



*Supplement of*

## **The atmospheric oxidizing capacity in China – Part 1: Roles of different photochemical processes**

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**Model validation.** Figure S1 shows the spatial distribution of the calculated monthly average surface maximum daily 8-hour average (MDA8) ozone, as well as the monthly mean concentrations of NO<sub>2</sub>, CO, and PM<sub>2.5</sub>. A statistical evaluation of the surface concentration of these air pollutants is provided in Table S3. Our model matches well with the spatial distribution of MDA8 ozone (Figure S1 a, b) in both January and July. High values of the correlation coefficient ( $R > 0.85$ ) and low values of the mean bias (MB) are indicative of good model performance. An overestimation of ozone is simulated in central and western China in July, which is related to the underestimation of NO<sub>2</sub> in these areas (Figure S1 c, d). A comparison between the diurnal variations of the model and measured mixing ratios of NO<sub>2</sub> (Figure S2 a) and ozone (Figure S2 b) is provided in four large metropolitan areas for January and July. The agreement is generally good in the case of NO<sub>2</sub>. The model, however, tends to overestimate the NO<sub>2</sub> concentrations during winter, specifically in Beijing and Shanghai. In the case of ozone, the agreement is rather good in the summer, but the model underestimates the urban concentrations during winter. An underestimation of the O<sub>3</sub>/NO<sub>2</sub> ratio indicates an underestimation of the NO<sub>2</sub> photolysis or an overestimation of the HO<sub>2</sub> concentration.

As shown in Figure S1e-h, both CO and PM<sub>2.5</sub> are overestimated in the urban areas of northern and central China. This bias can be partially explained by the uncertainties in the adopted emissions and chemicals boundary (Dai et al., 2020).

Surface mixing ratios for NO, HONO, and formaldehyde (HCHO) are displayed in Figure S4 and the diurnal variation of calculated concentrations of NO and HONO in four urban and four remote sites are shown in Figures S7 and S8. The spatial distribution of NO and HONO have a similar pattern, with high concentration values found in the northern urban areas, especially during winter. The HONO concentration is highest during nighttime, which is opposite to the case of NO, whose concentration is maximum during daytime. The maximum values of the calculated HONO concentrations are found in urban and rural areas within the range of 2.4 - 3.00 ppbv and 0.75 - 3.00 ppbv in January and July, respectively (Figure S8). A comparison between the values of HONO derived from local observations is compared with model estimates in Table S4. High values of surface HCHO are derived in the southern part of China, especially in summer. This region with high values of solar radiation and temperature exhibits high concentrations of isoprene (Figure S6b), which favors the formation of HCHO (Wang et al., 2021). Our calculated concentration of HCHO is consistent with the measured value of 6.5 ppbv in the summer at Chengdu (Yang et al., 2021) and of 5.8 - 6.8 and 4.9 ppbv in the Fall at Heshan (Tan et al., 2017) and Shenzhen (Yang et al., 2022), respectively.

The surface mixing ratios of OH, HO<sub>2</sub>, and NO<sub>3</sub> are in Figure S6. During winter, the high concentrations of OH are distributed over the South China Sea and along the southern coast of the country; in summer, the highest values are found in the North China Plain area and along ship tracks in the East China Sea. These distributions are partially relevant to the seasonal variations of water vapor and solar radiation and the distribution of concentrations of isoprene (Figure S5), NO (Figure S4

a, b), and HONO (Figure S4 c,d). In Beijing, the peak concentrations of calculated OH and HO<sub>2</sub> are  $1.5 \times 10^6 \text{ cm}^{-3}$  and  $1.0 \times 10^7 \text{ cm}^{-3}$  in winter and  $1.2 \times 10^7 \text{ cm}^{-3}$  and  $1.0 \times 10^9 \text{ cm}^{-3}$  in summer (Figure S10 and S9), respectively. These values are consistent with the observed peak concentrations of OH and HO<sub>2</sub> of  $1.5 \times 10^6 - 2.0 \times 10^6 \text{ cm}^{-3}$  and of  $3.0 \times 10^7 - 4.0 \times 10^7 \text{ cm}^{-3}$  in winter (Ma et al., 2019) and of  $8.0 \times 10^6 - 9.0 \times 10^6 \text{ cm}^{-3}$  and  $3 \times 10^8 - 15 \times 10^8 \text{ cm}^{-3}$  in summer (Yang et al., 2021) reported in this area. In Guangzhou sites, the maximum concentration of OH and HO<sub>2</sub> is  $10.0 \times 10^6 \text{ cm}^{-3}$  and  $7.0 \times 10^8 \text{ cm}^{-3}$  in summer, which is slightly higher than the peak value of  $4.5 \times 10^6 \text{ cm}^{-3}$  and  $4.0 \times 10^8 \text{ cm}^{-3}$  measured in autumn at Shenzhen (Yang et al., 2022). In Chengdu, the maximum concentrations of OH and HO<sub>2</sub> measured during summertime are  $1.0 \times 10^7 \text{ cm}^{-3}$  and  $1.0 \times 10^9 \text{ cm}^{-3}$ , respectively, which is also consistent with our calculated level ( $1.5 \times 10^7 \text{ cm}^{-3}$  and  $0.8 \times 10^9 \text{ cm}^{-3}$ ) (Yang et al., 2021).

As NO<sub>3</sub> can be rapidly photolyzed during the daytime, high concentrations of this radical are found during nighttime. Nitrate is formed from the reaction between NO<sub>2</sub> and O<sub>3</sub> (Brown and Stutz, 2012). The calculated concentrations of NO<sub>3</sub> radical are highest in January (Figure S6 e), specifically in northern China, and in July along the ship tracks in the sea near the coast of southern China. The highest calculated concentrations of nighttime NO<sub>3</sub> are found at the Wangdu site ( $1.2 \times 10^9 \text{ cm}^{-3}$ ) and at the Hok Tsui site ( $4.5 \times 10^8 \text{ cm}^{-3}$ ) in January and July (Figure S11), respectively. There is only a very small number of NO<sub>3</sub> measurements that are currently reported in China. However, Wang et al., (2023) have recently highlighted the increasing role of the NO<sub>3</sub> radical for the nighttime oxidation capacity in China, specifically during the 2014 – 2019 period.

The spatial distribution of the aerosol surface area density, shown in Figure S12, is an important parameter determining the rate at which heterogeneous reactions proceed. In the eastern plain of China, the calculated values are substantially higher in January by  $1 \times 10^{-5} - 2 \times 10^{-5} \text{ cm}^2/\text{cm}^3$  than in June by  $0.2 \times 10^{-5} - 0.5 \times 10^{-5} \text{ cm}^2/\text{cm}^3$ , which is consistent with the observed seasonal variation of the aerosol burden. The calculations of this parameter are based on the concentration of aerosol calculated in our model (Zaveri et al., 2008). Since the calculated PM<sub>2.5</sub> concentration appears to be slightly overestimated, the aerosol surface area density used to calculate the aerosol uptake may be overestimated.

Table S1. List of peroxy radicals (RO<sub>2</sub>), volatile organic compounds (VOCs), and aerosol species in the MOZART-MOSAIC mechanism in the WRF-Chem model.

