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Estimation of secondary $PM_{2.5}$ in China and the United States using a multi-tracer approach

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Table S1. Impacts of primary sulfate/nitrate emission uncertainty on the estimated secondary proportion of $PM_{2.5}$ [1] in China.

City	Change of sulfate/nitrate emission	
	10%	3%
Beijing	40.3	42.4
Tianjin	61.9	62.6
Shijiazhuang	44.8	45.2
Taiyuan	43.1	44.3
Hohhot	48.6	46.5
Shenyang	48.7	49.3
Changchun	47.9	46.7
Harbin	66.9	67.3
Shanghai	68.0	67.8
Nanjing	50.3	50.5
Hangzhou	45.6	45.3
Hefei	65.4	65.0
Fuzhou	64.8	62.1
Nanchang	62.5	62.7
Jinan	54.6	54.6
Zhengzhou	54.6	56.3
Wuhan	61.5	60.8
Changsha	65.9	64.9
Guangzhou	65.2	65.6
Nanning	65.2	65.2
Haikou	65.9	65.4
Chongqing	62.7	62.8
Chengdu	45.3	44.8
Guiyang	65.6	64.3
Kunming	70.4	70.5
Lhasa	56.1	55.1
Xian	52.6	53.0
Lanzhou	60.0	59.5
Xining	59.1	58.9
Yinchuan	59.5	60.1
Urumqi	72.1	70.3

[1] Based on the MEE observations in 2016.

Table S2. The comparison of two assumptions on weak correlation between secondary $PM_{2.5}$ and X-tracer in the secondary proportions of $PM_{2.5}$ (%) [1].

City	δ1 [2]	δ2 [3]	δ1 - δ2
Beijing	40.3	38.2	2.1
Tianjin	61.9	61.4	0.5
Shijiazhuang	44.8	42.8	2.0
Taiyuan	43.1	40.9	2.2
Hohhot	48.6	46.8	1.7
Shenyang	48.7	47.0	1.7
Changchun	47.9	46.3	1.6
Harbin	66.9	68.3	-1.4
Shanghai	68.0	67.8	0.2
Nanjing	50.3	47.6	2.7
Hangzhou	45.6	43.4	2.2
Hefei	65.4	65.1	0.3
Fuzhou	64.8	63.9	0.8
Nanchang	62.5	61.2	1.4
Jinan	54.6	52.7	1.9
Zhengzhou	54.6	52.8	1.9
Wuhan	61.5	60.4	1.1
Changsha	65.9	65.7	0.2
Guangzhou	65.2	64.9	0.3
Nanning	65.2	64.8	0.5
Haikou	65.9	66.4	-0.4
Chongqing	62.7	61.9	0.7
Chengdu	45.3	43.8	1.4
Guiyang	65.6	66.1	-0.5
Kunming	70.4	70.9	-0.4
Lhasa	56.1	55.1	1.0
Xian	52.6	50.7	1.9
Lanzhou	60.0	59.6	0.4
Xining	59.1	58.3	0.8
Yinchuan	59.5	58.6	0.9
Urumqi	72.1	73.3	-1.2

[1] Based on the MEE observations in 2016.

[2] δ1 is mean value of the estimated interval.

[3] δ2 is the specific value when r=0.

Table S3. List of 31 populous cities and 19 regional background cities and the corresponding averaged $PM_{2.5}$ concentrations (μg m^{-3}) in China during the studying period.

