Supporting Information

Impact of anthropogenic emissions on biogenic secondary organic aerosol: Observation in the Pearl River Delta, South China

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Text S1 SOA-tracer method for source apportionment

The SOA-tracer method is developed by Kleindienst and co-workers. Based on chamber experiments, they determine the mass fractions of tracers in SOA (f_{SOA}) and SOC (f_{SOC}) for individual precursor:

$$f_{SOA} = \frac{\sum_i [\mathrm{tr}_i]}{[SOA]}, \quad f_{SOC} = \frac{\sum_i [\mathrm{tr}_i]}{[SOC]}$$

where $\sum_i[\mathrm{tr}_i]$ is the sum of tracer concentrations for a precursor, and [SOA] and [SOC] are the measured SOA and SOC concentrations in chamber-generated SOA samples. The available f_{SOA} and f_{SOC} values were listed in Table S2. With these mass fractions in literatures and measured SOA tracers in the ambient air, SOA and SOC from different precursors have been estimated in different places of the world (Hu et al., 2008; Lewandowski et al., 2013; Stone et al., 2012; von Schneidemesser et al., 2009; Ding et al., 2014), with the assumption that the f_{SOA} and f_{SOC} values in the chamber samples are the same in the ambient air. In this study, the same set of SOA tracers reported by Kleindienst and co-workers were used for the SOC and SOA estimations (Table S2).

The uncertainty in the SOA-tracer method is induced from the analysis of organic tracers and the determination of conversion factors. The uncertainties in the tracers' analyses were estimated in the range of 15-157% (Table S2). The uncertainties in f_{SOA} were reported to be 25% for isoprene, 48% for monoterpenes, and 22% for β-caryophyllene (Kleindienst et al., 2007; Lewandowski et al., 2013). Considering these factors, the uncertainty of the estimating procedure was calculated through error propagation. The relative standard deviations (RSD) were 37% for SOA_I, 67% for SOA_M, and 158% for SOA_C. On average, the RSD of total BSOA (sum of the three BVOCs) was 59%.

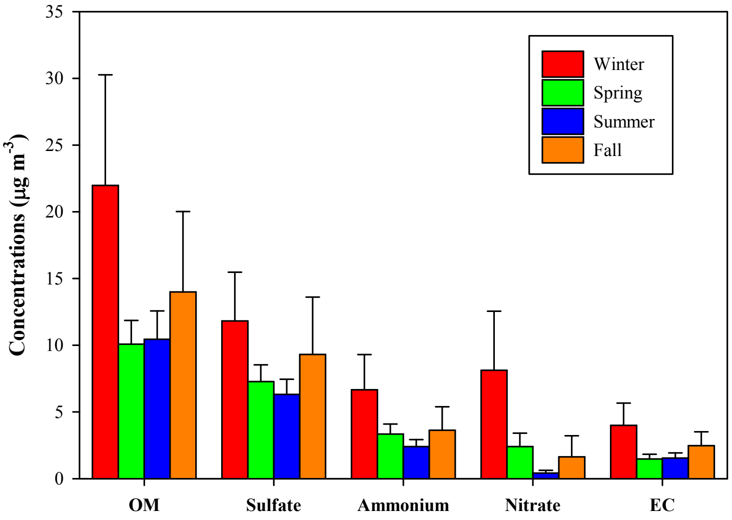

Figure S1 Seasonal variation of major components in $PM_{2.5}$. All the major components increased in winter and fall.

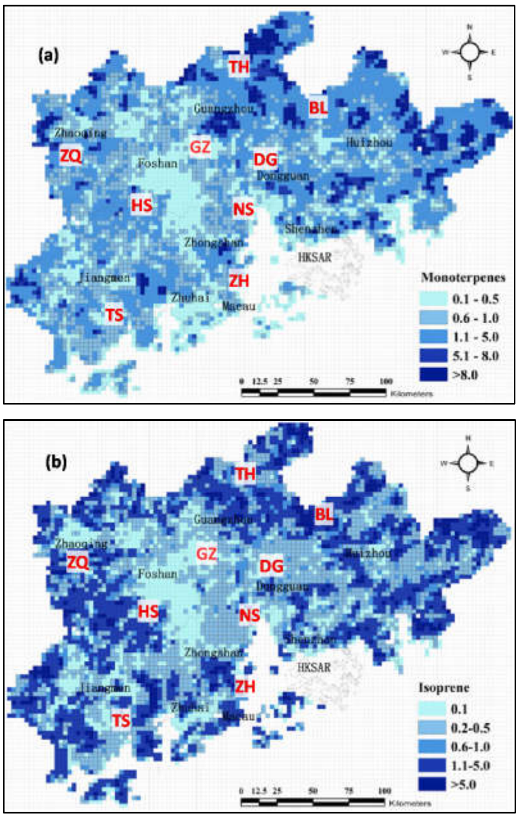

Figure S2 Spatial distribution of monoterpenes (a), and isoprene (b) emissions in the PRD (Zheng et al., 2010). The sampling sites are labeled.

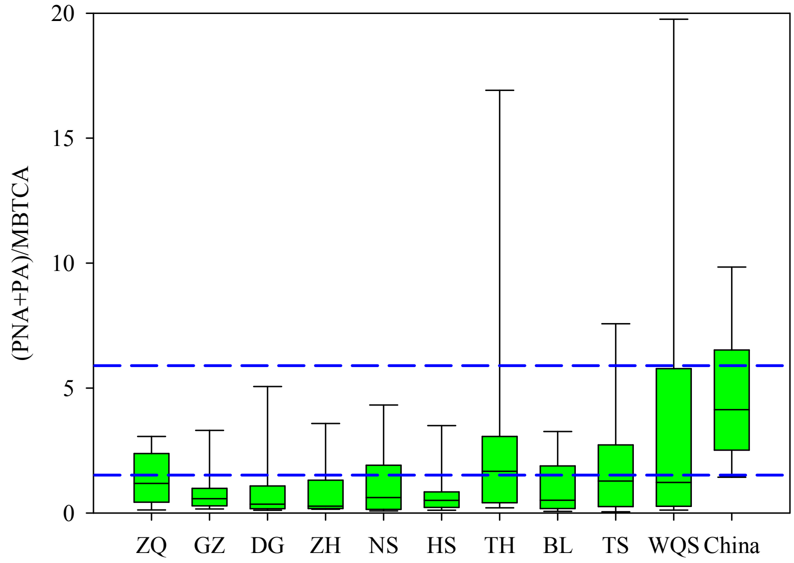

Figure S3 Spatial distribution of (PNA+PA)/MBTCA ratios at 9 sites in the PRD. The (PNA+PA)/MBTCA ratios between two blue dash lines (1.51−5.91) indicate fresh SOA_M from chamber studies (Eddingsaas et al., 2012; Offenberg et al., 2007). Box with error bars represent 10th, 25th, 75th, 90th percentiles at each site. The line in the box is the median at each site. The data at WQS site during 2008 in the PRD (Ding et al., 2012) and at 12 sites during 2012-2013 in China (Ding et al., 2016) were reported in our previous studies.

