Table S1. Summary of the location, sampling information of the 86 surface online OA measurements in China as well as the campaign-average mass concentrations of the observed OA and the PMF-derived POA and SOA.

Site #	Site Name / Region	References	Site Type	Sampling Period	Lon.	Lat.	OA ($\mu g\ m^{-3}$)	POA ($\mu g\ m^{-3}$)	SOA ($\mu g\ m^{-3}$)
1	IAP / NCP	J. K. Zhang et al. (2014)	urban	1/1/2013-2/1/2013	116.37	39.97	44.65	20.54	24.11
2	PKU / NCP	Hu et al. (2017)	urban	1/23/2013-3/2/2013	116.31	39.99	29.7	14.9	14.8
3	IAP / NCP	Y. Sun et al. (2016a)	urban	12/17/2013-1/17/2014	116.37	39.97	38.1	21.5	16.5
4	IAP / NCP	J. K. Zhang et al. (2015)	urban	1/1/2014-2/3/2014	116.37	39.97	27.27	8.45	18.82
5	IRSDE / NCP	Elser et al. (2016)	urban	1/9/2014-1/26/2014	116.38	40.00	43	35.67	7.4
6	IAP / NCP	Xu et al. (2019a)	urban	11/17/2014-12/13/2014	116.37	39.97	30.4	16.1	14.7
7	IAP / NCP	Zhou et al. (2018)	urban	12/1/2014-1/18/2015	116.37	39.97	31.8	18.4	14.3
8	THU / NCP	H. Li et al. (2019)	urban	12/6/2014-2/27/2015	116.30	40.00	30.4	17.9	12.6
9	IAP / NCP	J. K. Zhang et al. (2016a)	urban	12/10/2014-12/31/2014	116.37	39.97	20.25	12.14	8.1
10	NCNT / NCP	Huang et al. (2020)	urban	12/29/2014-2/28/2015	116.38	40.00	38.38	26.86	11.9
11	IAP / NCP	Y. Zhang et al. (2016)	urban	2/2/2015-4/1/2015	116.37	39.97	39	24.75	14.25
12	NCN / NCP	Duan et al. (2020)	urban	12/4/2015-2/6/2016	116.32	39.99	30.97	18.8	12.2
13	IAP / NCP	Xu et al. (2019a)	urban	11/17/2016-12/13/2016	116.37	39.97	36.4	18.3	19
14	IAP / NCP	Zhao et al. (2019)	urban	12/17/2016-12/30/2016	116.37	39.97	53.7	22.6	30.9
15	THU / NCP	H. Li et al. (2019)	urban	12/11/2017-2/2/2018	116.30	40.00	11.9	6.1	5.8
16	IAP / NCP	J. Li et al. (2020)	urban	11/10/2018-1/31/2019	116.37	39.97	11.5	6.7	4.8
17	IGDB / NCP	Huang et al. (2019)	urban	1/11/2014-2/18/2014	114.54	38.03	89	64.97	24.03
18	Handan / NCP	H. Li et al. (2017)	urban	12/4/2015-2/5/2016	114.50	36.57	82.50	50.61	31.87
19	PKU / NCP	Hu et al. (2017)	urban	3/30/2012-5/7/2012	116.31	39.99	14	7.2	6.8
20	PKU / NCP	Hu et al. (2016b)	urban	8/3/2011-9/15/2011	116.31	39.99	26.4	8.9	17.1
21	PKU / NCP	Hu et al. (2017)	urban	7/29/2012-8/29/2012	116.31	39.99	12.5	3.9	8.6
22	IAP / NCP	J. Zhang et al. (2015)	urban	8/1/2012-8/31/2012	116.37	39.97	13	5.9	7.2
23	IAP / NCP	Xu et al. (2017)	urban	6/3/2014-7/11/2014	116.37	39.97	18.1	7.78	10.32
24	THU / NCP	H. Li et al. (2018)	urban	6/30/2015-7/27/2015	116.30	40.00	12.2	4.15	8.05
25	NCN / NCP	Duan et al. (2020)	urban	7/1/2015-8/19/2015	116.32	39.99	19.27	4.9	14
26	NCN / NCP	Duan et al. (2019)	urban	8/15/2015-9/10/2015	116.32	39.99	13.8	3.9	9.8
27	IAP / NCP	Xu et al. (2019b)	urban	6/4/2017-7/13/2017	116.37	39.97	9.8	3.4	6.4
28	IAP / NCP	Xu et al. (2019b)	urban	5/20/2018-6/23/2018	116.37	39.97	12.7	3.7	9.2