Province	Populous city	$PM_{2.5}$	Regional background city	$PM_{2.5}$
Beijing	Beijing	69.4	-	-
Tianjin	Tianjin	69.5	-	-
Hebei	Shijiazhuang	92.0	Zhangjiakou	33.3
Shanxi	Taiyuan	62.8	-	-
InnerMogolia	Hohhot	42.0	Xilingol	16.2
Liaoning	Shenyang	57.5	-	-
Jilin	Changchun	51.6	Yanbian	31.6
Heilongjiang	Harbin	57.5	Daxinanling	20.4
Shanghai	Shanghai	45.6	-	-
Jiangsu	Nanjing	52.8	-	-
Zhejiang	Hangzhou	49.1	Zhoushan	26.4
Anhui	Hefei	61.6	Huangshan	28.3
Fujian	Fuzhou	27.8	Nanping	23.3
Jiangxi	Nanchang	40.9	-	-
Shandong	Jinan	75.3	Weihai	34.0
Henan	Zhengzhou	80.2	-	-
Hubei	Wuhan	62.0	-	-
Hunan	Changsha	57.9	-	-
Guangdong	Guangzhou	38.4	Shanwei	27.3
Guangxi	Nanning	39.0	Beihai	29.8
Hainan	Haikou	20.5	Sanya	16.0
Chongqing	Chongqing	50.8	-	-
Sichuan	Chengdu	59.4	Aba	15.1
Guizhou	Guiyang	36.7	Qianxinan	18.2
Yunnan	Kunming	28.8	Diqing	13.5
Xizang	Lhasa	22.5	Linzhi	10.5
Shannxi	Xi’an	68.8	-	-
Gansu	Lanzhou	50.1	Jiayuguan	30.9
Qinghai	Xining	49.0	Yushu	18.0
Ningxia	Yinchuan	44.7	Guyuan	35.6
Xinjiang	Urumqi	70.4	Altay	11.3

Table S4. List of $PM_{2.5}$ component measurements ($\mu g\ m^{-3}$) of China in previous studies.

City	Period	$PM_{2.5}$	SO_4^{2-}	NO_3^-	NH_4^+	SOA[1]	SPM/$PM_{2.5}$	Mean SPM/$PM_{2.5}$	Method	References
Beijing	2012 Summer	103	20.6	15.8	8.3	10.1	53.2%	**31% ~ 80%**	Offline	Tian et al., 2015
	2012 - 2013	72	9.3	11.9	5.3	9.6	50.3%		Offline	Liu et al., 2018b
	2013 Winter	159	25.4	19.0	15.6	32.2[a]	58.0%		Offline	Tao et al., 2015
	2013 Winter	143	23.9	20.2	16.5	15.4	53.1%		AMS	Huang et al., 2014b
	Jan 2014	153	9.6	12.1	6.7	33.8[c]	40.6%		Offline	Gao et al., 2018
	Apr 2014	115	10.7	10.7	11.4	15.2[c]	41.6%		Offline	Gao et al., 2018
	Jul 2014	96	25.6	25.6	14.1	11.1[c]	79.7%		Offline	Gao et al., 2018
	Oct 2014	139	21.1	45.5	13.9	23.0[c]	74.5%		Offline	Gao et al., 2018
	2014 Winter	138	21.0	26	14.1	17.4	51.8%		Offline	Lin et al., 2016
	Jun 2014 - Apr 2015	100	14.3	17.1	11.5	12.4[b]	55.6%		Offline	Huang et al., 2017
	May 2015 - Apr 2016	114	8.6	11.1	5.2	10.1	30.9%		Offline	Yu et al., 2019
	Jul 2015 - Apr 2016	81	9.6	12.4	8.6	7.7	47.1%		Offline	Xu et al., 2019
	Oct 2016	95	16.8	16.8	12.5	12.3[b]	61.5%		Offline	Zhang et al., 2018
	MTEA estimation [2]							**41%**	-	This study
Tianjin	Jun 2014 - Apr 2015	106	16.6	16.2	13.7	10.4[b]	53.8%	**41% ~ 54%**	Offline	Huang et al., 2017
	Jul 2015 - Apr 2016	86	12.1	13.9	10.5	7.3	51.0%		Offline	Xu et al., 2019
	Jul 2014	113	12.2	16.2	9.3	11.0[c]	43.0%		Offline	Gao et al., 2018