		Species
RO <sub>2</sub> (Emmons et al., 2010)		CH <sub>3</sub> O <sub>2</sub> , EO <sub>2</sub> , C <sub>2</sub> H <sub>5</sub> O <sub>2</sub> , PO <sub>2</sub> , CH <sub>3</sub> CO <sub>3</sub> , C <sub>3</sub> H <sub>7</sub> O <sub>2</sub> , RO <sub>2</sub> , ENEO <sub>2</sub> , ALKO <sub>2</sub> , MEKO <sub>2</sub> , ISOPO <sub>2</sub> , MACRO <sub>2</sub> , MCO <sub>3</sub> , PHENO <sub>2</sub> , C <sub>6</sub> H <sub>5</sub> O <sub>2</sub> , BENZO <sub>2</sub> , MALO <sub>2</sub> , BZOO, ACBZO <sub>2</sub> , TOLO <sub>2</sub> , DICARBO <sub>2</sub> , MDIALO <sub>2</sub> , XYLOLO <sub>2</sub> , XYLENO <sub>2</sub> , TERPO <sub>2</sub> , TERP <sub>2</sub> O <sub>2</sub> , NTERPO <sub>2</sub> , XO <sub>2</sub> , MBOO <sub>2</sub> , HMPROPO <sub>2</sub> , MBONO <sub>3</sub> O <sub>2</sub>
VOCs (Emmons et al., 2010)	Alkanes	C <sub>2</sub> H <sub>6</sub> , C <sub>3</sub> H <sub>8</sub> , BIGALK
	Alkenes	C <sub>2</sub> H <sub>4</sub> , C <sub>3</sub> H <sub>6</sub> , BIGENE, ISOPR, APIN, BPIN, LIMON
	Aromatics	BENZENE, TOLUNE, XYLENE, CRESOL
	Alkyne	C <sub>2</sub> H <sub>2</sub>
	Oxidized VOCs	HCHO, CH <sub>3</sub> CHO, POOH, CH <sub>3</sub> COOOH, PAN, MPAN, MACR, MVK, C <sub>2</sub> H <sub>5</sub> OOH, C <sub>3</sub> H <sub>7</sub> OOH, ROOH, CH <sub>3</sub> COCH <sub>3</sub> , CH <sub>3</sub> COCHO, XOOH, ONIT, ONITR, ISO-POOH, HYAC, GLYALD, MEK, BIGALD, GLYOXAL, ALKOOH, MEKOOH, TOLOOH, PHENOL, BEPOMUC, TEPOMUC, BIGALD1, BIGALD2, BIGALD3, BIGALD4, MBOOOH, HMPROP, TERPROD1, TERPROD2, TER-POOH, CH <sub>3</sub> OOH, MACROOH, PHENOOH, C <sub>6</sub> H <sub>5</sub> OOH, BENZOOH, BZOOH, BZALD, XYLOLOOH, XYLENOOH, TERP <sub>2</sub> OOH, CH <sub>3</sub> COOH, HYDRALD, CH <sub>3</sub> OH, C <sub>2</sub> H <sub>5</sub> OH, HCOOH, HOCH <sub>2</sub> OO
Others	CH <sub>4</sub>	
Aerosol species (Zaveri et al., 2008)		NA <sup>+</sup> , NH <sub>4</sub> <sup>+</sup> , Cl <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , NO <sub>3</sub> <sup>-</sup> , OIN, OC, BC, SOA

Table S2. Physical modules used in model simulations

Atmospheric process	Scheme
Cloud microphysics	Morrison double moment (Morrison et al., 2005)
Cumulus parameterization	Grell 3D Ensemble Scheme (Grell and Dévényi., 2002)
Land-surface physics	Noah Land Surface Model (Chen and Dudhia., 2001)
Longwave radiation	RRTM scheme (Mlawer et al., 1997)
Shortwave radiation	RRTM scheme (Mlawer et al., 1997)
Planetary boundary layer	Yonsei University PBL (Hong et al., 2006)
Photolysis	Madronich Fast Tropospheric Ultraviolet-Visible (FTUV) (Madronich et al., 1987., Tie et al., 2003)

Table S3. Statistical analysis of key quantities that characterize the surface concentrations of air pollutants averaged over the entire geographical area of China for January and July.

	SIM		OBS		Bias		R		NMB		NME		RMSE	
	Jan.	July	Jan.	July	Jan.	July	Jan.	July	Jan.	July	Jan.	July	Jan.	July
Ozone ( $\mu\text{g}/\text{m}^3$ )	47.8	76.4	41.3	72.5	6.5	3.6	0.9	0.9	0.2	0.1	0.2	0.1	6.5	3.6
NO <sub>2</sub> ( $\mu\text{g}/\text{m}^3$ )	19.5	12.4	29.1	13.9	-9.6	-1.4	0.5	0.9	-0.3	-0.1	0.3	0.2	9.6	1.4
CO (10 $\mu\text{g}/\text{m}^3$ )	61.9	30.7	48.7	28.2	13.1	2.5	0.8	0.7	0.3	0.1	0.3	0.1	13.1	2.5
PM <sub>2.5</sub> ( $\mu\text{g}/\text{m}^3$ )	80.9	32.6	65.7	24.6	15.1	8.1	0.7	0.3	0.2	0.3	0.3	0.3	15.1	8.1