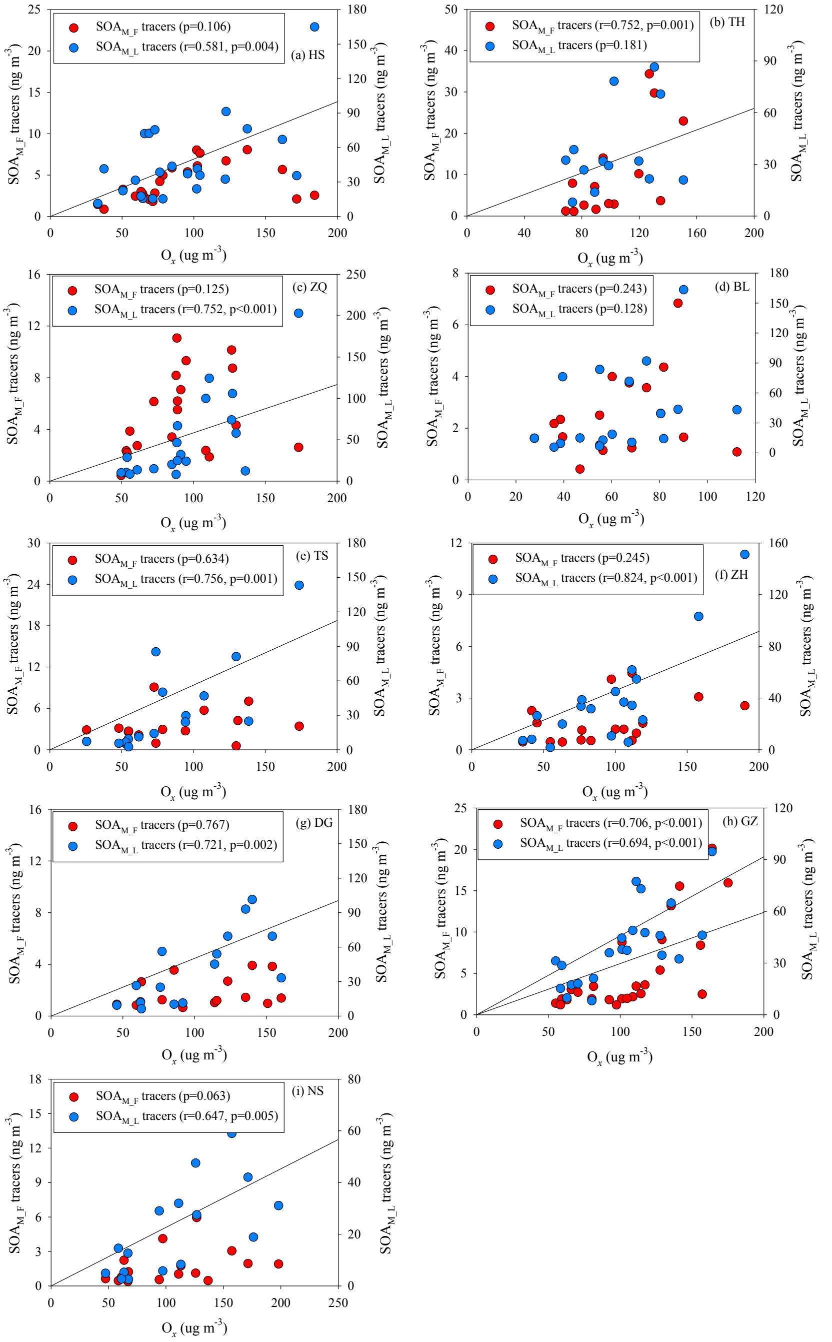

Figure S4 Correlations of SOA$_{M\_F}$ tracers and SOA$_{M\_L}$ tracers with O$_x$

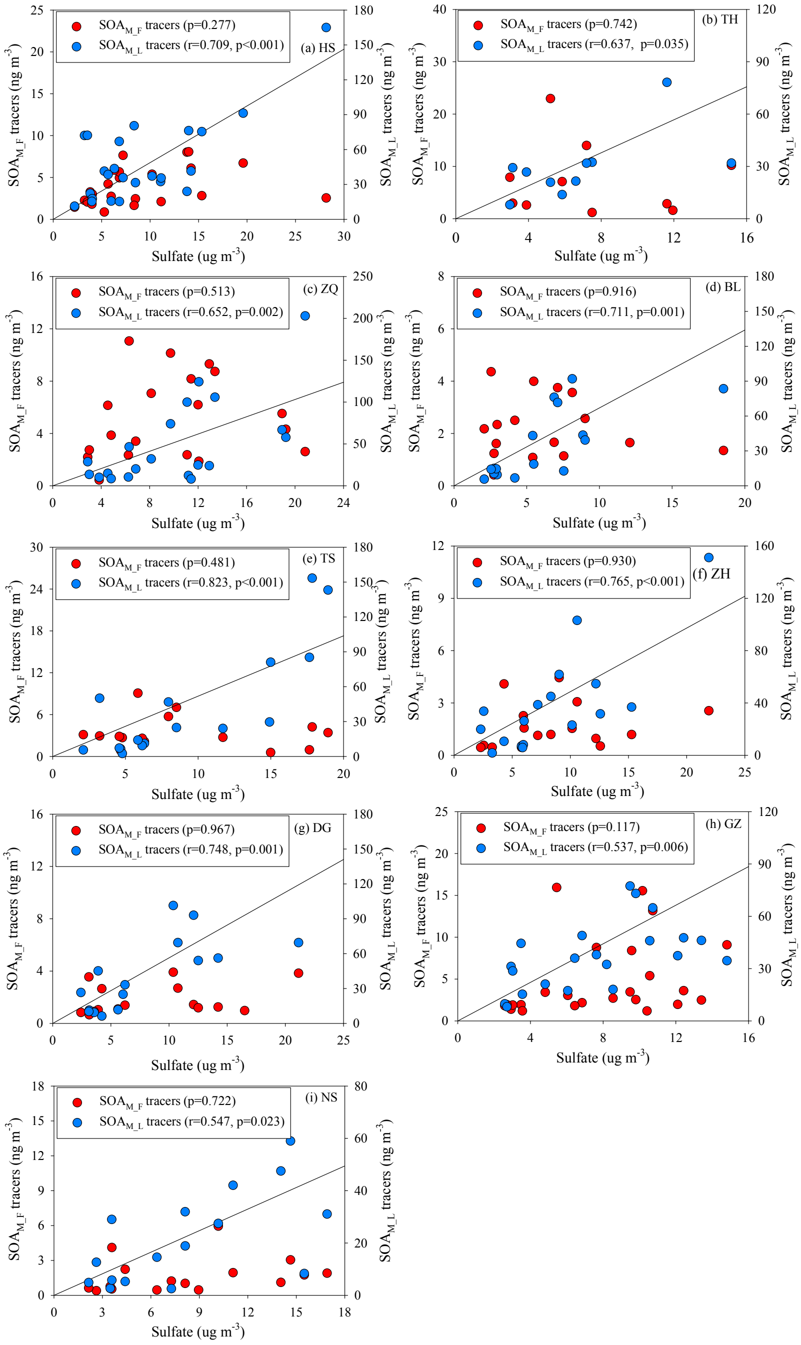

Figure S5 Correlations of SOA$_{M\_F}$ tracers and SOA$_{M\_L}$ tracers with sulfate