	Oct 2014	101	12.8	9.9	8.2	11.1[c]	41.4%		Offline	Gao et al., 2018
	2014 Winter	183	19.5	40.7	15.1	21.8[c]	53.1%		Offline	Gao et al., 2018
	May 2015 – Apr 2016	120	18.1	20.3	8.5	10.0	47.4%		Offline	Liu et al., 2018a
	MTEA estimation							**63%**		This study
Shijiazhuang	Jun 2014 - Apr 2015	155	25.5	23.4	18.8	17.7[b]	55.0%	**51% ~ 55%**	Offline	Huang et al., 2017
	Jul 2015 - Apr 2016	105	16.8	14.9	12.3	9.6	51.0%		Offline	Xu et al., 2019
	MTEA estimation							**49%**		This study
Shanghai	2012 Spring	70	15.3	8.6	6.4	5.7	51.4%	**26% ~ 71%**	Offline	Zhao et al., 2015
	2012 Summer	51	9.7	5.6	3.6	3.7	44.3%		Offline	Zhao et al., 2015
	2012 Fall	82	17.9	20.2	7.8	7.7	65.4%		Offline	Zhao et al., 2015
	2012 Winter	70	11.6	13.2	5.6	8.5	55.6%		Offline	Zhao et al., 2015
	2012 Spring	64	12.0	10.8	4.3	4.9	50.0%		Offline	Huang et al., 2014a
	2011- 2013 Spring	49	11.0	11.0	6.9	5.9	71.0%		Offline	Wang et al., 2016a
	2011- 2013 Summer	31	8.1	5.2	4.2	4.7	67.3%		Offline	Wang et al., 2016a
	2011- 2013 Fall	41	8.8	7.4	4.8	5.2	63.9%		Offline	Wang et al., 2016a
	2011- 2013 Winter	65	13.0	13.2	8.3	6.7	63.4%		Offline	Wang et al., 2016a
	2012 - 2013	68	13.6	11.9	5.8	8.6	58.7%		Offline	Liu et al., 2018b
	Oct - Nov 2013	75	12.9	15.0	6.6	4.2	51.6%		Offline	Ming et al., 2017
	Dec 2013 – Jan 2014	138	19.5	29.1	12.6	10.3	51.8%		Offline	Ming et al., 2017
	Mar 2014 – Apr 2014	96	12.3	10.4	5.5	4.5	34.1%		Offline	Ming et al., 2017
	Jun 2014 – Jul 2014	56	6.7	2.8	2.1	2.9	25.9%		Offline	Ming et al., 2017

	2013 Winter	91	10.8	12.4	7.5	21.8[b]	57.7%		AMS	Huang et al., 2014b
	Dec 2014 – Jan 2015	103	18.3	25.4	14.4	14.1[b]	70.1%		Offline	Du et al., 2017
	Mar 2015 – Apr 2015	74	8.7	11.2	5.7	9.2[b]	47.0%		Offline	Du et al., 2017
	MTEA estimation							**67%**		This study
Nanjing	Apr – May 2013	110	23.1	11.7	6.4	17.7[a]	53.5%	**52% ~ 79%**	Offline	Li et al., 2016
	Aug 2013	86	18.4	8.1	5.4	14.2[a]	53.6%		Offline	Li et al., 2016
	Oct 2013	77	12.6	7.3	3.8	36.8[a]	78.6%		Offline	Li et al., 2016
	Dec 2014 – Jan 2015	100	11.7	16.4	12.3	11.8[b]	52.2%		Offline	
	Mar 2015 – Apr 2015	83	21.4	16.1	7.9	9.1[b]	65.6%		Offline	Du et al., 2017
	MTEA estimation							**53%**		This study
Hangzhou	Oct 2013	36	9.7	5.3	6.0	6.5	76.4%	**76%**	Offline	Wu et al., 2016
	MTEA estimation							**53%**		This study
Guangzhou	Dec 2012 – Jan 2013	75	10.6	5.8	5.1	6.5	37.3%	**37% ~ 58%**	Offline	Liu et al., 2014
	2012 - 2013	75	13.1	7.2	4.8	8.4	44.6%		Offline	Liu et al., 2018b
	Nov 2012 – Dec 2013	61	9.3	5.7	4.2	11.9[b]	51.0%		Offline	Chen et al., 2016
	2013 Summer	51	8.9	4.9	4.0	6.6	47.8%		Offline	Cui et al., 2015
	2013 Fall/Winter	68	9.8	7.3	4.5	9.4	45.6%		Offline	Cui et al., 2015
	2013 Winter	69	12.7	8.9	6.9	11.4[b]	57.8%		AMS	Huang et al., 2014b
	2014 Spring	44	8.2	2.4	3.6	4.5	42.5%		Offline	Tao et al., 2017