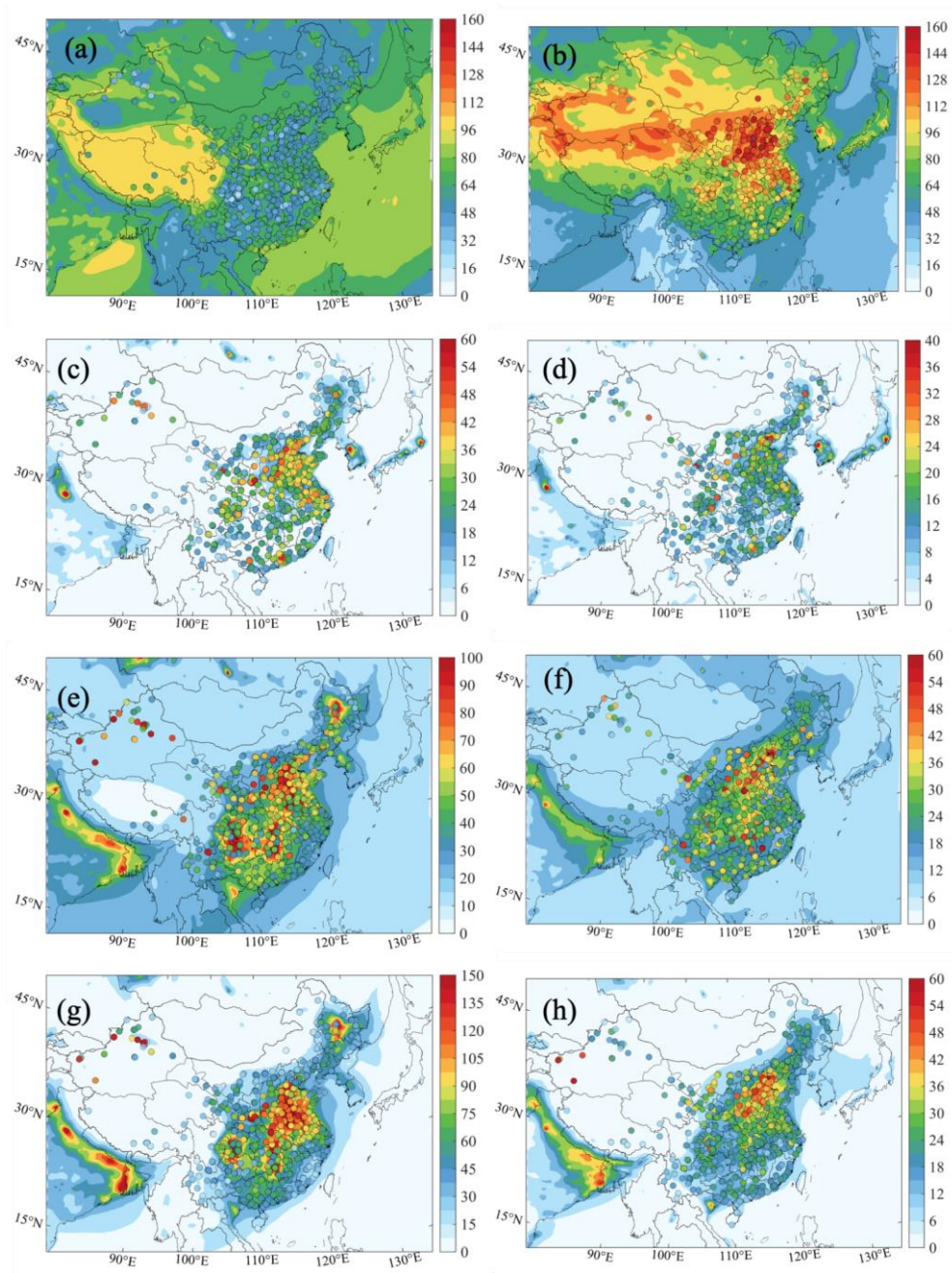
SIM and OBS represent the average of calculated and measured concentrations of chemicals [ $\mu\text{g}/\text{m}^3$ ]. Bias is the mean bias [ $\mu\text{g}/\text{m}^3$ ]; *R* is the correlation coefficient (unitless); NMB is the normalized mean bias (unitless); NME is the normalized mean error (unitless); RMSE is the root mean square error (unitless)

Table S4. Comparison between values of the maximum and average HONO mixing ratio [Units: ppbv] derived from local observations and calculated by our regional model.

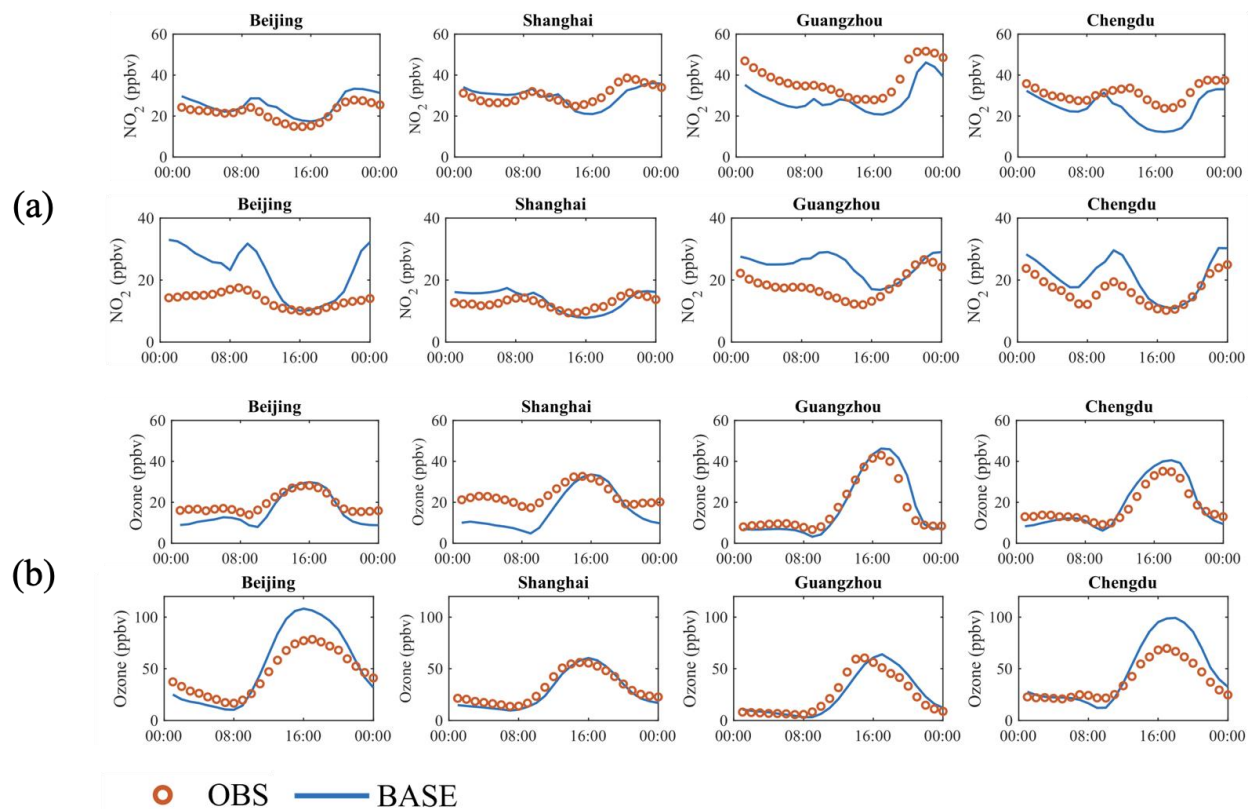
Location	Period	Maximum	Mean $\pm$ SD	References
Beijing (urban)	January 2016	-	1.05 $\pm$ 0.89	Wang et al., (2017)
	June-July 2016	-	1.38 $\pm$ 0.90	Wang et al., (2017)
	October 2018	2.50	-	Zhang et al., (2022)
	January 2018	3.00	1.30	This study
	July 2018	2.4	0.89	This study
Shanghai (urban)	Oct. 2004-Jan. 2005	-	1.10 $\pm$ 1.0	Cui et al., (2018)
	January 2018	2.40	1.10	This study
	July 2018	0.75	0.55	This study
Guangzhou (urban)	July 2016	-	1.03	Yang et al., (2017)
	Sep.-Nov. 2018	1.50	0.74 $\pm$ 0.70	Yu et al., (2022)
	January 2018	3.00	1.20	This study
	July 2018	1.95	1.05	This study
Chengdu (urban)	Aug.-Sep. 2019	2.00	1.3	Yang et al., (2021)
	January 2018	2.60	1.15	This study
	July 2018	2.15	0.95	This study
Wangdu (rural)	June 2018	1.56	-	Liu et al., (2019)
	January 2018	3.12	1.40	This study
	July 2018	2.40	1.00	This study
Heshan (rural)	Oct.-Nov. 2014	1.40	-	Yu et al., (2022)
	January 2018	1.05	0.35	This study
	July 2018	0.35	0.15	This study