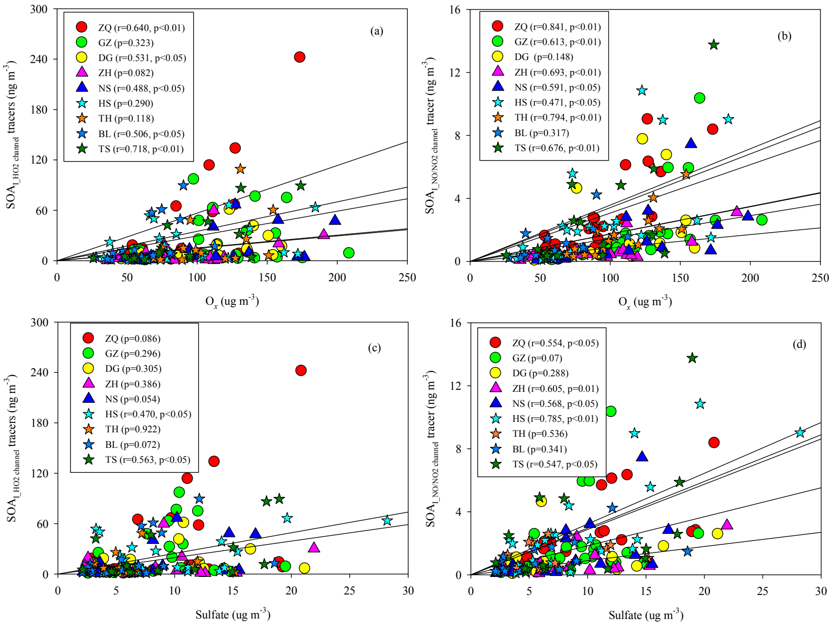

Figure S6 Correlations of SOA$_{HO2\text{-}channel}$ tracers and SOA$_{NO/NO2\text{-}channel}$ tracers with O$_x$ (a, b) and sulfate (c, d)

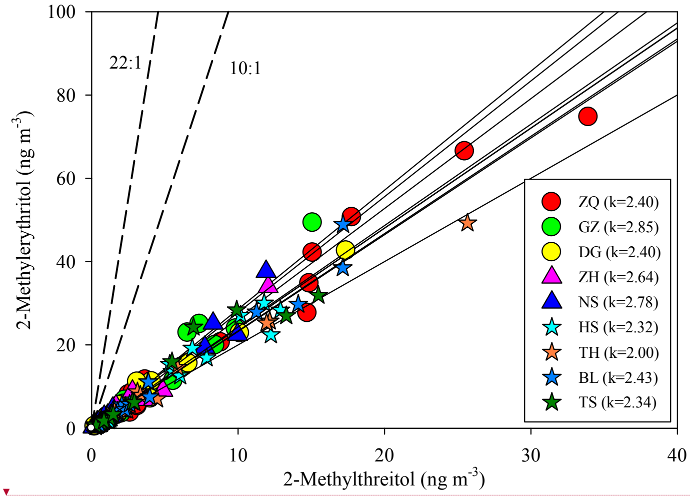

Figure S7 Significant correlations between 2-methyltetrol isomers at 9 sites in the PRD. K indicates the slope of each linear regression. The dash lines indicate the ratio range of 2-methyltetrol isomers in the SOA from isoprene ozonolysis (Riva et al., 2016).

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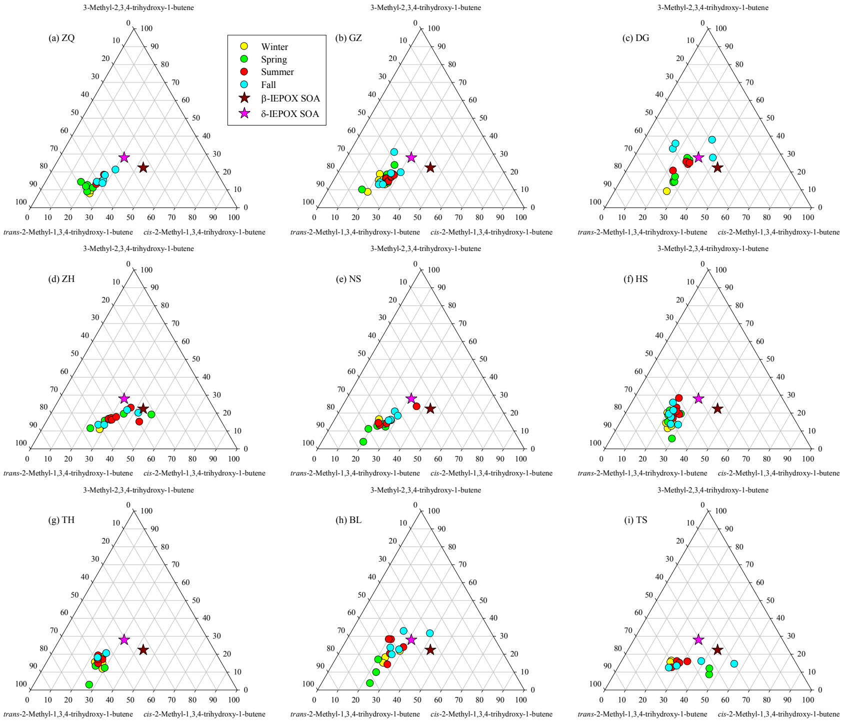

Figure S8 Intercomparison of C_5-alkene triols compositions at 9 sites and in β-IEPOX and δ-IEPOX derived SOA (Lin et al., 2012).

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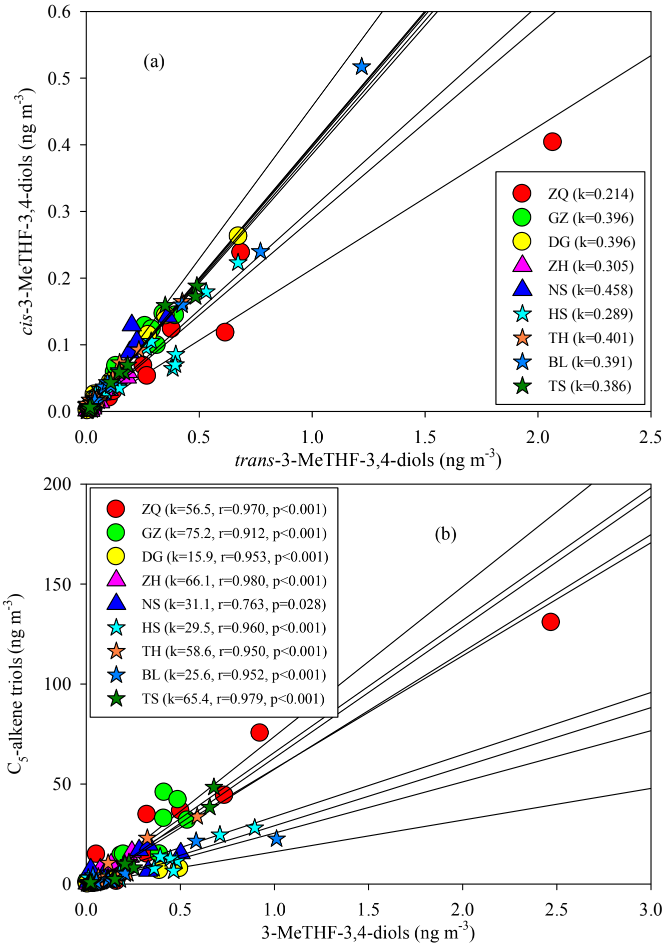

Figure S9 Significant correlations among the SOA_I tracers. K indicates the slope of each linear regression.