	2014 Summer	37	7.6	0.3	2.6	3.7	38.4%		Offline	Tao et al., 2017
	2014 Fall	48	11.4	1.0	4.4	4.7	44.8%		Offline	Tao et al., 2017
	2014 Winter	63	9.8	5.5	4.8	7.0	43.0%		Offline	Tao et al., 2017
	MTEA estimation							**66%**		This study
Xi'an	Dec 2012	137	13.5	9.8	6.6	21.6[b]	37.6%		Offline	Zhang et al., 2015
	2012 Spring	164	17.8	15.2	6.5	13.9	32.6%		Offline	Niu et al., 2016
	2012 Summer	109	25.0	10.1	6.6	8.8	46.3%		Offline	Niu et al., 2016
	2012 Fall	155	18.7	16.5	8.2	18.4	39.9%	**33% ~ 55%**	Offline	Niu et al., 2016
	Nov 2012 – Feb 2013	244	32.1	29.3	16.8	39.7	48.3%		Offline	Niu et al., 2016
	Dec 2014 – Nov 2015	113	15.2	16.6	8.4	21.3	54.7%		Offline	Dai et al., 2018
	MTEA estimation							**55%**		This study
Chengdu	Oct – Nov 2014	62	10.5	9.3	6.9	8.3[b]	56.5%		Offline	Wang et al., 2018
	Jan – Feb 2015	114	16.4	17.5	12.7	15.8[b]	54.7%		Offline	Wang et al., 2018
	Apr 2015	48	8.3	5.9	5.1	5.0[b]	50.6%	**44% ~ 57%**	Offline	Wang et al., 2018
	Jul 2015	45	9.7	3.9	4.2	5.9[b]	52.6%		Offline	Wang et al., 2018
	Jan 2015	48	6.1	3.7	2.4	8.7	43.5%		Offline	Li et al., 2017
	MTEA estimation							**46%**		This study
Chongqing	2012 - 2013	74	19.7	6.5	6.1	8.6	55.3%		Offline	Liu et al., 2018b
	Oct – Nov 2014	56	9.9	7.8	5.7	7.8[b]	55.7%		Offline	Wang et al., 2018
	Jan – Feb 2015	115	17.5	15.8	11.3	19.4[b]	55.7%	**44% ~ 56%**	Offline	Wang et al., 2018
	Apr 2015	58	10.4	5.9	5.2	8.0[b]	50.1%		Offline	Wang et al., 2018
	Jul 2015	54	11.1	1.6	4.0	6.8[b]	43.5%		Offline	Wang et al., 2018