Hok Tsui (background)	Sep.-Dec. 2012	-	$0.13 \pm 0.09$	Zha et al., (2014)
	January 2018	0.14	0.06	This study
	July 2018	0.05	0.02	This study

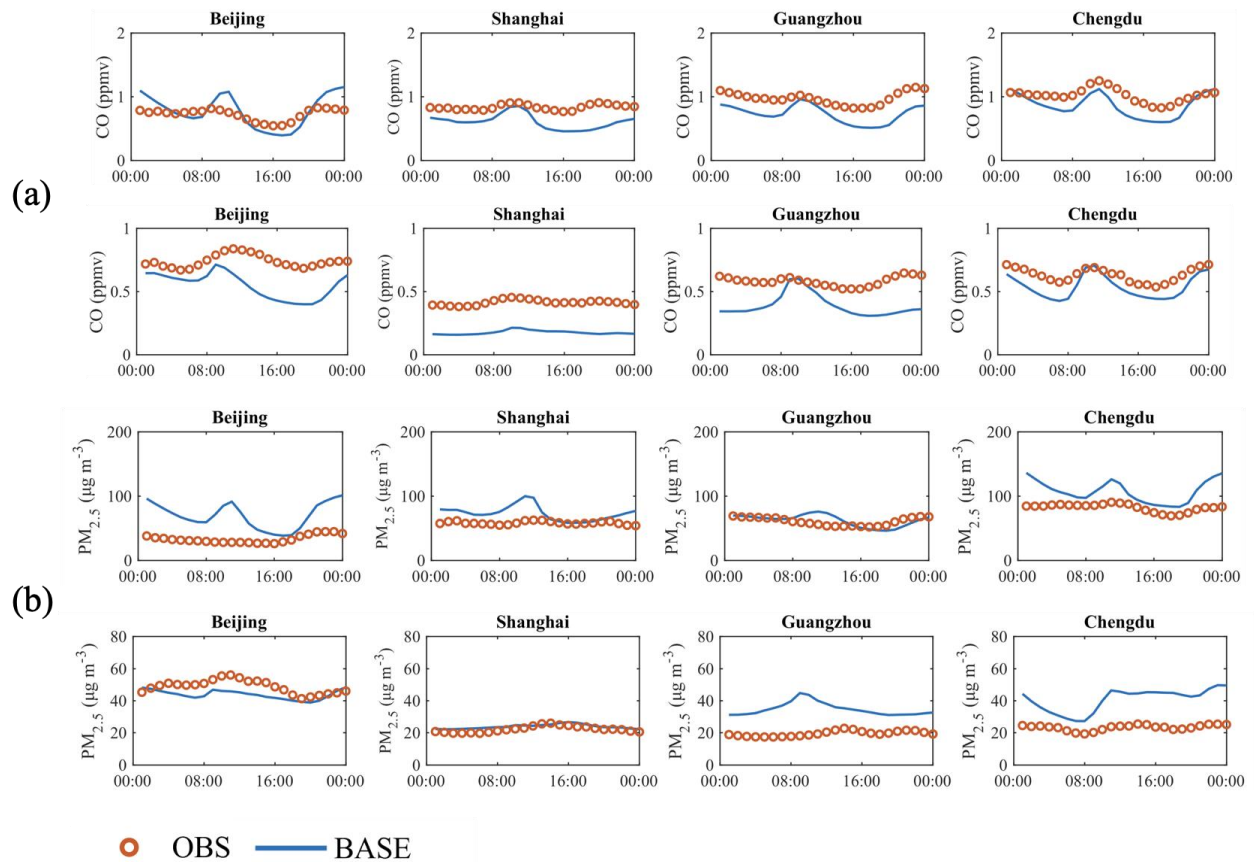




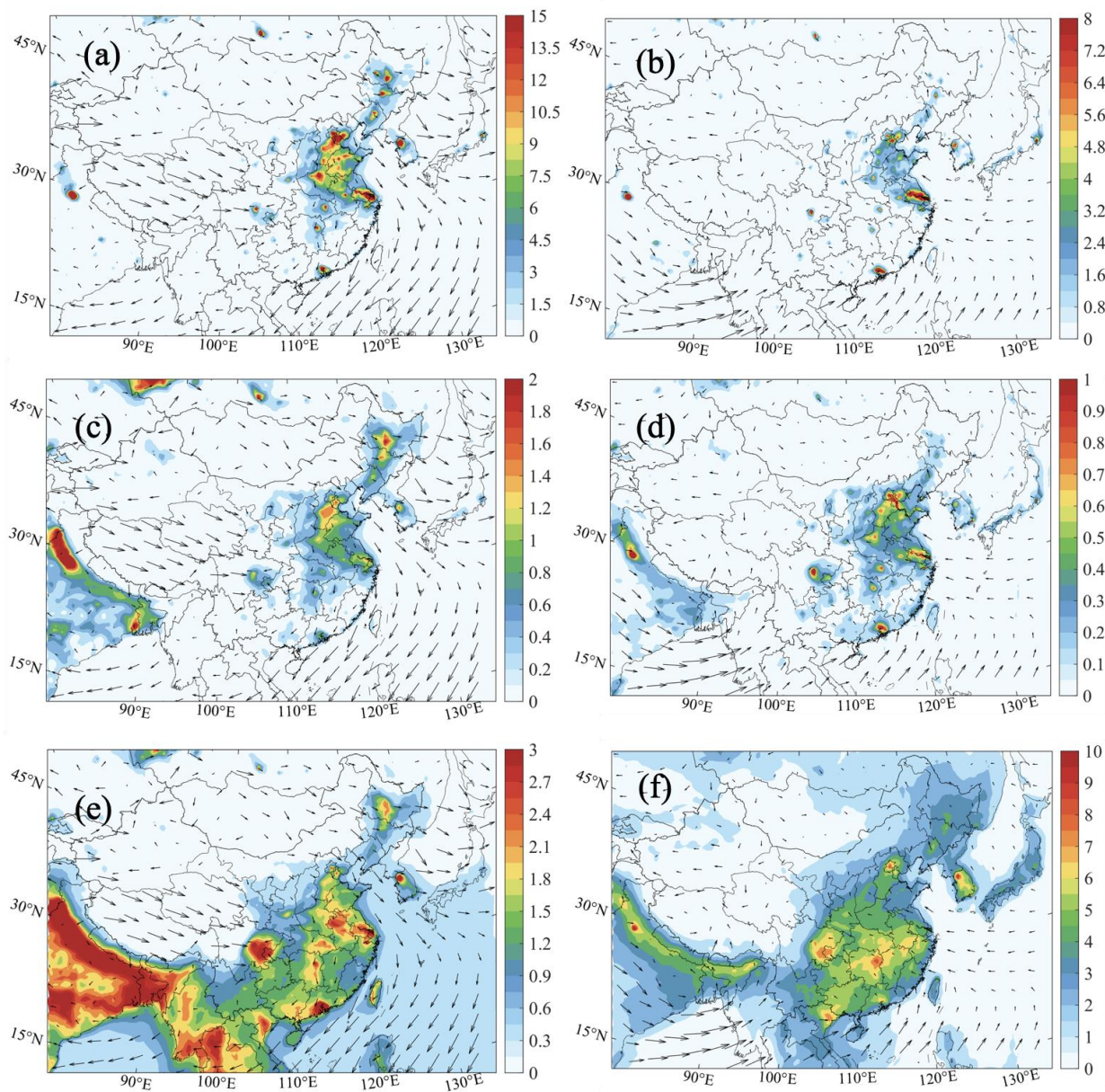
**Figure S1.** Spatial distribution of the simulated surface maximum daily 8-hour average (MDA8) ozone (a, b) [Unit:  $\mu\text{g m}^{-3}$ ], monthly average  $\text{NO}_2$  (c, d) [Unit:  $\mu\text{g m}^{-3}$ ], CO (e, f) [Unit:  $10 \mu\text{g m}^{-3}$ ], and  $\text{PM}_{2.5}$  (g, h) [Unit:  $\mu\text{g m}^{-3}$ ] concentrations. The left column panels are for January and the right column panels are for July 2018. In all cases, the units are  $\mu\text{g}/\text{m}^3$ . Circles with colors represent observed values at monitoring stations.