29	Xinxiang / NCP	H. Li et al. (2018)	urban	6/8/2017-6/25/2017	113.90	35.30	18.00	4.32	13.5
30	IAP / NCP	J. Zhang et al. (2015)	urban	10/1/2012-10/31/2012	116.37	39.97	27	10.8	16.5
31	PKU / NCP	Hu et al. (2017)	urban	10/13/2012-11/13/2012	116.31	39.99	18.2	9.7	8.6
32	IAP / NCP	J. K. Zhang et al. (2016a)	urban	10/1/2014-10/27/2014	116.37	39.97	34.57	15.56	19.01
33	IAP / NCP	W. Xu et al. (2015)	urban	10/10/2014-11/2/2014	116.37	39.97	29.4	15	14.9
34	IAP / NCP	J. K. Zhang et al. (2016b)	urban	10/17/2014-11/2/2014	116.37	39.97	44.44	17.33	27.11
35	IAP / NCP	Xu et al. (2017)	urban	10/14/2014-11/12/2014	116.37	39.97	29.58	14.84	14.74
36	IAP / NCP	Zhao et al. (2017)	urban	9/4/2015-9/30/2015	116.37	39.97	18.31	6.33	11.97
37	NCN / NCP	Duan et al. (2019)	urban	9/11/2015-10/10/2015	116.32	39.99	21.2	8.4	13
38	NCN / NCP	Duan et al. (2019)	urban	10/11/2015-12/04/2015	116.32	39.99	29.6	15.7	13.9
39	IAP / NCP	Zhou et al. (2018)	urban	11/13/2014-11/30/2014	116.37	39.97	38.1	23.8	13.1
40	Qingdao / NCP	Zhu et al. (2018)	urban	11/1/2013-11/30/2013	120.47	36.10	10.43	5.57	4.86
41	Xinglong / NCP	J. Li et al. (2020)	polluted regional	11/10/2018-1/31/2019	117.67	40.40	5.4	2.2	3.2
42	Gucheng / NCP	Kuang et al. (2020)	polluted regional	11/11/2018-12/24/2018	115.73	39.15	66.08	42.29	21.81
43	Changdao / NCP	Hu et al. (2013)	polluted regional	3/21/2011-4/24/2011	120.70	37.99	13.4	4.4	9.4
44	Xingtai / NCP	Y. Zhang et al. (2018)	suburban	4/30/2016-6/20/2016	114.37	37.18	11.59	2.54	9.04
45	Rizhao / NCP	Lei et al. (2020)	polluted regional	3/2/2019-3/29/2019	119.40	35.18	15.66	2.19	13.31
46	Xianghe / NCP	Y. Sun et al. (2016b)	polluted regional	6/1/2013-6/30/2013	116.96	39.80	28.3	8.2	19
47	BIPT / NCP	Chen et al. (2020)	suburban	8/16/2018-9/16/2018	116.33	39.73	12.24	3.79	8.44
48	Rizhao / NCP	Lei et al. (2020)	polluted regional	9/2/2018-9/29/2018	119.40	35.18	8.32	0.67	7.65
49	Nanjing / YRD	Tang et al. (2014)	urban	1/4/2013-1/31/2013	118.73	32.21	26.26	14.11	11.89
50	JEMC / YRD	Y. Zhang et al. (2015)	urban	12/1/2013-12/31/2013	118.77	32.05	38.4	15.74	22.66
51	NOC / YRD	J. Wang et al. (2016)	urban	4/13/2015-4/29/2015	118.73	32.01	12.69	5.64	7.04
52	Nanjing / YRD	Y. J. Zhang et al. (2015)	urban	6/1/2013-6/15/2013	118.76	32.08	15.4	3.3	11.2
53	JEMC / YRD	Y. J. Zhang et al. (2014)	urban	8/1/2013-8/31/2013	118.77	32.05	10.3	2.73	6.86
54	BJ / YRD	K. Li et al. (2018)	urban	8/5/2016-8/21/2016	120.21	30.21	17	6.7	10.7
55	Nanjing / YRD	Y. J. Zhang et al. (2015)	urban	10/15/2013-10/30/2013	118.76	32.08	20.3	7.2	13.1
56	JEMC / YRD	Y. Zhang et al. (2017)	urban	10/20/2015-11/19/2015	118.75	32.04	25.2	7.06	18.14
57	BJ / YRD	K. Li et al. (2018)	urban	9/7/2016-9/23/2016	120.21	30.21	18.5	6.1	12.1
58	Lin'an / YRD	Y. W. Zhang et al. (2015)	polluted regional	11/16/2013-12/18/2013	119.73	30.30	29	13.4	15.7

59	Dongguan / PRD	Zhu et al. (2018)	urban	12/12/2013-1/1/2014	113.75	23.03	23.3	7.36	15.75
60	PKUshenzhen / PRD	Cao et al. (2018)	urban	12/31/2015-1/23/2015	113.90	22.60	18.45	7.93	10.51
61	GIG / PRD	Guo et al. (2020)	urban	11/20/2017-1/5/2018	113.37	23.15	17.3	5.36	12.11
62	Mong Kok / PRD	Lee et al. (2015)	urban	3/7/2013-5/15/2013	114.17	22.32	12.8	7.9	4.9
63	Mong Kok / PRD	Lee et al. (2015)	urban	5/16/2013-7/19/2013	114.17	22.32	7.9	5.6	2.2
64	HKEPD / PRD	C. Sun et al. (2016)	urban	9/3/2013-12/31/2013	114.17	22.32	15.1	6.3	8.8
65	GPACS / PRD	Qin et al. (2017)	suburban	11/07/2014-1/3/2015	113.35	23.00	20.36	7.81	12.55
66	HKUST / PRD	Y. J. Li et al. (2015)	clean regional	1/19/2012-3/1/2012	114.26	22.34	5.1	0.98	4.12
67	HKUST / PRD	Y. J. Li et al. (2015)	clean regional	4/25/2011-6/1/2011	114.26	22.34	4	0.8	3.2
68	HKUST / PRD	Y. J. Li et al. (2015)	clean regional	9/1/2011-9/29/2011	114.26	22.34	4.1	0.75	3.35
69	HKUST / PRD	Y. J. Li et al. (2015)	clean regional	10/28/2011-12/15/2011	114.26	22.34	6	0.82	5.18
70	Xi'an / NW	Elser et al. (2016)	urban	12/13/2013-1/6/2014	108.88	34.23	128.5	100.32	21
71	LZU / NW	Xu et al. (2016)	urban	1/10/2014-2/4/2014	103.85	36.05	29.33	18.41	10.91
72	Baoji / NW	Y. C. Wang et al. (2017)	urban	2/26/2013-3/27/2014	107.14	34.35	29.7	16.63	13.07
73	CAREERI / NW	Xu et al. (2014)	urban	7/11/2012-8/7/2012	103.86	36.05	11.51	4.6	6.79
74	GMA / NW	X. Zhang et al. (2017)	urban	10/27/2014-12/3/2014	103.88	36.04	18.2	10.87	7.33
75	Sichuan Basin / OTR	Hu et al. (2016a)	suburban	12/3/2012-1/5/2013	104.64	30.15	21.5	6.2	15.4
76	Nan'ao island / OTR	Cao et al. (2019)	polluted regional	12/22/2015-1/16/2016	117.02	23.42	7.06	2.61	4.45
77	Xiamen / OTR	Cao et al. (2017)	suburban	5/1/2015-5/18/2015	118.05	24.60	13.07	3.61	9.46
78	Xinzhou / OTR	Q. Wang et al. (2016)	suburban	7/17/2014-9/5/2014	112.12	38.07	11.72	2.2	9.5
79	Mt. Yulong / OTR	Zheng et al. (2017)	remote	3/22/2015-4/14/2015	100.20	27.20	3.88	0.5	3.37
80	Lake Hongze / OTR	Zhu et al. (2016)	remote	3/19/2011-4/24/2011	118.33	33.23	9.8	3	6.8
81	Mt. Wuzhi / OTR	Zhu et al. (2016)	remote	3/18/2015-4/15/2015	109.49	18.84	4.9	0.01	4.89
82	QOMS / OTR	X. Zhang et al. (2018)	remote	4/12/2016-5/12/2016	86.95	28.36	2.39	1.04	1.34
83	WLG / OTR	Zhang et al. (2019)	remote	7/1/2017-7/31/2017	100.90	36.28	3.14	0.79	2.35
84	Nam Co / OTR	J. Wang et al. (2017a)	remote	5/30/2015-6/30/2015	90.98	30.77	0.71	0.13	0.59
85	Nam Co / OTR	Xu et al. (2018)	remote	5/31/2015-7/1/2015	90.95	30.77	1.36	0	1.36
86	NBS / OTR	Du et al. (2015)	remote	9/5/2013-10/15/2013	101.26	37.61	4.9	0.8	4.1

Table S2. Summary of the location, sampling information of the 49 surface online measurements of VOCs as well as the campaign-average mixing ratios of benzene, toluene, and xylene in China.