	MTEA estimation							**61%**		This study
Lanzhou	Dec 2012	120	11.8	7.2	6.7	21.2	39.1%	**18% ~ 41%**	Offline	Tan et al., 2016
	Jun – Jul 2013	34	4.3	1.9	1.9	5.8	40.9%		Offline	Tan et al., 2016
	Apr – May 2014	83	4.0	1.7	0.8	8.0	17.5%		Offline	Wang et al., 2016b
	Aug 2014	38	4.8	2.0	1.3	3.5	30.5%		Offline	Wang et al., 2016b
	Oct 2014	93	5.8	7.1	3.6	12.7	31.4%		Offline	Wang et al., 2016b
	Jan, Dec 2014	141	7.6	10.1	6.0	18.2	29.7%		Offline	Wang et al., 2016b
	MTEA estimation							**63%**		This study
Changsha	Sep – Oct 2013	102	19.4	2.6	8.7	12.5[b]	42.4%	**41% ~ 44%**	Offline	Tang et al., 2017
	Dec 2013 – Jan 2014	145	19.3	9.7	14.3	20.5[b]	44.0%		Offline	Tang et al., 2017
	Apr – May 2014	97	17.0	1.4	7.5	14.0[b]	41.1%		Offline	Tang et al., 2017
	Jul – Aug 2014	78	13.9	2.9	7.4	9.4[b]	43.1%		Offline	Tang et al., 2017
	MTEA estimation							**67%**		This study
Haikou	Jan 2015	17	3.1	0.5	1.0	2.3	40.1%	**32% ~ 40%**	Offline	Liu et al., 2017
	Mar 2015	9	1.6	0.2	0.5	1.2	38.8%		Offline	Liu et al., 2017
	Jul 2015	23	3.8	0.3	0.8	2.4	31.7%		Offline	Liu et al., 2017
	Sep 2015	47	7.9	3.1	3.0	2.8	35.7%		Offline	Liu et al., 2017
	MTEA estimation							**61%**		This study
Zhengzhou	Oct 2014	143	19.6	17.9	9.2	12.0	41.0%	**41% ~ 54%**	Offline	Jiang et al., 2017
	Dec 2014 – Jan 2015	191	23.5	26.5	19.8	22.6	48.4%		Offline	Jiang et al., 2017
	Apr 2015	138	19.7	20.3	14.4	11.3	47.6%		Offline	Jiang et al., 2017
	Jul 2015	110	24.2	14.3	13.9	7.3	54.3%		Offline	Jiang et al., 2017
	MTEA estimation							**60%**		This study

Shenyang	2013 – 2014	82	13.2	4.6	4.5	11.7	41.5%	**42%**	Offline	Liu et al., 2018b
	MTEA estimation							**51%**		This study
Lhasa	2013 – 2014	36	0.8	0.5	0.4	7.6	25.8%	**26%**	Offline	Liu et al., 2018b
	MTEA estimation							**64%**		This study

[1] SOA = 0.5*OM, OM = f * OC. Default f is 1.2. In case of a, b and c, the f is 1.8, 1.6 and 1.4 respectively.

[2] For period of 2014-2018.

Table S5. Impacts of anthropogenic emission uncertainty on the estimated secondary proportion of $PM_{2.5}$ [1] in China.

City	Secondary proportion of $PM_{2.5}$	Change of secondary proportion of $PM_{2.5}$	
		$a - 0.1$	$a + 0.1$
Beijing	40.3	-2.9	2.1
Tianjin	61.9	-3.0	0.7
Shijiazhuang	44.8	-1.7	0.4
Taiyuan	43.1	-2.8	1.1
Hohhot	48.6	3.2	-2.1
Shenyang	48.7	-1.7	0.7
Changchun	47.9	2.2	-1.1
Harbin	66.9	-0.5	0.4
Shanghai	68.0	-6.7	-0.2
Nanjing	50.3	0.6	0.2
Hangzhou	45.6	0.0	-0.3
Hefei	65.4	-4.2	-0.5
Fuzhou	64.8	3.9	-2.7
Nanchang	62.5	-1.0	0.2
Jinan	54.6	0.4	0.1
Zhengzhou	54.6	-3.6	1.7
Wuhan	61.5	-2.4	-0.8
Changsha	65.9	-2.5	-1.0
Guangzhou	65.2	-1.5	0.5
Nanning	65.2	-1.8	-0.1
Haikou	65.9	1.2	-0.5
Chongqing	62.7	-1.2	0.2
Chengdu	45.3	2.8	-0.5
Guiyang	65.6	1.7	-1.2
Kunming	70.4	-2.5	0.0
Lhasa	56.1	1.9	-1.0
Xian	52.6	-0.5	0.4
Lanzhou	60.0	1.5	-0.5
Xining	59.1	-1.0	-0.2
Yinchuan	59.5	-0.7	0.7
Urumqi	72.1	-6.8	-1.8

[1] Based on the MEE observations in 2016.

Table S6. Impacts of tracer concentration uncertainty on the estimated secondary proportion of $PM_{2.5}$ [1] in China (Unit: %).