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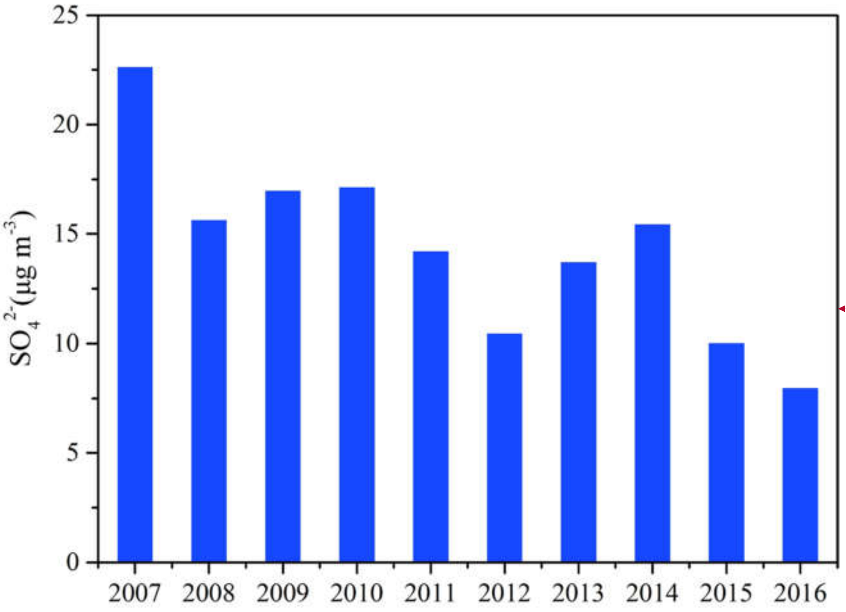

Figure S10 Long-term variation of sulfate in fall at WQS from 2007-2016.

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Table S1 Data summary of gaseous and particulate species in the air of PRD