Site #	Site Name	References	Sampling Period	Lon.	Lat.	Benzene (ppbv)	Toluene (ppbv)	Xylene (ppbv)
1	PKU	Wang et al. (2014)	12/29/2011-1/18/2012	116.31	40.00	2.34	2.67	
2	PKU	J. Li et al. (2019a)	1/1/2015-1/31/2015	116.33	39.99	1.30	1.20	
3	NCNST	K. Li et al. (2019)	1/31-2/15/2015	116.32	39.99	1.93	1.51	
4	RCEES	Liu et al. (2017)	12/15/2015-1/14/2016	116.30	40.00	1.81	1.67	1.12
5	PKU	Y. Shi et al. (2020)	12/01/2016-1/31/2017	116.33	39.99	3.27	3.63	1.50
6	PKU	Y. Shi et al. (2020)	12/2017	116.33	39.99	1.06	1.47	0.61
7	PKU	Y. Shi et al. (2020)	1/2018	116.33	39.99	1.03	1.26	0.51
8	TEPB	Liu et al. (2016)	12/2014-2/2015	117.15	39.10	0.54	0.27	0.38
9	UCAS	K. Li et al. (2019)	11/24-12/24/2014	116.67	40.41	0.91	0.73	
10	Wangdu	Zhang et al. (2020)	11/1/2017-1/21/2018	115.25	38.67	3.16	2.48	1.07
11	NUIST	An et al. (2017)	12/1/2013-2/28/2014	118.72	32.21	3.21	3.20	0.74
12	Ziyang	Li et al. (2014)	12/6/2012-1/4/2013	104.64	30.15	1.80	0.80	
13	BJ	L. Li et al. (2015)	5/2014	116.33	39.99	0.82	1.33	
14	PKU	J. Li et al. (2019a)	4/1/2015-4/30/2015	116.33	39.99	0.76	1.06	
15	TEPB	Liu et al. (2016)	3/2015-5/2015	117.15	39.10	0.24	0.30	0.21
16	SAES	Y. Liu et al. (2019c)	5/20-5/30/2017	121.72	31.28	0.42	1.31	1.52
17	WHU	Hui et al. (2020)	4/26-6/6/2017	114.36	30.54	0.49	0.71	0.43
18	PKUSZ	Yu et al. (2019)	4/1/2016-4/30/2016	113.90	22.60	0.60	3.12	1.07
19	PKU	M. Wang et al. (2015)	8/3/2011-9/13/2011	116.31	40.00	1.29	2.08	1.74
20	PKU	M. Wang et al. (2015)	8/1/2012-8/31/2012	116.31	40.00	0.98	2.25	1.75
21	PKU	M. Wang et al. (2015)	8/7/2013-8/25/2013	116.31	40.00	0.98	2.00	1.50
22	PKU	J. Li et al. (2019a)	7/1/2015-7/31/2015	116.33	39.99	0.80	1.55	
23	PKU	Li et al. (2016)	8/11/2015-9/3/2015	116.33	39.99	0.58	1.20	0.68
24	TEPB	Liu et al. (2016)	6/2015-8/2015	117.15	39.10	8.26	1.54	0.89
25	Gucheng	L. Li et al. (2015)	6/5/2013-7/16/2013	115.67	39.13	1.08	1.31	
26	Quzhou	L. Li et al. (2015)	6/11/2014-7/9/2014	115.02	36.86	0.81	0.48	
27	NUIST	An et al. (2017)	6/1-8/31/2013	118.72	32.21	1.86	1.47	0.74
28	Zhaohui	K. Li et al. (2017)	7/1/2013-8/15/2013	120.16	30.29	0.39	1.28	1.51
29	Nanling	Gong et al. (2018)	7/15/2016-8/17/2016	112.90	24.69	0.05	0.15	0.04
30	PKU	Wu et al. (2016)	10/1-10/16/2014	116.33	39.99	1.21	1.78	1.65
31	PKU	J. Li et al. (2015)	10/18-11/22/2014	116.33	39.99	1.62	2.43	1.24

32	PKU	J. Li et al. (2019a)	10/1/2015-10/31/2015	116.33	39.99	1.01	1.66	
33	TEPB	Liu et al. (2016)	11/2014; 9/2015-10/2015	117.15	39.10	2.12	1.18	1.06
34	Shenyang	Z. Ma et al. (2019)	8/20-9/16/2017	123.42	41.78	1.41	1.63	0.88
35	Pixian	Deng et al. (2019)	8/28/2016-10/7/2016	103.87	30.80	0.79	1.78	1.17
36	CJZ	J. Li et al. (2018)	8/24/2015-9/22/2015	106.50	29.62	1.10	1.20	0.70
37	JYS	J. Li et al. (2018)	8/24/2015-9/22/2015	106.38	29.83	0.60	0.40	
38	NQ	J. Li et al. (2018)	8/24/2015-9/22/2015	106.59	29.43	1.00	0.80	0.90
39	Heshan	M. Song et al. (2019)	10/22-11/22/2014	112.93	22.73	1.04	2.96	2.39
40	RCEES	Q. Li et al. (2020)	3/1/2016-1/31/2017	116.50	39.80	1.00	1.20	1.00
41	BNU	Liu et al. (2020)	1,4,7,10/2016	116.37	39.97	0.92	0.98	0.80
42	Langfang	C. Song et al. (2019)	4/2016-3/2017	116.76	39.57	0.86	0.84	0.62
43	NUIST	An et al. (2014)	3/2011-2/2012	118.72	32.21	3.14	2.10	1.37
44	WH	Lyu et al. (2016)	2/2013-10/2014	114.37	30.54	1.70	2.00	0.60
45	WHU	Yang et al. (2019)	9/2016-8/2017	114.62	30.88	0.73	0.95	0.37
46	Qingyang	Song et al. (2018)	10/27/2016-9/30/2017	104.08	30.93	0.95	1.80	1.71
47	LEMS	Jia et al. (2016)	1/2013-12/2013	103.92	36.05	1.94	1.01	1.23
48	GPACS	Zou et al. (2015)	6/2011-5/2012	113.35	23.00	0.62	4.59	2.07
49	Mong Kok	Z. Li et al. (2020)	1/2013-12/2014	114.17	22.32	0.44	1.74	0.63

Table S3. Summary of the locations, sampling information, and campaign-average mixing ratios of the 28 surface HONO measurements as well as the corresponding simulation results in the Sp_base and Sp_R1+2 simulations.

