Supplement of

Modeling SOA contributions of VOC, IVOC and SVOC emissions and large uncertainties associated with OA aging

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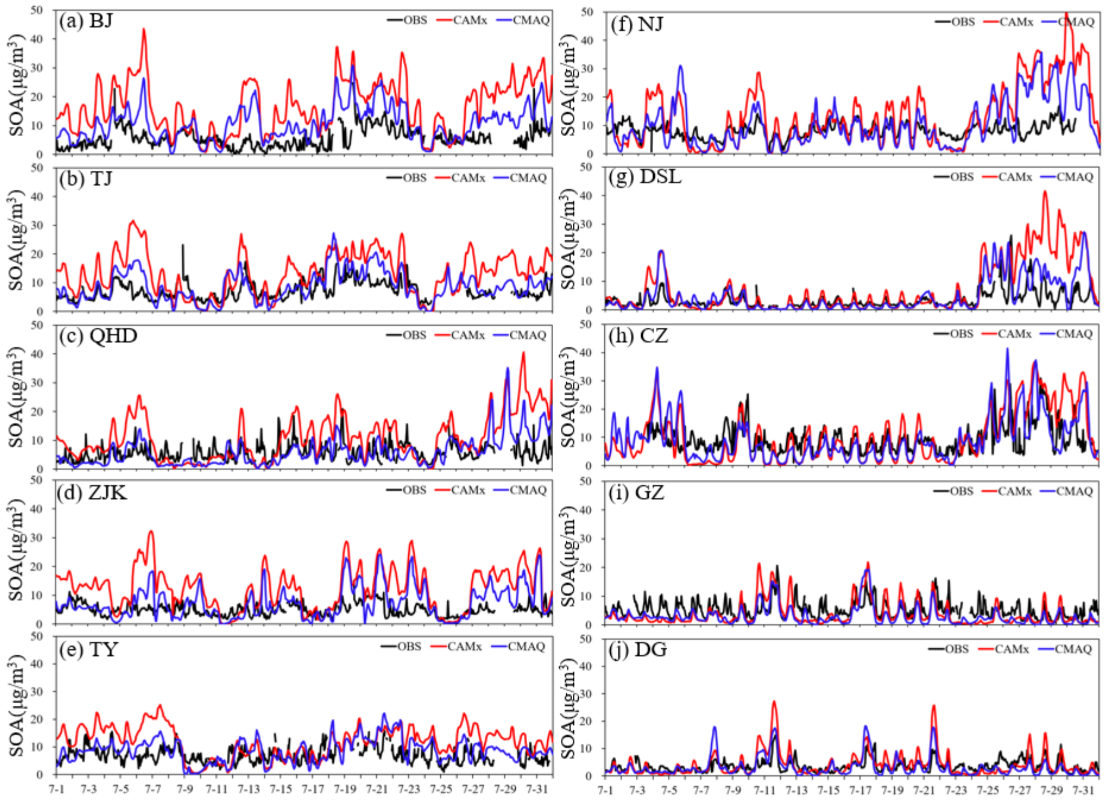

Figure S1 Time series of hourly observed and simulated SOA concentration for CMAQ and CAMx including S/IVOC emissions at different sites in July 2018 (see site information in Table S3).

