39	0.42
0.2	0.17	0.17	0.20	0.22	0.23	0.25	0.28	0.32	0.37	0.39	0.42
0.3	0.17	0.17	0.20	0.22	0.23	0.27	0.28	0.33	0.37	0.40	0.42
0.4	0.17	0.18	0.20	0.22	0.23	0.25	0.28	0.33	0.37	0.38	0.49
0.5	0.17	0.18	0.20	0.22	0.24	0.24	0.29	0.33	0.38	0.38	0.45
0.6	0.17	0.18	0.20	0.22	0.24	0.26	0.29	0.34	0.36	0.45	0.54
0.7	0.17	0.18	0.20	0.20	0.24	0.22	0.26	0.35	0.37	0.41	0.56
0.8	0.17	0.18	0.20	0.22	0.24	0.26	0.29	0.35	0.32	0.48	0.42
0.9	0.17	0.18	0.20	0.20	0.23	0.22	0.29	0.33	0.37	0.40	0.42

1	0.17	0.18	0.19	0.21	0.19	0.20	0.20	0.36	0.36	0.39	0.50
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Table S3(b). The O/C ratio for CCOA with different a value sets using ME-2 method in Dongguan.

a value for HOA a value for CCOA	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1
0	0.12	0.12	0.14	0.14	0.14	0.14	0.14	0.14	0.14	0.14	0.14
0.1	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.14	0.13
0.2	0.13	0.12	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.13
0.3	0.13	0.12	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.13	0.13
0.4	0.13	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.16	0.16
0.5	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.12
0.6	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.18	0.12	0.19
0.7	0.13	0.12	0.12	0.12	0.12	0.12	0.12	0.12	0.13	0.12	0.20
0.8	0.14	0.13	0.14	0.14	0.14	0.14	0.14	0.14	0.14	0.14	0.14
0.9	0.18	0.18	0.18	0.18	0.18	0.18	0.18	0.19	0.19	0.19	0.19
1	1.25	1.25	1.19	0.38	1.22	1.22	1.00	0.98	0.99	0.81	0.94

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Table S3(c). The O/C ratio for COA with different a value sets using ME-2 method in Dongguan.

a value for HOA a value for CCOA	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1
0	0.17	0.16	0.17	0.16	0.16	0.16	0.16	0.17	0.17	0.17	0.17
0.1	0.17	0.16	0.16	0.16	0.17	0.17	0.17	0.17	0.18	0.18	0.17
0.2	0.17	0.17	0.18	0.16	0.17	0.17	0.18	0.18	0.18	0.18	0.18
0.3	0.18	0.18	0.17	0.17	0.17	0.16	0.18	0.18	0.18	0.18	0.18
0.4	0.18	0.17	0.18	0.17	0.17	0.17	0.18	0.18	0.18	0.50	0.16
0.5	0.18	0.18	0.18	0.17	0.17	0.15	0.17	0.18	0.18	0.44	0.18
0.6	0.18	0.19	0.18	0.17	0.18	0.18	0.17	0.18	0.16	0.18	0.15
0.7	0.18	0.17	0.19	0.18	0.18	0.18	0.18	0.18	0.16	0.18	0.15
0.8	0.19	0.18	0.17	0.17	0.18	0.18	0.18	0.18	0.18	0.18	0.18

0.9	0.18	0.18	0.18	0.16	0.18	0.18	0.14	0.19	0.15	0.18	0.14
1	0.11	0.12	0.13	0.17	0.14	0.14	0.10	0.17	0.10	0.12	0.12

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Table S3(d). The O/C ratio for BBOA with different a value sets using ME-2 method in Dongguan.

a value for CCOA \ a value for HOA	0	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1
0	0.59	0.59	0.59	0.59	0.56	0.58	0.55	0.52	0.54	0.50	0.49
0.1	0.59	0.61	0.60	0.59	0.57	0.57	0.55	0.55	0.51	0.50	0.50
0.2	0.61	0.61	0.60	0.61	0.60	0.57	0.56	0.54	0.52	0.51	0.50
0.3	0.61	0.61	0.62	0.61	0.60	0.58	0.56	0.55	0.53	0.51	0.51
0.4	0.62	0.62	0.61	0.62	0.61	0.60	0.57	0.55	0.53	0.48	0.54
0.5	0.62	0.61	0.62	0.62	0.61	0.57	0.58	0.55	0.52	0.46	0.50
0.6	0.62	0.61	0.61	0.62	0.59	0.59	0.58	0.55	0.53	0.50	0.52
0.7	0.62	0.62	0.62	0.61	0.58	0.59	0.58	0.54	0.54	0.51	0.51
0.8	0.61	0.60	0.62	0.60	0.58	0.57	0.55	0.53	0.51	0.49	0.51
0.9	0.61	0.59	0.60	0.61	0.57	0.58	0.48	0.53	0.48	0.50	0.49
1	0.38	0.39	0.39	0.53	0.37	0.37	0.37	0.48	0.36	0.37	0.37

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Table S4. The correlation parameters (R^2) between POA factors resolved from ME-2 and their tracers in Dongguan.

a value for CCOA \ a value for HOA	0	0.1	0	0.1
	HOA vs. BC		CCOA vs. PAHs	
0	0.53	0.52	0.57	0.58
0.1	0.54	0.54	0.60	0.59
0.2	0.55	0.54	0.57	0.61
0.3	0.56	0.57	0.61	0.61
0.4	0.56	0.57	0.62	0.61

0.5	0.55	0.56	0.62	0.61
0.6	0.57	0.57	0.62	0.62
0.7	0.58	0.57	0.63	0.62
0.8	0.59	0.60	0.63	0.63
0.9	0.60	0.60	0.63	0.64
1	0.61	0.60	0.64	0.63
