



Supplement of

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

Qiao Zhu et al.

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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
using unconstrained PMF method in Dongguan

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



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



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







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





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	0	0.1	0	0.1	0	0.1	0	0.1	0	0.1	0	0.1
a value for HOA	HO	DA	CC	OA	CO	DA	BB	OA	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

$\overline{}$	BBOA	CCOA	НОА	COA	LO-OOA	MO-OOA	CCOA vs PAHs	HOA vs BC
<i>a</i> value for BBOA							(R ²)	(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

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.

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 <i>a</i> value for CCOA	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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<i>a</i> value for a value for CCOA	e HOA 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

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

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

<i>a</i> value <i>a</i> value for CCOA	HOA ⁰	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	0.9	1
0	0.17	0.16	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

1	0.11	0.12	0.13	0.17	0.14	0.14	0.10	0.17	0.10	0.12	0.12
	Table S3(o	l). The O	/C ratio for	r BBOA w	vith differe	nt <i>a</i> value s	sets using	ME-2 met	hod in Do	ngguan.	
<i>a</i> value <i>a</i> value for D CCOA	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

0.18

0.14

0.19

0.15

0.18

0.14

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0.18

0.9

102

0.18

0.18

0.16

0.18

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

a value for	<i>a</i> value for HOA	0	0.1	0	0.1
CCOA	\searrow	HOA vs. BC		CCOA vs. PA	AHs
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

Table S5. Comparison of the correlations between POAs and their tracers and Q/Qexp values using four reference

BBOA MS in N	AE-2 in Qingdao.
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Reference MS	HOA vs. BC	BBOAvs.C ₂ H ₄ O ₂ ⁺	COA vs. $C_6H_{10}O^+$	CCOA vs.PAHs	Q/Q _{exp}
	(R ²)	(\mathbf{R}^2)	(R ²)	(\mathbf{R}^2)	
This study	0.65	0.81	0.82	0.94	1.72
BBOA in Europe	0.60	0.75	0.65	0.80	1.93
(Crippa et al.,2014)					
BBOA in Xi'an	0.61	0.78	0.69	0.79	1.84
and Beijing					
(Elser et al.,2016)					
BBOA in Chinese	0.62	0.80	0.84	0.90	1.85
BB simulation					
(He et al.,2010)					

Table S6. Comparison of the correlations between POAs and their tracers and Q/Qexp values using four reference HOA

107 MS in ME-2 in Dongguan.

Reference MS	HOA vs. BC BBOAvs.C ₂ H ₄ O ₂ ⁺		COA vs. C ₆ H ₁₀ O ⁺	CCOA vs.PAHs	Q/Qexp
	(R ²)	(\mathbf{R}^2)	(R ²)	(\mathbf{R}^2)	
This study	0.60	0.71	0.93	0.64	2.85
HOA in Paris	0.57	0.71	0.86	0.60	3.42

(Crippa et al.,2013)					
HOA in Xi'an and	0.58	0.70	0.85	0.57	3.40
Beijing					
(Elser et al.,2016)					
HOA in Chinese	0.60	0.70	0.84	0.60	3.41
vehicle emission					
simulation					
(Zheng et al.,2016)					

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