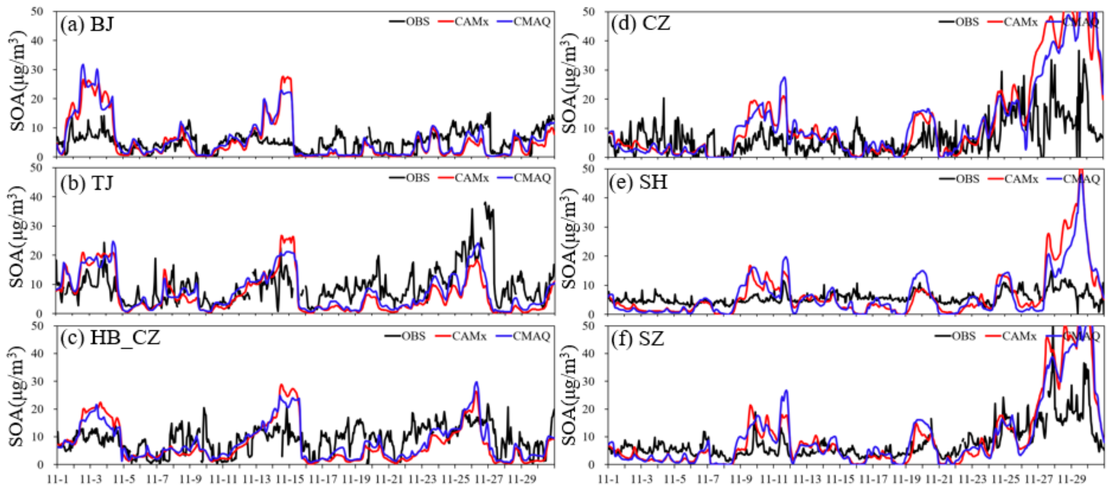

Figure S2 Time series of hourly observed and simulated SOA concentration for CMAQ and CAMx including S/IVOC emissions at different sites in November 2018 (see site information in Table S3).

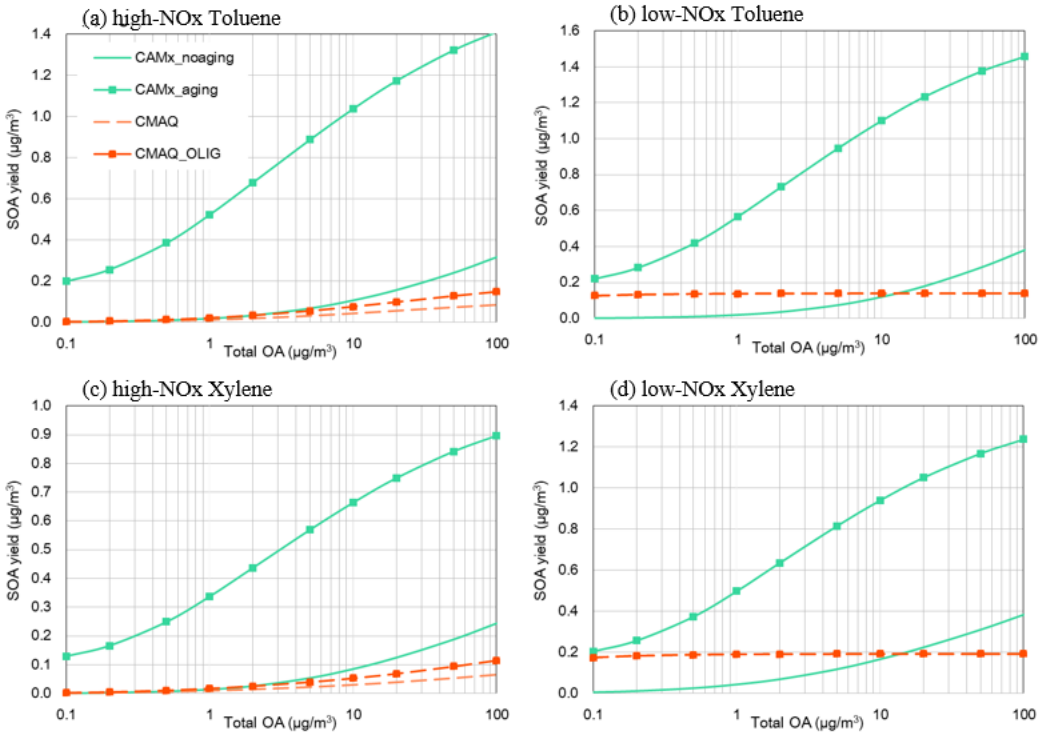

Figure S3 Conceptual SOA mass yields (μg/m^3 SOA formed per μg/m^3 precursor reacted) from toluene (a, b) and xylene (c, d) under high- (a, c) and low-NO*x* (b, d) conditions by different model configurations.

Emission inventory

In the study, we combined the emissions of CB05 in the MEIC inventory with the emissions of specific VOCs in SAPRC07 to obtain the emissions of specific VOCs in the CB6 inventory (**Table S1**). For the emission inventory used by CMAQ, in order to increase the variety of its precursors, some specific VOCs in the inventory have been changed to obtain a more accurate inventory. We mainly adjusted the following species for the CMAQ emissions inventory, such as naphthalene (NAPH) = 0.002×XYL, xylene and other polyalkyl aromatics except naphthalene (XYLMN) = 0.998×XYL, tracer for alkanes that can form secondary organic aerosol (SOAALK) = 0.108×Paraffin carbon bond (PAR).