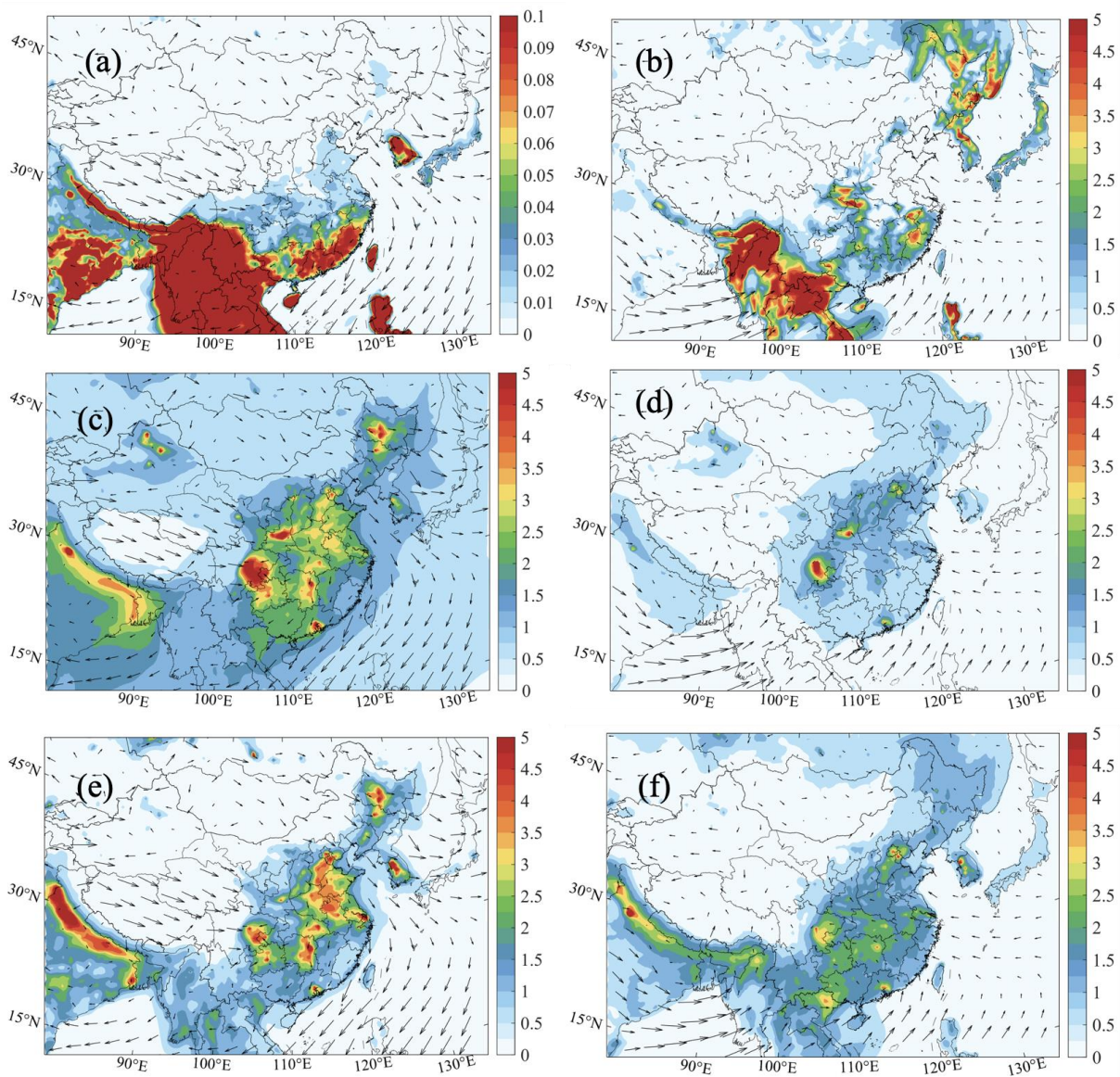
**Figure S2.** Diurnal variation of observed and simulated mixing ratio [Unit: ppbv] of NO<sub>2</sub> (a) and ozone (b) in four city sites in January (first and third rows) and July (second and fourth rows) of 2018.