	Zhaoqing (ZQ, urban site)					**Guangzhou (GZ, urban site)**					**Dongguan (DG, urban site)**					**Nansha (NS, sub-urban site)**					**Zhuhai (ZH, sub-urban site)**				
	Winter	Spring	Summer	Fall	Annual	Winter	Spring	Summer	Fall	Annual	Winter	Spring	Summer	Fall	Annual	Winter	Spring	Summer	Fall	Annual	Winter	Spring	Summer	Fall	Annual
Temperature (^{o}C)	15.1	22.8	29.1	24.2	22.7(12.8-31.3)	15.7	23.0	29.9	26.6	24.0(11.2-31.8)	18.1	23.0	30.3	25.3	24.9(15.9-32.0)	19.6	22.2	29.5	25.5	25.6(16.0-30.9)	16.6	22.5	29.5	24.4	24.2(14.7-31.1)
RH (%)	54	61	63	59	59(34-71)	52	59	64	55	58(26-83)	57	61	64	60	61(30-78)	72	67	68	63	67(33-82)	76	72	75	74	74(42-85)
SO_2 (μg m^{-3})	22.0	31.1	19.7	29.5	25.5(4.09-21.9)	23.4	15.7	7.38	15.4	15.1(3.43-41.3)	27.8	14.5	13.4	15.6	16.2(5.04-33.2)	22.9	12.8	9.30	20.5	14.4(4.04-36.5)	8.55	6.05	5.77	10.4	7.33(2.14-14.6)
NO_2 (μg m^{-3})	40.4	24.1	15.4	36.7	29.1(2.45-82.7)	84.8	49.3	36.3	61.2	57.2(29.7-155)	73.7	34.0	24.2	35.4	36.9(10.5-102)	61.4	27.6	19.0	44.4	31.4(8.08-91.7)	50.7	21.7	14.3	34.2	26.8(7.08-65.0)
NO (μg m^{-3})	14.7	4.65	4.06	5.88	7.31(2.00-35.4)	31.8	6.73	5.24	6.65	12.7(1.13-126)	25.1	3.64	6.77	6.14	7.88(1.5-42.4)	13.5	2.48	2.00	6.16	4.04(0.56-13.5)	6.75	na	na	na	6.75(5.63-7.53)
NO_x (μg m^{-3})	62.9	31.3	21.5	45.7	40.3(6.70-121)	134	59.5	44.3	71.4	76.8(35.2-349)	112	39.6	34.6	44.8	49.0(13.6-167)	83.0	32.1	22.7	54.4	38.3(10.3-111)	57.5	na	na	na	57.4(47.6-72.1)
O_3 (μg m^{-3})	55.2	64.3	81.9	59.6	65.2(11.8-145)	52.9	54.1	51.2	62.2	54.7(18.8-115)	42.3	65.6	80.0	66.9	66.6(31.5-123)	64.8	80.2	80.8	78.6	79.0(21.3-149)	49.7	48.7	66.3	106	67.5(18.3-155)
O_x (μg m^{-3})	95.7	88.5	97.2	96.3	94.4(49.9-173)	138	103	87.5	123	112(55.3-208)	116	99.7	104	102	103(46.0-160)	126	108	99.8	123	110(47.7-198)	100	70.4	80.6	140	94.3(35.7-190)
CO (mg m^{-3})	1.18	0.69	0.66	0.73	0.81(0.21-1.66)	1.21	1.04	0.73	0.78	0.94(0.52-1.81)	1.15	0.72	0.71	0.61	0.73(0.32-1.52)	0.85	0.74	0.66	0.69	0.70(0.37-1.21)	1.11	0.66	0.62	0.59	0.70(0.48-1.14)
OC (μgC m^{-3})	21.5	8.26	8.73	9.60	12.0(4.66-32.1)	15.9	6.55	5.90	10.2	9.59(3.12-33.5)	20.2	6.51	6.28	7.09	8.34(3.46-27.7)	17.1	5.37	5.43	9.18	7.20(1.94-19.6)	9.82	4.41	4.50	8.55	6.05(1.94-17.5)
EC (μgC m^{-3})	5.70	1.50	1.80	2.34	2.83(0.79-8.41)	4.35	1.58	1.60	2.51	2.51(0.79-11.7)	6.76	1.80	2.12	3.39	2.99(0.84-8.39)	5.32	1.22	1.48	2.71	1.99(0.44-6.61)	2.82	1.07	0.96	2.55	1.59(0.44-5.16)
SO_4^{2-} (μg m^{-3})	12. 8	8.27	8.86	10.1	10.0(2.92-20.9)	12.0	6.72	5.78	9.55	8.44(2.61-19.5)	17.7	6.87	6.52	8.09	8.52(2.45-21.2)	14.1	7.48	6.18	10.6	8.32(2.18-16.9)	14.0	6.80	5.48	12.3	8.47(2.33-21.9)
NO_3^- (μg m^{-3})	10.8	4.05	0.54	1.31	4.18(0.14-21.5)	9.11	1.82	0.57	1.29	3.22(0.15-23.3)	12.3	2.92	0.75	0.79	2.88(0.23-16.2)	14.4	1.59	0.50	1.06	1.81(0.23-14.4)	4.97	1.28	0.16	1.53	1.38(0.04-5.91)
NH_4^+ (μg m^{-3})	7.92	4.44	3.70	3.86	4.98(1.10-14.3)	7.10	2.98	2.26	3.87	4.03(0.95-12.8)	8.58	3.03	2.09	2.79	3.41(0.62-9.75)	11.5	3.54	2.33	3.91	3.69(0.93-11.5)	6.35	2.51	2.23	4.54	3.34(0.60-8.10)
Cl^- (μg m^{-3})	1.42	0.50	0.08	0.19	0.55(0.03-2.89)	0.86	0.37	0.13	0.14	0.37(0.06-1.78)	1.84	0.24	0.14	0.06	0.36(0.03-2.21)	1.33	0.44	0.09	0.20	0.30(0.04-1.44)	0.27	0.25	0.05	0.06	0.14(0.01-0.52)
Na^+ (μg m^{-3})	0.83	0.34	0.29	0.36	0.45(0.08-2.66)	0.66	0.23	0.26	0.40	0.39(0.08-1.13)	0.73	0.38	0.51	0.38	0.46(0.11-0.96)	0.60	1.46	0.27	0.43	0.68(0.15-2.30)	0.48	0.46	0.28	0.34	0.37(0.09-0.71)
K^+ (μg m^{-3})	0.83	0.38	0.23	0.33	0.44(0.11-1.32)	0.70	0.21	0.25	0.36	0.38(0.01-2.16)	1.45	0.36	0.20	0.35	0.45(0.14-1.95)	0.81	0.30	0.11	0.30	0.26(0.04-0.81)	0.46	0.22	0.09	0.24	0.21(0.02-0.58)
Mg^{2+} (μg m^{-3})	0.10	0.05	0.04	0.04	0.06(0.02-0.35)	0.08	0.04	0.03	0.04	0.05(0.01-0.18)	0.08	0.06	0.07	0.04	0.06(0.02-0.12)	0.07	0.15	0.11	0.08	0.11(0.03-0.21)	0.06	0.09	0.04	0.04	0.06(0.01-0.13)
Ca^{2+} (μg m^{-3})	0.59	0.31	0.37	0.44	0.44(0.07-1.14)	0.49	0.34	0.24	0.30	0.34(0.17-0.88)	0.42	0.32	0.32	0.30	0.33(0.11-0.79)	0.32	0.46	0.41	0.44	0.43(0.17-1.03)	0.22	0.29	0.07	0.17	0.18(0.01-0.85)
$PM_{2.5}$ (μg m^{-3})	60.7	25.4	27.3	36.9	37.5(11.7-85.6)	64.5	31.4	20.7	35.1	37.6(10.1-131)	102	39.5	26.0	32.9	41.9(14.7-125)	78.1	24.2	24.6	36.8	31.2(7.74-78.9)	51.8	27.1	17.4	43.9	30.5(9.46-83.3)
3-Hydroxyglutaric acid	21.5	18.0	35.9	20.1	23.8(3.32-89.5)	18.5	13.4	19.9	32.7	20.9(2.77-54.0)	36.5	17.6	27.6	20.2	23.2(3.73-73.6)	28.0	7.80	11.0	9.26	10.5(0.62-27.9)	16.4	6.13	15.0	33.7	16.9(0.70-61.7)
3-Hydroxy-4,4-dimethylglutaric acid	10.6	12.3	24.2	15.5	15.6(1.51-57.4)	8.09	8.39	14.8	24.4	13.7(nd-35.8)	19.3	12.8	23.9	18.1	18.0(1.17-60.5)	12.6	6.39	10.4	5.60	7.93(0.27-28.5)	14.8	6.22	12.7	30.5	15.1(0.56-53.4)
cis-Pinonic acid	5.04	3.76	2.77	4.37	3.98(0.30-10.5)	6.11	5.67	2.19	6.77	4.99(0.36-20.3)	1.65	3.26	0.39	1.66	1.84(nd-11.8)	0.57	3.48	0.97	1.54	1.85(0.06-12.8)	0.37	3.01	0.83	2.02	1.62(0.14-10.3)
Pinic acid	1.40	1.03	1.40	2.33	1.54(0.10-5.10)	0.99	0.76	0.57	1.48	0.92(0.11-3.47)	0.87	0.94	0.46	0.76	0.75(0.12-2.73)	0.51	0.77	0.52	0.71	0.65(0.05-2.69)	0.37	0.31	0.52	0.63	0.45(nd-1.82)
3-Methyl-1,2,3-butanetricarboxylic acid	6.65	4.43	17.8	8.47	9.32(0.90-55.5)	7.70	5.69	10.0	14.5	9.44(0.55-25.3)	6.67	3.05	7.96	10.3	6.99(0.17-23.5)	6.76	2.34	6.90	6.80	5.52(0.07-21.0)	2.87	1.42	5.44	15.4	6.11(0.17-35.5)
Sum of SOA_M tracers	45.1	39.4	82.0	50.8	54.3(10.0-205)	41.4	33.9	47.4	79.9	50.0(9.79-118)	65.0	37.7	60.3	51.0	50.9(8.57-156)	48.5	20.8	29.8	23.9	26.5(3.24-67.3)	34.7	17.1	34.5	82.3	40.3(1.89-153)
cis-3-Methyltetrahydrofuran-3,4-diol	0.02	0.03	0.13	0.09	0.06(nd-0.40)	0.02	0.04	0.06	0.10	0.06(nd-0.14)	0.02	0.02	0.04	0.09	0.04(nd-0.26)	nd	nd	0.07	0.11	0.09(0.02-0.14)	nd	0.01	0.02	0.03	0.01(nd-0.05)
trans-3-Methyltetrahydrofuran-3,4-diol	0.04	0.08	0.64	0.25	0.25(nd-2.06)	0.04	0.05	0.16	0.21	0.12(nd-0.39)	0.05	0.04	0.10	0.22	0.11(nd-0.67)	nd	nd	0.13	0.31	0.21(0.01-0.67)	0.01	0.02	0.06	0.08	0.04(nd-0.18)
cis-2-Methyl-1,3,4-trihydroxy-1-butene	0.59	1.63	11.3	6.26	4.93(0.11-33.2)	0.39	0.85	3.32	4.50	2.27(0.02-10.6)	0.49	0.28	0.76	0.37	0.45(0.02-1.54)	0.43	0.23	1.72	2.50	1.37(0.05-5.25)	0.12	0.26	1.36	1.85	0.95(0.04-4.24)
3-Methyl-2,3,4-trihydroxy-1-butene	0.21	0.82	5.82	3.19	2.50(0.07-17.0)	0.24	0.46	1.85	2.49	1.26(0.01-5.85)	0.20	0.21	0.68	0.70	0.46(0.01-2.52)	0.33	0.12	1.01	1.65	0.85(nd-3.19)	0.05	0.13	0.75	0.89	0.49(0.01-2.31)
trans-2-Methyl-1,3,4-trihydroxy-1-butene	1.62	4.71	25.9	13.5	11.4(0.17-80.4)	0.95	1.86	7.49	11.5	5.43(0.11-29.2)	1.21	0.58	1.59	1.06	1.06(0.01-3.90)	1.24	0.64	3.46	5.11	2.85(0.09-11.3)	0.24	0.53	2.27	3.91	1.81(0.02-9.74)
2-Methylglyceric acid	2.02	3.31	3.00	3.84	3.04(0.24-9.02)	1.32	1.04	0.79	4.02	1.70(0.09-10.3)	1.56	1.41	0.94	3.09	1.83(0.07-7.75)	1.25	0.73	1.33	2.30	1.43(0.09-7.43)	0.47	0.37	0.78	1.26	0.71(0.09-3.11)
2-Methylthreitol	1.36	7.08	16.7	6.56	7.93(0.68-33.9)	0.91	1.65	4.92	5.23	3.22(0.20-15.0)	1.45	2.08	3.16	6.34	3.60(0.22-17.3)	0.97	0.54	4.08	4.17	2.88(0.11-11.9)	0.20	0.82	3.38	2.48	1.93(0.09-12.0)
2-Methylerythritol	3.31	15.5	42.4	15.5	19.1(1.59-74.7)	1.94	3.87	15.7	11.6	8.58(0.41-49.3)	2.97	4.56	8.67	14.7	8.56(0.48-42.6)	1.77	1.23	12.5	9.83	7.77(0.27-37.6)	0.44	1.86	9.49	4.69	4.79(0.25-33.9)
Sum of SOA_I tracers	9.18	33.1	106	49.2	49.3(4.86-250)	5.80	9.77	34.2	39.6	22.6(0.89-97.9)	7.95	9.18	15.9	26.2	16.0(0.95-68.6)	6.00	3.49	24.2	24.0	17.0(0.83-69.9)	1.53	3.99	18.1	15.2	10.8(0.54-62.2)
β-Caryophyllenic acid	9.76	3.70	4.02	3.40	5.22(0.40-14.3)	11.4	4.33	4.75	7.78	7.07(nd-20.5)	9.05	2.32	2.35	2.22	3.13(0.07-10.8)	8.40	1.81	1.20	1.47	1.88(0.20-8.40)	3.69	0.67	1.03	3.05	1.82(nd-5.53)
SOA_M (μg m^{-3})	1.02	0.90	1.86	1.15	1.23(0.22-4.65)	0.94	0.77	1.08	1.81	1.13(0.22-2.69)	1.48	0.86	1.37	1.16	1.15(0.19-3.55)	1.10	0.47	0.68	0.54	0.60(0.07-1.52)	0.79	0.39	0.78	1.87	0.91(0.04-3.48)
SOA_I (μg m^{-3})	0.11	0.41	0.99	0.41	0.47(0.05-1.85)	0.07	0.10	0.34	0.33	0.21(0.01-1.04)	0.09	0.13	0.20	0.38	0.22(0.01-1.07)	0.06	0.04	0.28	0.26	0.19(0.01-0.83)	0.02	0.05	0.22	0.13	0.11(0.01-0.76)
SOA_C (μg m^{-3})	0.90	0.34	0.37	0.31	0.47(0.03-1.31)	1.05	0.40	0.44	0.71	0.64(nd-1.88)	0.83	0.21	0.22	0.20	0.28(0.01-0.99)	0.77	0.17	0.11	0.14	0.17(0.01-0.77)	0.34	0.06	0.09	0.28	0.16(nd-0.50)
BSOA (μg m^{-3})	2.03	1.65	3.22	1.88	2.19(0.45-7.40)	2.06	1.27	1.85	2.86	1.99(0.45-4.21)	2.40	1.20	1.79	1.75	1.66(0.26-4.47)	1.93	0.68	1.07	0.94	0.96(0.12-2.24)	1.14	0.50	1.09	2.28	1.20(0.08-4.25)