Table S1 Consolidation of CB05 and SARPC07 species into CB6

No.	CB6	CB05	SAPRC07	No.	CB6	CB05	SAPRC07
1	Acetone (ACET)		✓	11	Ethyne (ETHY)		ACYE
2	Acetaldehyde (ALD2)	✓		12	Ethanol (ETOH)	✓	
3	Aldehydes (ALDX)	✓		13	Formaldehyde (FORM)	✓	
4	BENZ		✓	14	XYL	✓	
5	CH4		✓	15	Internal olefin carbon bond (IOLE)	✓	
6	Ethene (ETH)	✓		16	ISOP	✓	
7	Ethane (ETHA)	✓		17	Ketone carbon bond (KET)		Methyl ethyl ketone (MEK)
8	Methanol (MEOH)	✓		18	Propane (PRPA=0.8ALK2)		ALK2
9	PAR(=PAR-3ACET-BENZ-2ETHY-KET-1.5PRPA)			19	Terminal olefin carbon bond (OLE)	✓	
10	TERP	✓		20	TOL	✓	

Table S2 Scaling factors adopted to allocate S/IVOC

Emission surrogate	CMAQ-species	Phase mode	Scale factor
IVOC	PCVOC	GAS	0.0047
SVOC	VLVPO1	GAS	0
	VSVPO1	GAS	0.045
	VSVPO2	GAS	0.14
	VSVPO3	GAS	0.18
	VIVPO1	GAS	0.50
	ALVPO1	FINE	0.09
	ASVPO1	FINE	0.045
	ASVPO2	FINE	0
	ASVPO3	FINE	0
	AIVPO1	FINE	0

Table S3 Location of 16 monitoring sites and $(OC/EC)_{min}$ value for each sites

Month	Site	Site	Longitude (°)	Latitude (°)	$(OC/EC)_{min}$
Jul.	Beijing	BJ	116.4	39.8	0.7
	Tianjin	TJ	117.2	39.0	1.1
	Qinhuangdao	QHD	119.6	39.9	0.7
	Zhangjiakou	ZJK	114.8	40.8	1.1
	Taiyuan	TY	112.5	37.8	1.4
	Nanjing	NJ	118.7	32.0	0.4
	Changzhou	CZ	119.9	31.7	0.3
	Dianshan Lake	DSL	120.9	31.0	1.0
	Guangzhou	GZ	113.2	23.1	0.8
	Dongguan	DG	113.7	23.0	1.1
Nov.	Beijing	BJ	116.4	39.0	1.2
	Tianjin	TJ	117.2	39.0	1.3
	Cangzhou	HB_CZ	116.7	38.2	1.0
	Changzhou	CZ	119.9	31.7	1.1
	Shanghai	SH	121.5	31.2	1.4
	Suzhou	SZ	120.6	31.3	0.4

Table S4. Definition of model performance evaluation metrics

No.	Statistics (abbreviation)	Definition	Note
1	Correlation coefficient (R)	$\frac{\sum[(P_j-\bar{P})\times(O_j-\bar{O})]}{\sqrt{\sum(P_j-\bar{P})^2\times\sum(O_j-\bar{O})^2}}$	Unitless, -1≤R≤1
2	Mean bias (MB)	$\frac{\sum(\mathrm{P_j}-\mathrm{O_j})}{\mathrm{N}}$	concentration unit
3	Normalize mean bias (NMB)	$\frac{\sum(P_j-O_j)}{\sum O_j}\times 100$	-100%≤NMB≤+∞
4	Index of agreement (IOA)	$1-\frac{\sum(P_j-O_j)^2}{\sum(\lvert P_j-\bar{O}\rvert+\lvert O_j-\bar{O}\rvert)^2}$	Unitless, -1≤IOA≤1
5	2x the proportion of data within the line (FAC2)	$0.5\leq\frac{\mathrm{Pj}}{\mathrm{Oj}}\leqslant 2$	0≤FAC2≤100%
6	Fractional Bias (FB)	$\mathrm{FB}=\frac{1}{n}\left\{\frac{\sum_1^n(\mathrm{P}-\mathrm{O})}{\sum_1^n\left[\frac{(\mathrm{P}+\mathrm{O})}{2}\right]}\right\}$	-100%≤FB≤100%

Pj and Oj are simulated value and observed value, respectively. $\bar{\mathrm{P}}$ and $\bar{\mathrm{O}}$ are the mean of the simulated value and the average of the observed value, respectively.