City	Secondary proportion of $PM_{2.5}$	Change of secondary proportion of $PM_{2.5}$	
		1.1 * CO concentration & 0.9 * PMC concentration	0.9 * CO concentration & 1.1 * PMC concentration
Beijing	40.3	-0.01	0.01
Tianjin	61.9	-0.32	-0.52
Shijiazhuang	44.8	-0.26	-0.28
Taiyuan	43.1	0.22	0.17
Hohhot	48.6	-0.03	-0.01
Shenyang	48.7	-0.06	-0.06
Changchun	47.9	0.03	0.04
Harbin	66.9	0.22	-0.59
Shanghai	68.0	-1.51	-1.90
Nanjing	50.3	0.00	0.03
Hangzhou	45.6	-0.42	-0.46
Hefei	65.4	-1.57	-1.73
Fuzhou	64.8	-0.25	-0.44
Nanchang	62.5	-0.33	-0.42
Jinan	54.6	-0.04	-0.02
Zhengzhou	54.6	0.14	0.14
Wuhan	61.5	-1.45	-1.49
Changsha	65.9	-1.60	-1.74
Guangzhou	65.2	0.00	-0.28
Nanning	65.2	-0.22	-0.47
Haikou	65.9	-0.15	-0.09
Chongqing	62.7	-0.23	-0.31
Chengdu	45.3	0.42	0.44
Guiyang	65.6	-0.22	-0.50
Kunming	70.4	-0.40	-0.69
Lhasa	56.1	0.07	0.05
Xian	52.6	-0.04	-0.01
Lanzhou	60.0	0.15	0.02
Xining	59.1	-0.56	-0.60
Yinchuan	59.5	0.02	-0.06
Urumqi	72.1	-2.70	-2.85

[1] Based on the MEE observations in 2016.

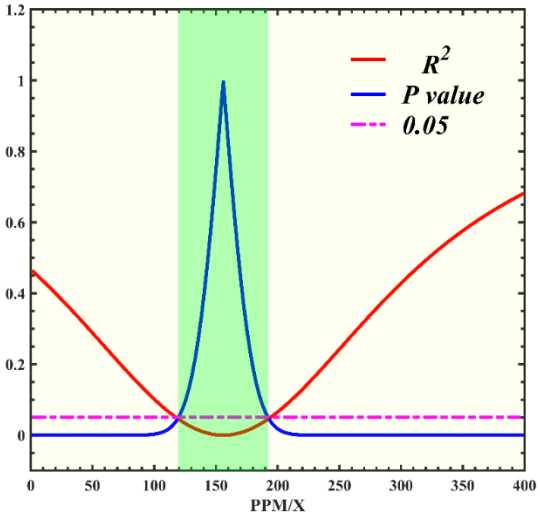

Figure S1. An illustration for scanning the aim interval (green shadow) in the MTEA approach.