“na” means not available and “nd” means not detected.

Table S1 Data summary of gaseous and particulate species in the air of PRD (continued)

	Tianhu (TH, rural site)					**Boluo (BL, rural site)**					**Heshan (HS, rural site)**					**Taishan (TS, rural site)**					**9 sites average**				
	Winter	Spring	Summer	Fall	Annual	Winter	Spring	Summer	Fall	Annual	Winter	Spring	Summer	Fall	Annual	Winter	Spring	Summer	Fall	Annual	Winter	Spring	Summer	Fall	Annual
Temperature (°C)	13.2	19.9	27.0	24.4	20.5(11.0-29.4)	16.4	20.5	28.6	23.4	22.7(13.9-31.4)	13.1	21.8	29.0	23.1	21.4(10.5-31.0)	16.4	23.0	29.1	23.4	22.9(14.0-31.1)	16.0	22.1	29.1	24.5	23.2(10.5-32.0)
RH (%)	na	na	na	na	na	75	75	70	71	72(60-85)	58	64	70	63	63(39-86)	75	76	71	75	74(54-84)	58	60	61	58	59(26 -86)
SO$_2$ (μg m^{-3})	11.9	9.75	8.70	13.5	10.5(5.34-16.9)	15.1	10.1	10.6	15.6	13.0(5.13-20.3)	31.3	23.2	11.7	29.5	24.4(5.60-46.8)	11.6	4.35	5.46	7.15	7.14(0.95-17.0)	19.4	14.2	10.2	17.5	14.9(7.14-25.5)
NO$_2$ (μg m^{-3})	10.6	8.54	10.8	3.37	8.98(2.95-16.2)	14.1	17.6	11.4	10.7	12.9(3.78-21.4)	45.7	26.7	5.85	43.4	31.4(3.68-60.2)	38.7	14.3	13.9	18.5	21.3(6.47-49.4)	46.7	24.9	16.8	32.0	28.5(8.98-57.2)
NO (μg m^{-3})	0.25	0.32	1.72	0.70	0.87(0.13-2.47)	1.47	1.30	0.75	0.83	1.03(0.27-2.82)	3.49	3.77	1.40	3.67	3.15(0.68-12.4)	3.58	0.47	0.63	1.13	1.45(0.08-5.78)	11.2	2.60	2.51	3.46	5.03(0.87-12.7)
NO$_x$ (μg m^{-3})	11.8	9.82	14.2	5.08	10.8(4.68-19.8)	17.2	20.4	14.7	12.8	15.7(8.65-26.6)	51.6	33.0	17.37	48.7	38.5(9.72-72.4)	44.0	16.1	15.8	19.8	23.9(8.08-57.0)	63.8	30.0	22.4	38.5	39.0(10.8-76.8)
O$_3$ (μg m^{-3})	99.2	84.6	90.2	139	97.2(52.8-150)	29.6	38.0	69.8	55.8	50.6(18.2-97.3)	48.7	75.4	61.0	60.4	61.3(12.8-135)	32.9	65.6	77.4	87.9	65.9(10.9-147)	52.8	64.1	73.2	79.6	67.7(50.6-97.2)
O$_x$ (μg m^{-3})	110	93.2	101	143	106(69.0-154)	43.7	55.6	81.2	66.5	63.5(27.8-112)	94.4	102	66.9	104	92.8(33.3-184)	71.6	80.0	91.2	106	87.2(25.8-173)	99.5	88.9	90.0	112	96.1(63.5-112)
CO (mg m^{-3})	0.62	0.57	0.56	0.32	0.54(0.26-0.87)	0.85	0.67	0.51	0.54	0.62(0.30-1.06)	1.10	0.88	0.87	0.82	0.91(0.50-1.22)	0.88	0.56	0.54	0.78	0.68(0.30-1.04)	1.00	0.73	0.65	0.65	0.74(0.54-0.94)
OC (μgC m^{-3})	8.05	5.38	6.30	8.05	6.49(3.64-10.4)	7.40	6.63	7.03	8.67	7.52(2.64-16.7)	12.9	6.82	6.31	12.6	9.65(2.74-22.4)	10.8	6.79	8.28	12.8	9.67(4.24-23.1)	13.7	6.30	6.53	8.74	8.50(1.93-33.4)
EC (μgC m^{-3})	1.44	0.96	1.04	na	1.13(0.40-2.04)	2.98	1.99	1.92	2.08	2.22(0.54-8.22)	3.38	1.86	1.36	3.51	2.52(0.52-6.19)	3.22	1.28	1.66	3.14	2.32(0.64-6.86)	4.00	1.47	1.55	2.47	2.23(0.40-11.6)
SO$_4^{2-}$ (μg m^{-3})	9.83	6.32	5.11	na	7.18(2.99-15.1)	4.38	10.11	6.40	5.97	6.45(2.10-18.5)	10.4	6.96	5.43	13.9	9.17(2.24-28.2)	11.3	5.98	7.09	13.3	9.41(2.12-18.9)	11.8	7.28	6.32	9.31	8.44(2.10-28.2)
NO$_3^-$ (μg m^{-3})	1.00	1.27	0.09	na	0.88(0.01-2.93)	2.69	3.50	0.48	0.60	1.56(0.11-8.66)	8.33	3.01	0.30	5.32	4.23(0.15-16.3)	9.49	2.23	0.40	2.81	3.73(0.12-23.7)	8.12	2.41	0.42	1.64	2.65(0.01-23.7)
NH$_4^+$ (μg m^{-3})	3.80	2.52	1.94	na	2.80(1.11-5.88)	2.43	4.58	2.42	2.38	2.79(0.77-8.96)	5.92	3.50	2.14	6.06	4.40(0.85-13.8)	6.32	3.01	2.57	5.22	4.28(0.44-11.3)	6.66	3.34	2.41	3.63	3.74(0.44-14.2)
Cl$^-$ (μg m^{-3})	0.05	0.07	0.02	na	0.04(nd-0.15)	0.54	0.40	0.05	0.08	0.23(0.01-1.77)	1.20	0.46	0.05	0.32	0.50(0.01-1.85)	1.54	0.26	0.06	0.24	0.52(0.01-4.20)	1.00	0.33	0.07	0.14	0.33(nd-4.20)
Na$^+$ (μg m^{-3})	0.57	0.48	0.14	na	0.42(0.04-0.75)	0.10	0.26	0.43	0.25	0.27(0.03-1.00)	0.71	0.31	0.24	0.59	0.46(0.18-1.29)	0.49	0.28	0.35	0.70	0.45(0.18-0.96)	0.57	0.47	0.31	0.38	0.44(0.03-2.66)
K$^+$ (μg m^{-3})	0.43	0.24	0.12	na	0.27(0.06-0.57)	0.27	0.24	0.19	0.20	0.22(0.07-0.71)	1.02	0.35	0.32	0.81	0.62(0.11-1.50)	0.49	0.16	0.42	0.52	0.39(0.09-1.02)	0.72	0.27	0.21	0.34	0.36(0.01-2.16)
Mg^{2+} (μg m^{-3})	0.08	0.07	0.02	na	0.05(nd-0.10)	0.02	0.03	0.03	0.04	0.03(0.01-0.07)	0.04	0.03	0.02	0.04	0.03(nd-0.09)	0.02	0.03	0.03	0.04	0.02(nd-0.06)	0.06	0.06	0.04	0.04	0.05(nd-0.34)
Ca^{2+} (μg m^{-3})	0.46	0.42	0.10	na	0.35(0.04-0.85)	0.18	0.16	0.25	0.34	0.24(0.10-0.62)	0.34	0.13	0.13	0.36	0.24(0.02-0.66)	0.20	0.19	0.19	0.31	0.22(0.12-0.40)	0.36	0.29	0.23	0.29	0.30(nd-1.14)