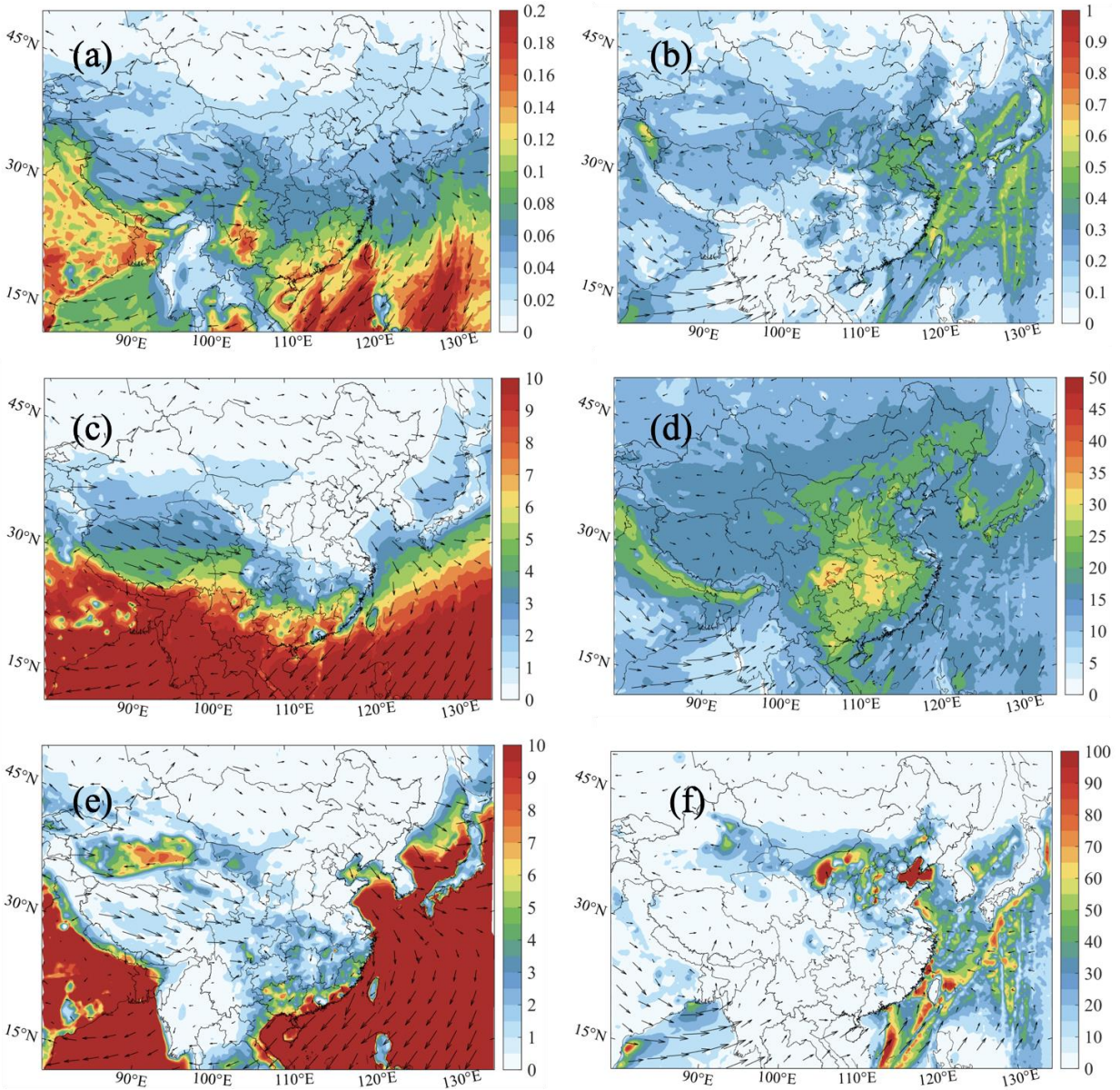
**Figure S3.** Diurnal variation of observed (circles in orange) and simulated (line in blue) mixing ratio of CO [Unit: ppmv] (a) and PM<sub>2.5</sub> [Unit: µg m<sup>-3</sup>](b) in four city sites in January (first and third rows) and July (second and fourth rows) of 2018.



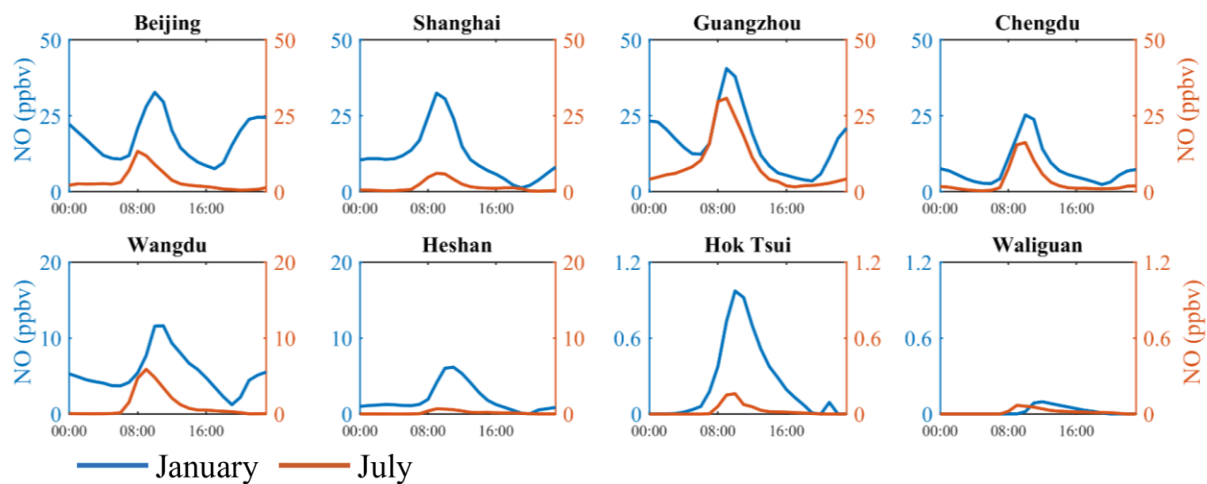
**Figure S4.** Spatial distribution of the simulated surface mixing ratio [Unit: ppbv] of NO (a, b), HONO (c, d), and HCHO (e, f) average for daytime (08:00 - 19:00 Local Standard Time (LST)) in January and July 2018.



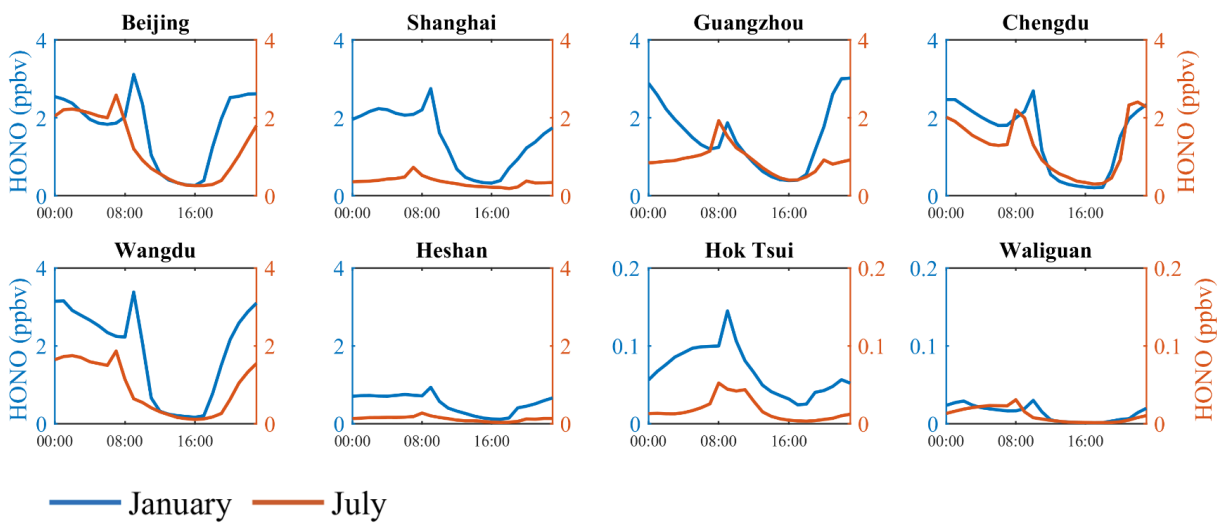
**Figure S5.** Spatial distribution of simulated mixing ratio [Units: ppbv] of isoprene (a, b), ethene (c, d), and ethane (e, f) average for daytime in January and July 2018.