Site #	Site Name	References	Sampling Period	Lon.	Lat.	Observed HONO (ppbv)	Sp_base simulation		Sp_R1+2 simulation	
							HONO (ppbv)	Sim/ Obs	HONO (ppbv)	Sim/ Obs
Winter										
1	EESCAS	J. Wang et al. (2017b)	1/3-1/27/2016	116.34	40.01	1.03	0.71	0.68	1.04	1.01
2	Jinan	D. Li et al. (2018)	12/1/2015-2/29/2016	117.05	36.67	1.14	0.95	0.84	1.64	1.44
3	Jinan	L. Wang et al. (2015)	11/26/2013-1/5/2014	117.05	36.67	0.36	1.12	3.15	1.93	5.42
4	YelRD	Gu et al. (2020)	2/8-3/24/2017	118.98	37.76	0.26	0.41	1.59	0.60	2.34
5	NUIST	Zheng et al. (2020)	12/1-12/31/2015	118.71	32.21	1.28	0.85	0.66	1.62	1.27
6	SORPES	Y. Liu et al. (2019b)	12/2017-2/2018	118.95	32.12	0.67	0.80	1.19	1.43	2.12
7	Tung Chung	Z. Xu et al. (2015)	2/3-3/9/2012	113.93	22.30	0.90	0.06	0.07	0.21	0.23
8	Zhengzhou	Hao et al. (2020)	1/9-1/31/2019	113.52	34.80	2.51	1.56	0.62	2.20	0.88
Spring										
9	EESCAS	J. Wang et al. (2017b)	4/1-5/14/2016	116.34	40.01	1.05	0.15	0.14	0.40	0.38
10	Jinan	D. Li et al. (2018)	3/1-5/31/2016	117.05	36.67	1.11	0.45	0.41	1.05	0.95
11	Changzhou	X. Shi et al. (2020)	4/3-4/24/2017	119.90	31.80	1.52	0.32	0.21	0.78	0.51
12	SORPES	Y. Liu et al. (2019b)	3-5/2018	118.95	32.12	0.45	0.43	0.94	1.10	2.42
13	SORPES	Nie et al. (2015)	4/17-6/24/2012	118.95	32.12	0.75	0.30	0.40	0.90	1.19
14	Tung Chung	Z. Xu et al. (2015)	5/1-5/31/2012	113.93	22.30	0.40	0.15	0.38	0.56	1.40
Summer										
15	Wangdu	Y. Liu et al. (2019a)	6/8-7/5/2014	115.18	38.68	0.92	0.22	0.24	0.66	0.72
16	EESCAS	J. Wang et al. (2017b)	6/20-7/25/2016	116.34	40.01	1.39	0.15	0.11	0.50	0.36
17	Jinan	D. Li et al. (2018)	6/1-8/31/2016	117.05	36.67	0.76	0.42	0.55	1.09	1.42
18	YelRD	Gu et al. (2020)	6/1-7/10/2017	118.98	37.76	0.17	0.06	0.36	0.23	1.36
19	SORPES	Y. Liu et al. (2019b)	11/2017; 9-11/2018	118.95	32.12	1.04	0.45	0.44	1.04	1.00
20	SORPES	Y. Liu et al. (2019b)	6-8/2018	118.95	32.12	0.66	0.31	0.46	0.86	1.30
21	Tung Chung	Z. Xu et al. (2015)	8/3-9/7/2011	113.93	22.30	0.68	0.10	0.14	0.43	0.64
22	IEECAS	Huang et al. (2017)	7/24-8/6/2015	108.87	34.22	1.11	0.06	0.06	0.31	0.28
Autumn										
23	EESCAS	J. Wang et al. (2017b)	9/22-10/21/2016	116.34	40.01	2.27	0.31	0.13	0.62	0.27
24	ICCAS	Tong et al. (2015)	10/28-11/2/2014	116.32	39.99	1.45	0.37	0.26	0.64	0.44
25	UCAS	Tong et al. (2015)	10/28-11/2/2014	116.60	40.40	0.75	0.17	0.23	0.27	0.36
26	ICCAS	Jia et al. (2020)	8/23-9/15/2018	116.32	39.99	1.24	0.21	0.17	0.66	0.53
27	Jinan	D. Li et al. (2018)	9/1-11/30/2015	117.05	36.67	1.75	0.80	0.46	1.51	0.86
28	Tung Chung	Z. Xu et al. (2015)	11/1-12/3/2011	113.93	22.30	0.92	0.08	0.09	0.29	0.32

Table S4. Summary of the locations, sampling information, and the maxima in the diurnal profiles of the hourly mean concentrations from 10 surface OH and HO_2 measurements in China as well as the corresponding model results in the Sp_base and Sp_R1+2 simulations.

Site #	Site Name/ Region	References	Sampling Period	Lon.	Lat.	OH (10^6 cm^{-3})			HO_2 (10^8 cm^{-3})		
						Obs	Sp_base	Sp_R1+2	Obs	Sp_base	Sp_R1+2
Winter											
1	Huairou/ NCP	Tan et al. (2018)	1/6-2/2/2016; 2/20-3/5/2016	116.68	40.41	3.1	1.1	2.8	0.8	0.2	0.5
2	IAP/ NCP	Slater et al. (2020)	11/16-11/20/2016; 12/2-12/8/2016	116.37	39.97	2.7	1.3	3.0	0.4	0.2	0.6
3	IAP/ NCP	Ma (2019)	11/15-12/9/2016	116.37	39.97	2.3	1.1	2.8	0.6	0.2	0.5
4	PKU/ NCP	X. Ma et al. (2019)	11/16-12/24/2017	116.31	40.00	1.8	1.2	2.7	0.4	0.2	0.5
Summer											
5	Wangdu/ NCP	Tan et al. (2017)	6/7-7/8/2014	115.2	39.5	8.3	6.3	12.9	8.3	2.6	5.2
6	IAP/ NCP	Whalley et al. (2021)	5/21-6/26/2017	116.37	39.97	8.6	3.5	7.6	2.9	3.5	5.9
7	Tiazhou/ YRD	Yang (2021)	5/23-6/17/2018	119.9	32.3	11.1	9.0	14.2	11.4	4.4	6.6
8	Chengdu/ Southwest	Yang et al. (2021)	8/10-8/25/2019	103.8	30.4	10.0	9.8	19.7	10.1	4.0	7.6
Autumn											
9	Heshen/ PRD	Tan et al. (2019)	10/19-11/22/2014	112.9	22.7	4.4	7.0	18.4	3.2	1.8	5.6
10	Shenzhen/ PRD	Yang (2021)	10/5-10/28/2018	113.9	22.6	4.5	7.6	11.8	4.4	5.4	7.8

Table S5. Descriptions of the implemented new sources of HONO.