PM$_{2.5}$ (μg m^{-3})	33.9	21.6	18.9	na	25.0(9.98-43.0)	31.4	30.5	24.5	28.7	28.4(11.0-72.5)	63.1	29.8	21.4	54.9	42.2(6.78-112)	55.3	21.1	24.5	52.6	38.3(7.68-114)	60.1	27.8	22.8	35.7	34.7(6.78-131)
3-Hydroxyglutaric acid	21.0	18.1	22.0	35.6	22.2(4.44-52.4)	14.1	16.4	24.3	16.5	18.1(3.64-74.8)	16.3	14.7	24.0	35.7	22.6(2.72-89.4)	19.0	5.25	23.4	42.0	22.4(1.44-79.2)	21.2	13.1	22.6	27.3	20.1(10.5-23.8)
3-Hydroxy-4,4-dimethylglutaric acid	10.7	15.2	15.0	35.5	16.6(1.65-36.8)	6.20	13.9	18.4	19.1	14.9(0.77-53.6)	7.87	12.5	19.6	26.7	16.6(2.11-61.0)	7.49	4.36	15.5	25.7	13.2(nd-47.9)	10.8	10.2	17.2	22.3	14.7(7.93-18.0)
cis-Pinonic acid	11.8	6.26	4.03	29.3	10.2(0.66-34.3)	1.80	1.16	1.57	2.22	1.74(0.28-5.34)	6.81	3.32	2.01	2.41	3.63(0.56-18.5)	7.55	3.91	2.01	1.82	3.82(0.08-26.1)	4.64	3.76	1.86	5.79	3.75(1.62-10.2)
Pinic acid	1.70	0.92	1.33	6.58	1.99(0.19-7.69)	0.73	0.49	0.66	0.94	0.72(0.12-1.80)	1.17	0.65	0.77	1.45	1.01(0.14-3.17)	0.63	0.95	1.02	1.44	1.00(0.13-2.43)	0.93	0.76	0.81	1.81	1.01(0.45-1.99)
3-Methyl-1,2,3-butanetricarboxylic acid	4.48	3.76	5.37	18.3	6.31(0.75-18.8)	6.89	3.68	12.1	10.4	8.91(0.35-34.5)	3.18	7.44	14.1	9.73	8.62(0.92-18.1)	6.98	2.08	6.68	13.8	7.39(0.40-25.8)	5.80	3.76	9.60	12.0	7.63(5.52-9.44)
Sum of SOA$_M$ tracers	49.7	44.3	47.7	125	57.4(15.5-134)	29.7	35.6	57.0	49.2	44.5(7.51-164)	35.3	38.6	60.5	76.0	52.5(12.3-167)	41.7	16.5	48.6	84.9	47.9(4.82-157)	43.5	31.6	52.0	69.2	47.1(26.5-57.4)
cis-3-Methyltetrahydrofuran-3,4-diol	0.01	0.02	0.04	0.13	0.03(nd-0.16)	0.02	0.02	0.07	0.14	0.07(nd-0.51)	0.02	0.01	0.05	0.10	0.04(nd-0.22)	0.02	0.06	0.05	0.12	0.06(nd-0.18)	0.02	0.03	0.06	0.10	0.05(0.01-0.09)
trans-3-Methyltetrahydrofuran-3,4-diol	0.02	0.04	0.08	0.33	0.08(nd-0.42)	0.03	0.04	0.22	0.35	0.18(nd-1.21)	0.03	0.03	0.24	0.30	0.15(nd-0.67)	0.04	0.13	0.13	0.25	0.13(0.01-0.48)	0.03	0.05	0.20	0.26	0.14(0.04-0.25)
cis-2-Methyl-1,3,4-trihydroxy-1-butene	0.28	0.31	1.32	7.20	1.48(0.05-8.95)	0.19	0.10	1.77	1.55	1.00(0.01-5.64)	0.31	0.27	1.66	2.91	1.28(0.04-7.09)	0.24	0.12	2.01	5.88	2.19(0.03-12.0)	0.34	0.45	2.80	3.67	1.77(0.45-4.93)
3-Methyl-2,3,4-trihydroxy-1-butene	0.17	0.19	0.89	5.49	1.07(nd-6.86)	0.13	0.08	1.41	1.18	0.78(nd-4.53)	0.16	0.22	1.53	2.38	1.07(nd-5.23)	0.14	0.04	1.06	2.82	1.15(nd-5.94)	0.18	0.25	1.67	2.31	1.07(0.46-2.50)
trans-2-Methyl-1,3,4-trihydroxy-1-butene	0.67	0.71	2.98	15.6	3.29(0.13-17.8)	0.44	0.30	3.70	3.17	2.11(0.04-12.3)	0.77	0.71	3.70	7.29	3.11(0.10-17.1)	0.56	0.16	4.28	13.3	5.21(0.05-30.3)	0.86	1.13	6.15	8.28	4.03(1.06-11.4)
2-Methylglyceric acid	1.43	1.03	1.23	4.80	1.68(0.31-5.54)	0.77	0.69	1.89	4.78	2.26(0.23-13.5)	1.50	1.23	2.27	5.87	2.71(0.10-10.8)	1.15	1.48	2.44	5.10	2.54(0.15-13.7)	1.28	1.25	1.63	3.90	1.99(0.71-3.04)
2-Methylthreitol	0.98	1.76	6.12	18.8	4.98(0.26-25.6)	1.12	1.60	7.80	6.91	4.87(0.34-17.1)	0.87	1.22	7.25	6.91	4.06(0.25-12.9)	1.10	1.33	5.93	7.78	4.03(0.26-15.4)	1.00	2.01	6.60	7.24	4.17(1.93-7.93)
2-Methylerythritol	1.85	3.46	14.2	37.2	10.4(0.36-49.2)	2.44	3.05	19.9	15.8	11.6(0.75-48.9)	2.07	2.62	19.4	14.1	9.54(0.70-30.0)	2.44	2.81	18.0	16.0	9.79(0.59-31.8)	2.14	4.32	17.8	15.5	10.0(4.79-19.1)
Sum of SOA$_I$ tracers	5.41	7.50	26.9	89.6	23.0(1.45-113)	5.13	5.87	36.7	32.7	22.6(1.54-93.8)	5.74	6.30	36.1	39.8	21.9(1.38-77.3)	5.68	5.95	33.9	51.2	24.1(1.19-103.)	5.82	9.46	36.9	40.8	23.0(10.8-49.3)
β-Caryophyllenic acid	9.17	3.84	2.96	5.17	5.20(1.77-11.8)	3.71	2.33	2.33	2.30	2.64(0.45-8.47)	6.73	2.38	2.99	5.17	4.31(1.45-10.3)	5.49	0.11	2.88	4.79	3.31(nd-11.4)	7.49	2.39	2.73	3.93	3.84(1.82-7.07)
SOA$_M$ (μg m^{-3})	1.13	1.01	1.08	2.84	1.30(0.35-3.05)	0.67	0.81	1.29	1.12	1.01(0.17-3.73)	0.80	0.88	1.37	1.72	1.19(0.28-3.79)	0.95	0.38	1.10	1.93	1.08(0.10-3.56)	0.99	0.72	1.18	1.57	1.07(0.60-1.30)
SOA$_I$ (μg m^{-3})	0.07	0.10	0.34	0.97	0.27(0.01-1.25)	0.07	0.08	0.47	0.44	0.29(0.02-1.11)	0.07	0.08	0.46	0.43	0.25(0.01-0.82)	0.07	0.09	0.42	0.46	0.25(0.01-0.85)	0.07	0.12	0.41	0.42	0.25(0.11-0.47)
SOA$_C$ (μg m^{-3})	0.84	0.35	0.27	0.47	0.47(0.16-1.08)	0.34	0.21	0.21	0.21	0.24(0.04-0.77)	0.62	0.22	0.27	0.47	0.39(0.13-0.94)	0.50	0.01	0.26	0.44	0.30(nd-1.05)	0.69	0.22	0.25	0.36	0.35(0.16-0.64)
BSOA (μg m^{-3})	2.04	1.46	1.70	4.28	2.05(0.53-4.33)	1.08	1.11	1.98	1.76	1.55(0.26-5.35)	1.49	1.18	2.11	2.62	1.84(0.56-5.43)	1.52	0.47	1.79	2.82	1.65(0.16-5.24)	1.74	1.06	1.84	2.35	1.68(0.96-2.19)