Table S5 Summary of existing S/IVOC emission inventory in China

Reference	Region	Methods	Sector	IVOC emissions	SVOC emissions	S/IVOC emissions	Contribution (%)
Liu et al. 2017	China	Emission factor	On-road	200.4 (Gg/year)	/	/	/
Miao et al. 2021	China	Naphthalene×66	MEIC sectors	3.8 (Tg/year)	/	/	/
		POA×1.5		5.7	/	/	/
		NMVOC-based		6.6	/	/	/
		Sector and subsector specified	Residential	11.0	/	/	/
Wu et al. 2019	PRD	POA ratio	Industry	/	/	114.6 (Gg/year)	35.4
			Residential			8.4	2.6
			On-road			134.4	41.6
			Off-road			4.8	1.5
			Dust			46.8	14.5
			Biomass burning			14.4	4.5
			Total			323.4	100
Cai et al. 2019	Northern China	Emission factor	Residential coal	/	/	53.9 (Gg/year)	/
Zhu et al. 2020	China	POA ratio	Biomass burning	734 (Gg/year)	/	/	/
Li et al. 2020	BTH	POA ratio	Transportation Industry Residential	IVOC-low 0.09 (Tg/year); IVOC-medium 0.36 (Tg/year)	2×POA	/	/

Wu et al. 2021	China	POA ratio	Industry	/	/	4.6 (Tg/year)	48.0
			Residential			2.9	30.2
			Transportation			1.2	12.7
			Power plants			5.2×10^{-5}	0.0
			Shipping			0.7	7.8
			Total			9.6	100
Qian et al. 2021	China	Emission factor	Residential solid fuel combustion	175.9±16.0Gg	/	/	/
Huang et al. 2021	YRD	Emission factor and POA ratio	Industry	156.1/156.1 (Gg/year)	/	/	49.9/21.4
			Residential combustion	1.3/90.3			0.4/12.4
			On-road	16.0/263.6			5.1/36.1
			Off-road	74.4/74.4			23.8/10.2
			Dust	37.7/37.7			12.0/5.2
			Biomass burning	15.0/95.6			4.8/13.1
			Cooking	12.4/12.4			4.0/1.7
			Total	312.8/730.1			100
An et al. 2022	YRD	POA ratio	Industry process	/	/	249.3 (Gg/year)	22.1
			Industry solvent-use			489.4	43.4
			Mobile			362.9	28.4
			Residential			127.3	5.2
			Agriculture			17.8	0.9
			Total			1,246.7	100

Chang et al. 2022	China	POA ratio	Power and industrial boiler	91.1 (Gg/year)	26.7 (Gg/year)	/	2.3/2.3
			Industry process	115.5	75.4		2.9/6.5
			Domestic fossil fuel combustion	765.7	351.4		19.4/30.1
			Domestic biomass burning	493.3	317.2		12.5/27.1
			On-road gasoline vehicles	83.4	4.0		2.1/0.3
			On-road diesel vehicles	89.4	19.8		2.3/1.7
			Off-road transportation	178.9	14.7		4.5/1.3
			Biomass open burning	871.9	347.5		22.1/29.7
			Domestic VCP	681.1	6.9		17.3/0.6
			Industrial VCP	568.2	5.7		14.4/0.5
			Total	3,938.5	1,169.3		100
Wang et al. 2022	China	Emission factor	Mobile	241.2(Gg/year)	/	/	/

Table S6 Evaluation results of $PM_{2.5}$ simulation in selected areas

Month	Region	CAMx		CMAQ	
		MB (μg/m³)	NMB (%)	MB (μg/m³)	NMB (%)
Jul.	Northeast	3.1	15.1	-3.3	-16.2
	NCP	8.2	25.4	-5.3	-16.0
	YRD	6.6	27.5	-1.1	-5.3
	PRD	3.2	17.8	-1.5	-9.0
	Central China	8.8	38.4	4.0	17.2
	Sichuan Basin	10.6	53.4	3.9	20.9
Nov	Northeast	-1.6	-4.3	10.5	9.3
	NCP	-3.9	-5.2	-8.2	-11.4
	YRD	28.4	54.8	15.5	30.2
	PRD	15.8	46.3	4.4	13.4
	Central China	30.3	70.9	18.2	41.3
	Sichuan Basin	24.8	59.6	8.8	21.4