New sources	Description	Reference
Traffic emission	$E_{\mathrm{HONO,traffic}} = 0.017 \times E_{\mathrm{NO}_x,\mathrm{traffic}}$	Rappengluck et al. (2013)
Soil emission	$E_{\mathrm{HONO,soil}} = \mathrm{HONO}_f \times E_{\mathrm{NO}_x,\mathrm{soil}} \times f_{\mathrm{SWC}} \times \mathrm{CRF}(\mathrm{LAI}, \mathrm{meteology}, \mathrm{biome})$ $E_{\mathrm{NO}_x,\mathrm{soil}}$ and canopy reduction factor (CRF): Hudman et al. (2012) The soil water content adjustment factor (f_{SWC}) and the proportions of the emissions of HONO to NO_x (HONO_f): Rasool et al. (2019)	Hudman et al. (2012) Rasool et al. (2019)
Biomass burning emission	Burned area data: GFED4 $EF_{\mathrm{HONO,bb}}$: Andreae (2019)	Giglio et al. (2013) Andreae (2019)
Heterogeneous reaction of NO_2 on the ground	$\mathrm{NO_2} \xrightarrow{surface} \mathrm{HONO} \quad (k_g)$ $k_{\mathrm{g}} = \frac{1}{8} \times v_{\mathrm{NO_2}} \times \frac{S_{\mathrm{g}}}{V} \times \gamma_{\mathrm{g-NO_2}}$ $\gamma_{\mathrm{g-NO_2}} = \begin{cases} 1 \times 10^{-6} & (\mathrm{night}) \\ 2 \times 10^{-5} \times \frac{J(\mathrm{NO_2})}{2 \times 10^{-3}} & (\mathrm{day}) \end{cases}$ $\frac{S_{\mathrm{g}}}{V} = \begin{cases} 0.1 & (\mathrm{urban\ area}) \\ \frac{2 \times \mathrm{LAI}}{\mathrm{H}} & (\mathrm{other\ areas}) \end{cases}$ Land cover type: MCD12C1 v6	Li et al. (2010); Zheng et al. (2020)
Photolysis of nitrate	$\mathrm{Nitrate} \xrightarrow{h\nu} 0.67\mathrm{HONO} + 0.33\mathrm{NO_2} \quad (J_{Nitrate})$ $J_{Nitrate} = 100 \times J_{HNO_3}$	Kasibhatla et al. (2018)

Table S6. The ratios of IVOCs to NMVOCs emissions from different subsectors.

Subsectors	IVOCs/NMVOCs	Reference
Gasoline	0.032	Lu et al. (2018)
Diesel	0.527	Lu et al. (2018)
Coal	0.352	Cai et al. (2019)
Biomass burning	0.164	Lim et al. (2019)
Solvent use	0.240	Khare and Gentner (2018)

Table S7. Annual mean emissions of IVOC6, IVOC5, and IVOC4 in China in 2014 (unit: Tg year^{-1}). The values in parentheses are the IVOCs emissions when the residential emission is multiplied by a factor of 7.

Sectors	Corresponding subsectors	IVOC6	IVOC5	IVOC4	Total IVOCs
Power	Coal	0.01	0.01	0.01	0.03
Industry	Coal Solvent	2.51	1.43	0.83	4.77
Residential	Coal Biofuel	0.60 (3.10)	0.44 (1.72)	0.17 (0.83)	1.21 (5.64)
Transportation	Gasoline Diesel	0.23	0.18	0.15	0.56
Total	-	3.35 (5.85)	2.05 (3.33)	1.15 (1.81)	6.56 (11.00)

Table S8. The SOA yield parameterization for IVOCs under low and high NO_x conditions in this study compared with the literature parameterization. The saturation concentration (C^*) is in unit of μg m^{-3}. Only one-step oxidation is considered in the parameterizations.

Parameterization	Category	mass-based stoichiometric coefficients (α) for C^*						SOA yield at 10 μg m^{-3}	Reference
		nonvolatile	0.1	1	10	100	1000		
low NO_x condition									
Pye and Seinfeld (2010) (Base)	IVOCs	0.73	-	-	-	-	-	0.73	Chan et al. (2009)
high NO_x condition									
Pye and Seinfeld (2010) (Base)	IVOCs	-	0	0.039	0.296	0.235	-	0.21	Chan et al. (2009)
Koo et al. (2014)	IVOCs	-	-	0.030	0.194	0.264	0.376	0.15	Murphy and Pandis (2009)
Jathar et al. (2014)	*n*-C_{13} proxy	-	0.014	0.059	0.220	0.400	-	0.21	Presto et al. (2010)
	n-C_{15} proxy	-	0.044	0.071	0.410	0.300	-	0.34	
Lu et al. (2020)	IVOC6-ALK	-	0.009	0.045	0.118	0.470	-	0.15	Presto et al. (2010); Zhao et al. (2015); Zhao et al. (2016)
	IVOC5-ALK	-	0.051	0.061	0.394	0.494	-	0.35	
	IVOC4-ALK	-	0.068	0.083	0.523	0.239	-	0.43	
	IVOC3-ALK	-	0.067	0.086	0.544	0.198	-	0.43	
	IVOC6-ARO	-	0.022	0.109	0.251	0.005	-	0.25	
	IVOC5-ARO	-	0.143	0.021	0.329	0.358	-	0.36	
This study	IVOC6	-	0.011	0.052	0.201	0.296	-	0.19	Presto et al. (2010)
	IVOC5	-	0.049	0.078	0.439	0.271	-	0.36	
	IVOC4	-	0.063	0.089	0.550	0.200	-	0.44	

Table S9. The observed campaign-average mass concentrations of primary IVOCs at the SAES site and the corresponding model results in the Cp_R1+2 and Cp_R1+2+3 simulations.