**Figure S6.** Spatial distribution of simulated surface mixing ratio [Units: pptv] of OH (a, b) and HO<sub>2</sub> (c, d) averaged for daytime and NO<sub>3</sub> (e, f) averaged for nighttime (19:00 - 06:00 LST) in January and July of 2018.

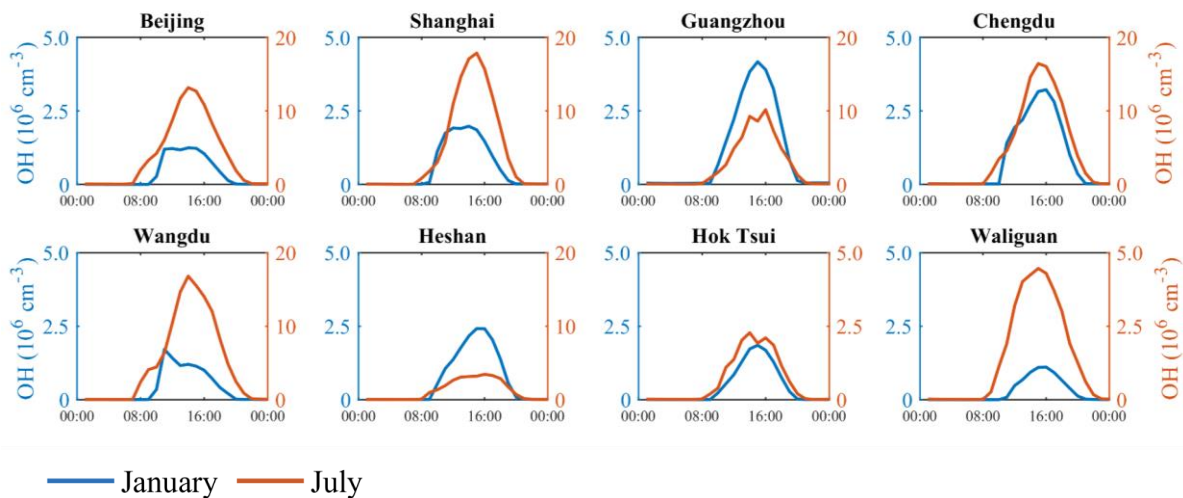


**Figure S7.** Diurnal variation of the simulated mixing ratio of NO [Unit: ppbv] in different sites in January (in blue, left axis) and July (in orange, right axis) of 2018.

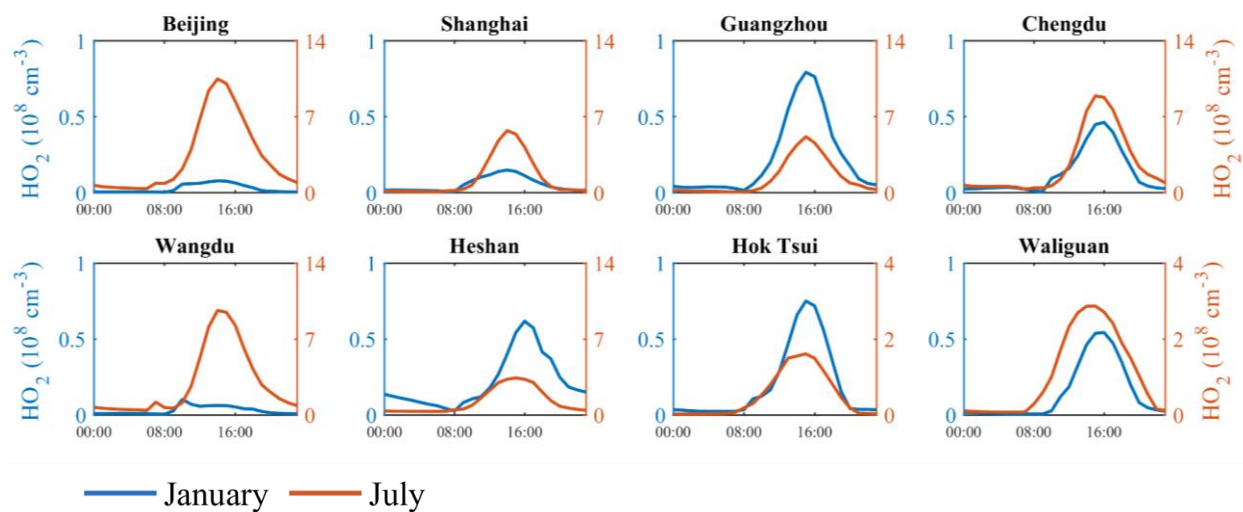


**Figure S8.** Diurnal variation of the simulated mixing ratio of HONO [Units: ppbv] in different sites in January (in blue, left axis) and July (in orange, right axis) of 2018.