Table S7 Evaluation results of SOA simulation in selected sites

Mon	Site	Observed average (μg/m³)	CAMx				CMAQ			
			Simulated average (μg/m³)	MB (μg/m³)	NMB (%)	FB (%)	Simulated average (μg/m³)	MB (μg/m³)	NMB (%)	FB (%)
Jul.	BJ	6.3	16.6	10.1	159	80	10.7	4.2	66	45
	TJ	7.1	13.2	6.1	84	40	8.5	1.4	19	-1
	QHD	5.5	10.9	5.1	91	76	6	0.1	1	-15
	ZJK	5	12.4	7.2	145	59	7.4	2.3	45	9
	TY	6.9	11.8	5	72	42	8.7	1.8	27	15
	NJ	8.3	13.5	4.7	89	7	10.5	2.1	25	-8
	DSL	3.5	7.3	4	108	10	4.8	1.2	34	-18
	CZ	9.3	10	0.9	10	-28	9	-0.5	-6	-28
	GZ	5.3	3.7	-1.6	-30	-54	3.1	-2.2	-42	-71
	DG	3.5	4.1	-0.5	15	-10	3.2	-0.4	-12	-35
Nov.	BJ	5.3	5.4	0.1	1	-44	6.1	0.8	14	-19
	TJ	9	6.5	-2.8	-31	-58	7.6	-1.8	-19	-35
	HB_CZ	8.8	7.4	-1.6	-18	-35	8.1	-0.9	-10	-16
	CZ	6.8	12.4	5.6	83	9	12.1	5.3	79	16
	SH	5.7	6.7	1	18	-34	6.5	0.8	13	-39
	SZ	7.7	10	2.3	30	-26	10	2.3	30	-24

Table S8 Domain averaged concentration for each OA component by selected region (μg/m³)

Month	Region	CAMx					CMAQ				
		POA	ASOA	BSOA	IVOCs-SOA	SVOCs-SOA	POA	ASOA	BSOA	IVOCs-SOA	SVOCs-SOA
July	Northeast	0.8	2.1	3.5	2.0	0.2	0.5	0.6	4.0	1.8	0.2
	NCP	1.6	5.7	2.8	4.8	0.3	0.9	1.4	3.6	4.0	0.4
	YRD	1.1	2.9	4.9	2.6	0.3	0.7	0.8	5.8	2.1	0.3
	PRD	0.5	1.1	4.0	0.8	0.2	0.3	0.4	4.8	0.8	0.2
	Central China	1.3	1.7	10.8	2.2	0.4	1.0	0.8	14.8	1.8	0.5
	Sichuan Basin	1.9	1.8	4.8	2.5	0.5	1.2	0.8	5.3	2.1	0.4
November	Northeast	6.5	0.5	0.1	1.2	0.2	6.7	0.7	0.8	1.4	0.2
	NCP	5.8	1.9	0.3	4.3	0.4	5.8	1.5	1.8	3.9	0.4
	YRD	3.8	4.1	0.9	5.6	0.3	3.7	2.4	3.3	5.0	0.4
	PRD	1.7	3.7	4.0	2.9	0.3	1.3	1.9	5.8	3.4	0.4
	Central China	3.1	4.2	2.0	5.4	0.4	2.8	2.1	4.1	5.1	0.4
	Sichuan Basin	4.3	3.5	1.4	3.8	0.3	3.3	1.5	2.4	3.0	0.3

Table S9 Molar yields (ppm/ppm) of different precursors under high- and low-NOx conditions in CAMx and CMAQ (values are obtained directly from the source code)

Model	Precursor	High NOx yields under different C^* ($\mu g/m^3$)					
		0.01	1	10	100	1000	Sum
CAMx	Benzene	0	0.001	0.079	0.148	0.222	0.450
	Toluene	0	0.006	0.145	0.281	0.432	0.864
	Xylene	0	0.001	0.127	0.201	0.301	0.630
CMAQ	Benzene	0	0.034	0	0.392	0	0.426
	Toluene	0	0.016	0.051	0.047	0	0.114
	Xylene	0	0.015	0.023	0.06	0	0.098
Model	Precursor	Low NOx yields under different C^* ($\mu g/m^3$)					
		0.01	1	10	100	1000	Sum
CAMx	Benzene	0	0.035	0.108	0.185	0.268	0.596
	Toluene	0	0.006	0.145	0.437	0.281	0.869
	Xylene	0	0.048	0.195	0.252	0.364	0.859
CMAQ	Benzene	0.146	0	0	0	0	0.146
	Toluene	0.14	0	0	0	0	0.140
	Xylene	0.193	0	0	0	0	0.193

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