Site	Reference	Period	Observation ($\mu g\ m^{-3}$)	Cp_R1+2 simulation		Cp_R1+2+3 simulation	
				Sim ($\mu g\ m^{-3}$)	Sim/ Obs	Sim ($\mu g\ m^{-3}$)	Sim/ Obs
SAES (Shanghai)	Y. Li et al. (2019)	December 5, 2016 to January 3, 2017	35.1	9.3	0.26	15.6	0.44
		July 16 to August 8, 2017	4	1.9	0.48	-	-

Table S10. The values of EF_{SOAP}/EF_{CO} used in models and the maximum of ΔOA/ΔCO obtained in the experiments based on oxidation flow reactor (OFR). The values in parentheses are the ΔOA/ΔCO uncorrected for losses of low volatility vapors in the experiments.

Site/campaign name	Site/campaign condition	Simulating/Sampling period	EF_{SOAP}/EF_{CO} (g kg^{-1}) or Maximum ΔOA/ΔCO (μg m^{-3} $ppmv^{-1}$)	Reference
		Model		
	China	2014	69 in Sp_base, R1, R1+2 80 in Sp_R1+2+3	This study
	World	2013	69	Pai et al. (2020)
MILAGRO	Mexico City region	15 to 31 March 2006	80	Hodzic and Jimenez (2011)
CalNex	Urban outflow from Los Angeles	15 May to 15 June 2010	69	Hayes et al. (2015); Woody et al. (2016)
SEAC^{4}RS	Summertime aircraft in the southeast US	1 August to 31 October 2013	69	Kim et al. (2015)
WINTER	Wintertime aircraft campaign in the northeastern US	1 February to 15 March 2015	80	Shah et al. (2019)
WINTER	Wintertime aircraft campaign in the northeastern US	1 February to 15 March 2015	69	Schroder et al. (2018)
		OFR experiment		
CalNex	Urban outflow from Los Angeles	29 May to 10 June 2010	45-68 (43)	Ortega et al. (2016)
KORUS-AQ	Aircraft campaign over South Korea and the Yellow Sea	May to June 2016	129 ± 16.5	Nault et al. (2018)
Changping	Suburban near Beijing	12 to 21 June 2016	59	J. Li et al. (2019b)
Hong Kong	The east coast of Hong Kong	11 to 21 October 2016	22	
Pittsburgh	A highway tunnel	May 2013	91	Tkacik et al. (2014)
Hong Kong	1 m from a major road	24 Dec 2017 to 15 Jan 2018	48 (38 ± 65)	T. Liu et al. (2019)
Durham	10 m from the edge of Interstate 40	1 to 15 July 2015 5 to 25 February 2016	180 (62 ± 18) 110 (20)	Saha et al. (2018)
Beijing	The 4th Ring Road	November 2018	44	Liao et al. (2021)

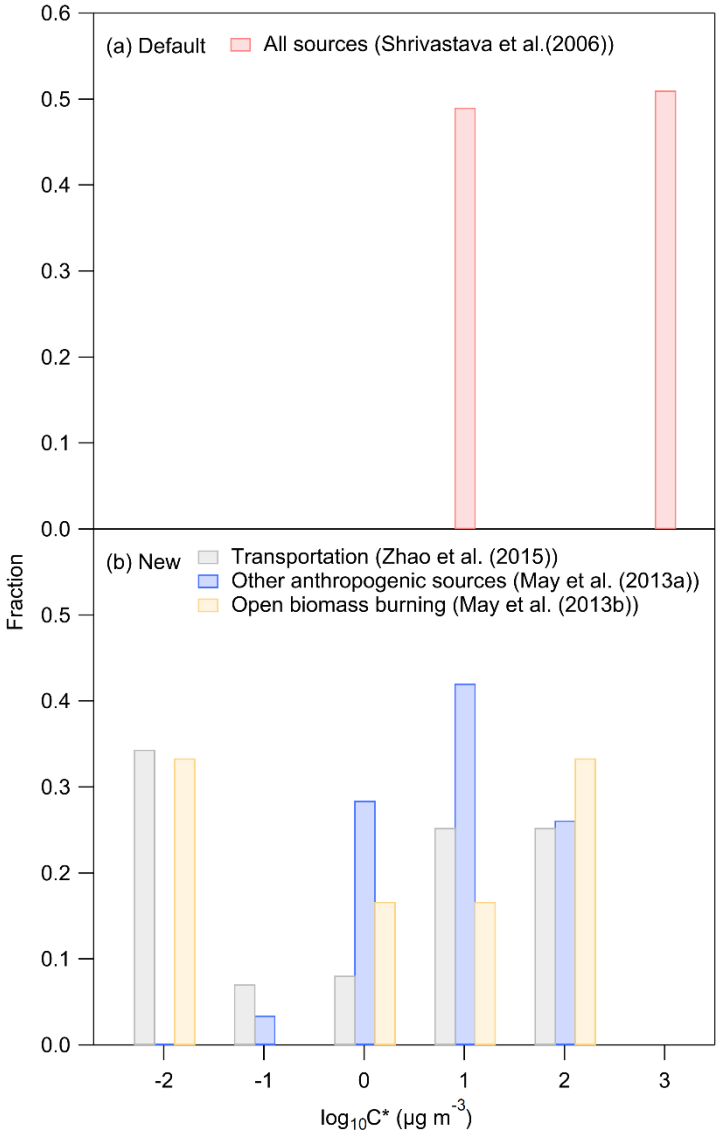

Figure S1. The (a) default and (b) new volatility distributions of primary SVOCs that used in the Cp_base and Cp_R1 schemes, respectively.