“na” means not available and “nd” means not detected.

Table S2 SOA tracers and f_{SOA} and f_{SOC} values for SOA estimation

	Monoterpenes [a]	Isoprene [b]	β-Caryophyllene [b]
SOA Tracers [c]	PNA (15%) [d] PA (34%) [d] MBTCA (62%) [d] HGA (96%) [d] HDMGA (67%) [d]	2-MTLs (41%) [d] 2-MGA (43%) [d] 3-MeTHF-3,4-diols (52%) C_5-alken triols (93%)	CA (157%) [d]
f_{SOA} (μg μg^{-1})	0.044 (48%) [e]	0.063 (25%) [e]	0.0109 (22%) [e]
f_{SOC} (μg μgC^{-1})	0.059	0.155	0.023

[a] The f_{SOA} and f_{SOC} values for monoterpenes are calculated based on the data reported by Offenberg et al. (2007). [b] The f_{SOA} and f_{SOC} values for isoprene, and β-caryophyllene are reported by Kleindienst et al. (2007). [c] The numbers in brackets are uncertainties in tracer measurement. [d] These tracers are used to calculate f_{SOA} and estimate ambient SOA. [e] The numbers in brackets are the uncertainties of f_{SOA} values reported by Kleindienst et al. (2007).

Table S3 Correlation analysis of HO_2-channel SOA_I tracers with O_3

	Coefficient (r)	*p*-value
3-MeTHF-3,4-diols	0.343	<0.001
C_5-alkene triols	0.388	<0.001
2-Methyltetrols	0.386	<0.001
HO_2-chanle SOA_I tracers	0.409	<0.001

Table S4 Correlations among HO_2-channel SOA_I tracers

	3-MeTHF-3,4-diols	C_5-alkene triols	2-Methyltetrols
3-MeTHF-3,4-diols	1	0.789	0.792
C_5-alkene triols		1	0.787
2-Methyltetrols			1

All the correlations are significant (p<0.001)

Table S5 Rate constants and lifetimes of SOA precursors

	α-Pinene	**β-Pinene**	**Isoprene**	**β-Caryophyllene**
	Rate constants at 298 K (cm^3 molecules^{-1} s^{-1}) [a]			
OH	5.25×10^{-11}	7.88×10^{-11}	9.99×10^{-11}	1.97×10^{-10}
O_3	9.01×10^{-17}	1.50×10^{-17}	1.28×10^{-17}	1.16×10^{-14}
	Lifetimes (hrs) [b]			
OH	0.53	0.35	0.28	0.14
O_3	3.64	21.9	25.7	0.03

[a] Rate constants are provided by MCMv3.2 (http://mcm.leeds.ac.uk/MCMv3.2).
[b] Lifetimes are estimated using summer average concentration of OH radical (~1×10^7 molecules cm^{-3}) in the PRD (Hofzumahaus et al., 2009), and annual average O_3 concentration (67.7 μg m^{-3}) in Table S1.

Table S6 Concentrations of isoprene SOA products at HS and TS sites

	HS 20150701	TS 20150701
2-Methyltetrol sulfates (ng m^{-3})	6.65	2.99
C_5-alkene triols (ng m^{-3})	11.5	10.8
2-Methyltetrols (ng m^{-3})	41.8	31.2
3-MeTHF-3,4-diols (ng m^{-3})	0.482	0.227

Table S7 Correlations of BSOA with sulfate and O_x during fall-winter in 2008 at WQS

	Sulfate (2008-WQS)			O_x (2008-WQS)	
	Slope	*p*-value	% [a]	Slope	*p*-value
SOA_M	0.023	0.005	50	-	0.551
SOA_I	0.032	<0.001	76	-	0.509
SOA_C	0.032	<0.001	87	-	0.139
BSOA	0.087	<0.001	69	-	0.563

[a] Percentages of SOA reduction at 50% decline of sulfate or O_x

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