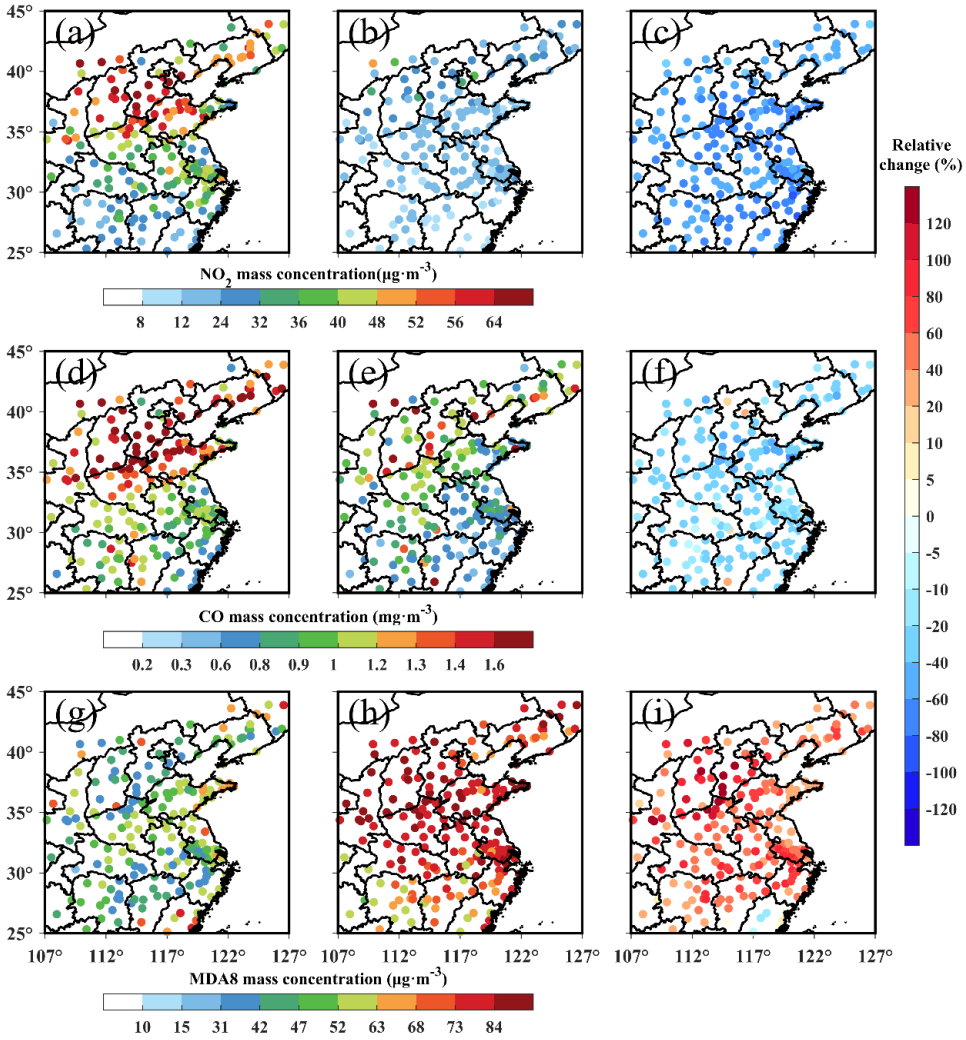

Figure S2. The spatial distribution of (a-b) NO_2, (d-e) CO and (g-h) maximum daily 8-h average O_3 (MDA8) mass concentrations before (01~23 Jan 2020) and during (23-Jan ~ 17-Feb 20) COVID-19 national lockdown. The right panel (c, f, and g) indicates relative change, i.e. (post-lockdown – pre-lockdown)/pre-lockdown.

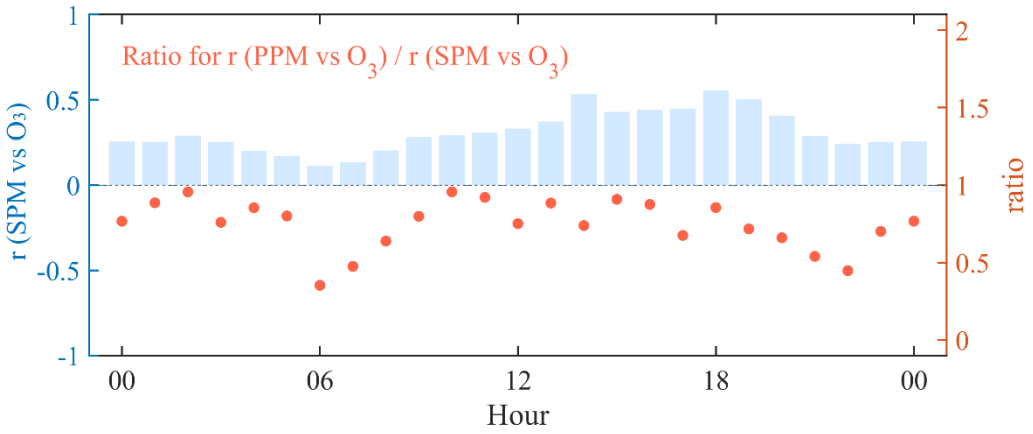

Figure S3. The nationwide correlation between PM versus O_3. Bule bars denote the correlation coefficients between SPM and O_3. The red dots indicate the ratios of the correlation coefficient of PPM vs O_3 to that of SPM vs O_3.

Reference

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