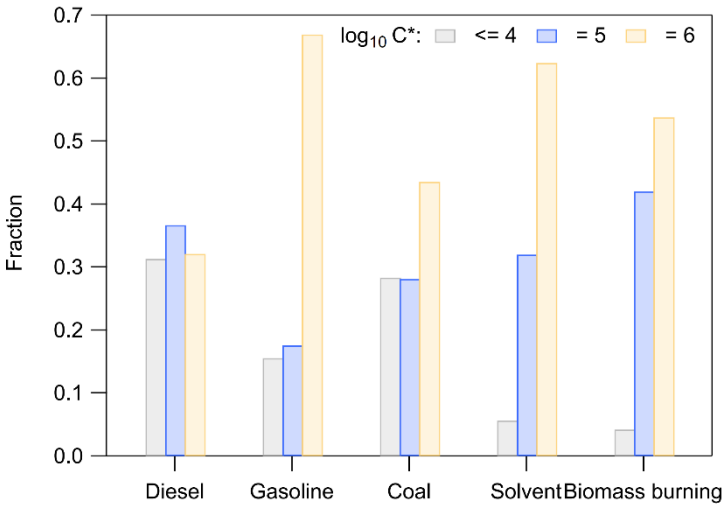

Figure S2. The volatility distributions of primary IVOCs from diesel, gasoline, coal combustion, solvent uses, and biomass burning. The volatility distributions from diesel and gasoline are adopted from Lu et al. (2018). For coal combustion, solvent uses, and biomass burning, the volatility distributions are provided by Cai et al. (2019), Khare and Gentner (2018), and Lim et al. (2019), respectively.

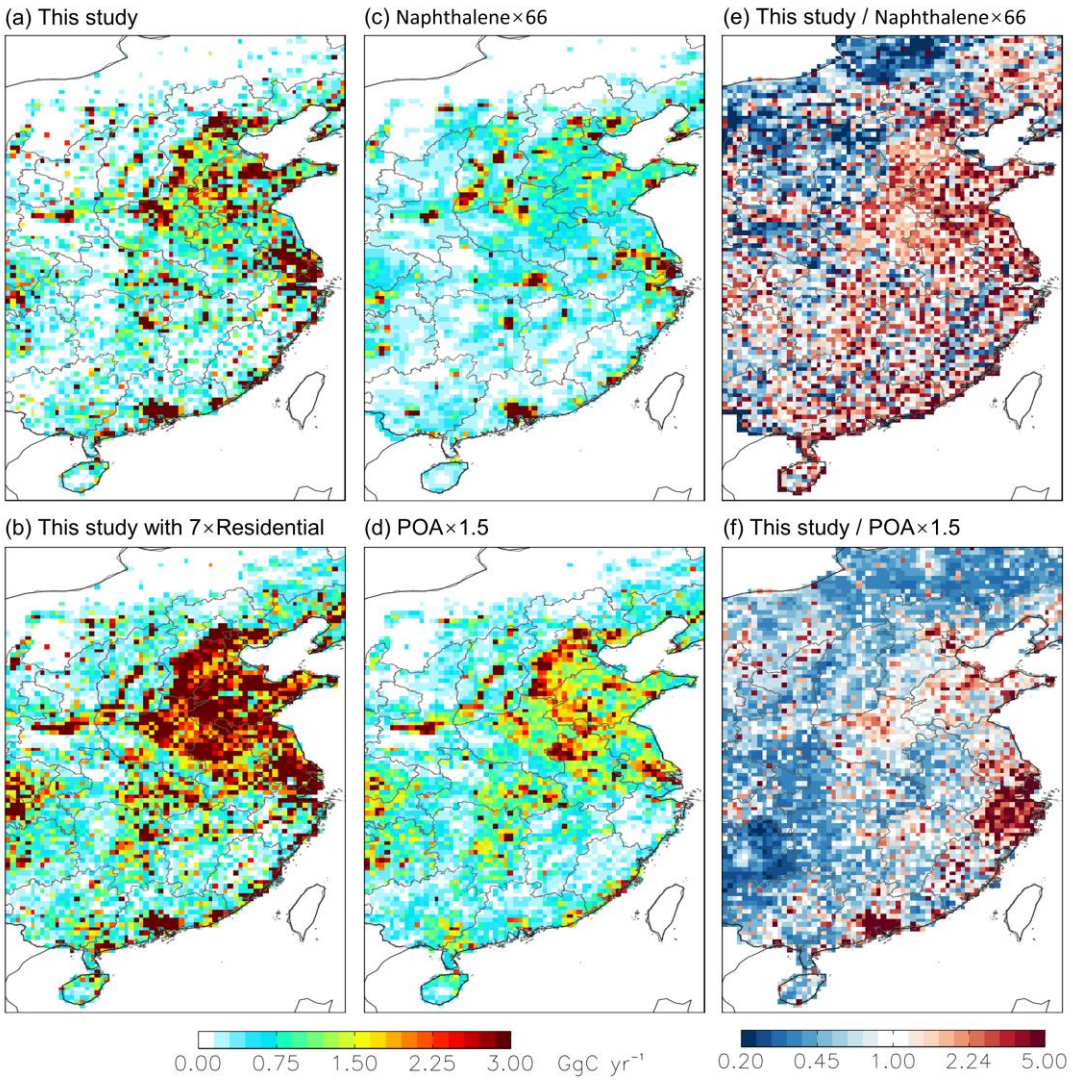

Figure S3. Annual emissions of IVOCs in 2014 that are estimated by (a) the new method developed in this study, (b) the new method with 7-fold emissions from the residential sector during the heating season, (c) the naphthalene-based, and (d) the POA-based methods as well as (e-f) the emission ratios of the new method to naphthalene-based and the new method to POA-based estimates.

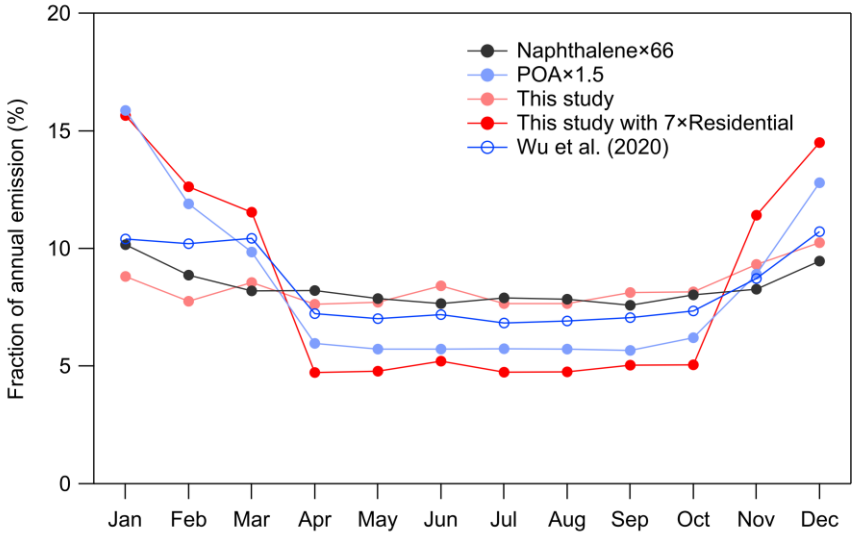

Figure S4. Comparisons of monthly profiles of the IVOC emissions in China in 2014.

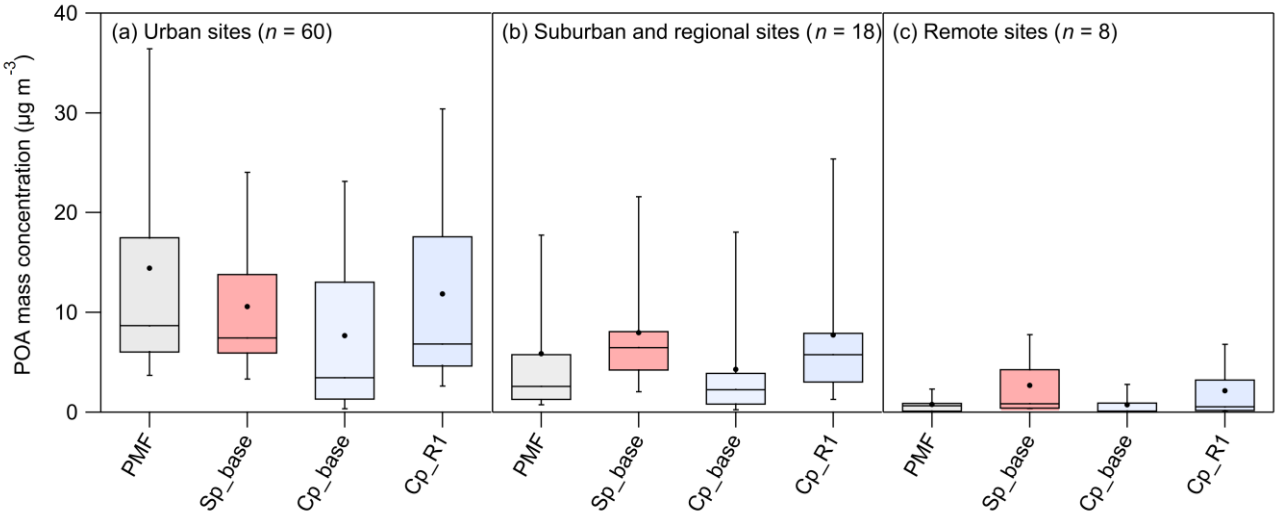

Figure S5. The PMF-derived and simulated campaign-average mass concentrations of POA at (a) urban, (b) suburban and regional, and (c) remote sites. The upper and lower edges of the boxes, the whiskers, the middle lines, and the solid dots denote the 25th and 75th percentiles, the 5th and 95th percentiles, the median values, and the mean values of the POA concentrations.

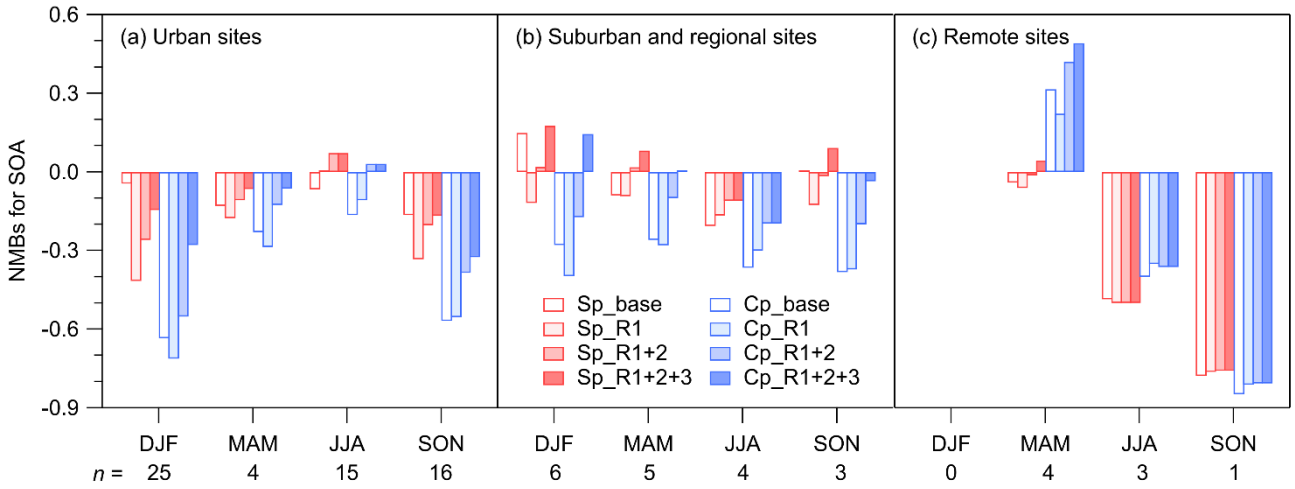

Figure S6. NMBs for SOA simulations at (a) urban, (b) suburban and regional, and (c) remote sites for different seasons. The model results were compared to the PMF-derived SOA results in China.

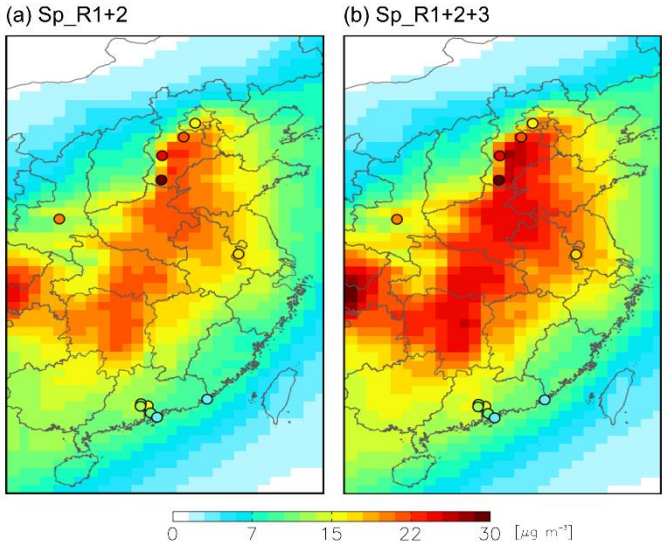

Figure S7. Winter-mean mass concentrations of SOA simulated by the Sp_R1+2 and Sp_R1+2+3 schemes. The circles are shown for the PMF-derived SOA concentrations from measurements.

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