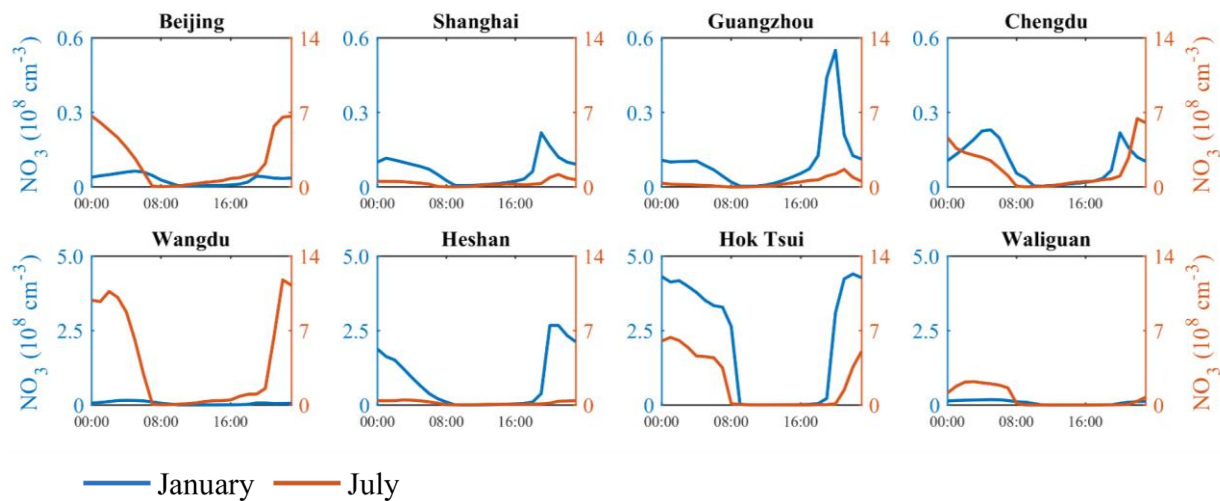




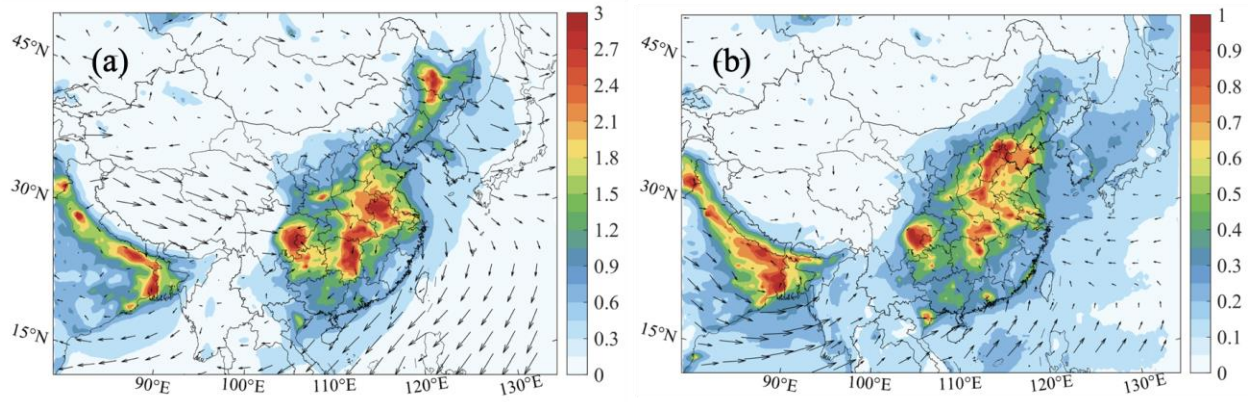
**Figure S9.** Diurnal variation of simulated mixing ratio [Unit:  $10^6 \text{ cm}^{-3}$ ] of OH radical in different sites in January (in blue, left axis) and July (in orange, right axis) of 2018.



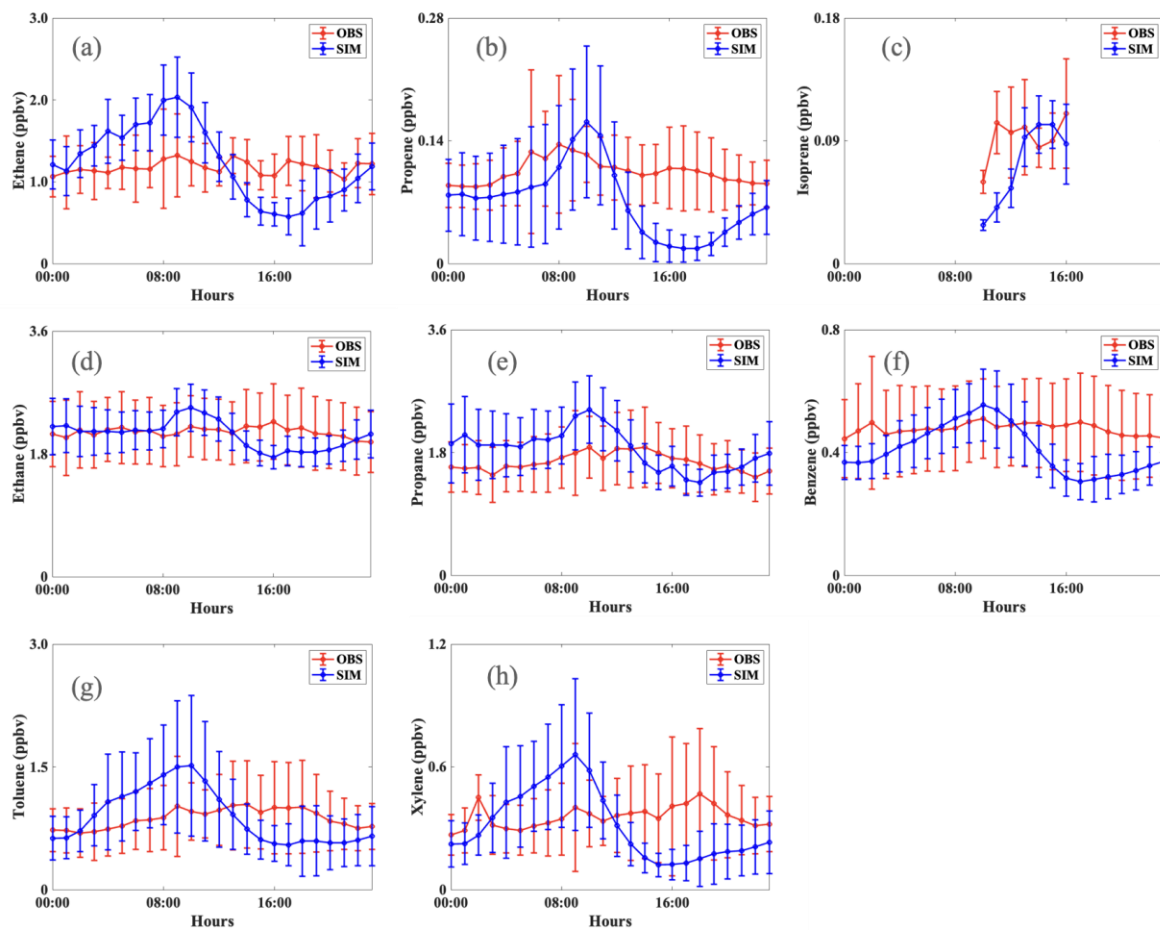
**Figure S10.** Diurnal variation of simulated mixing ratio [Unit:  $10^8 \text{ cm}^{-3}$ ] of HO<sub>2</sub> radical in different sites in January (in blue, left axis) and July (in orange, right axis) of 2018.



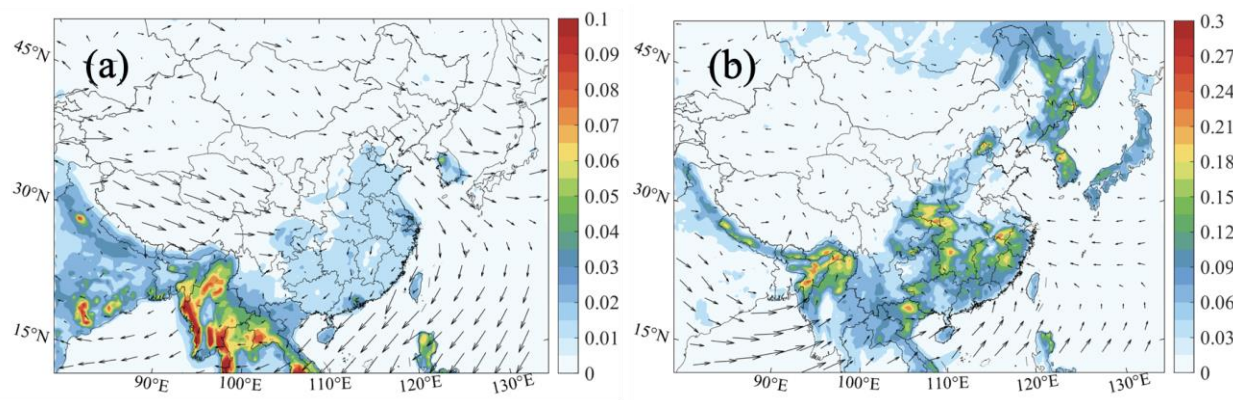
**Figure S11.** Diurnal variation of simulated mixing ratio [Unit:  $10^8 \text{ cm}^{-3}$ ] of  $\text{NO}_3$  radical in different sites in January (in blue, left axis) and July (in orange, right axis) of 2018.



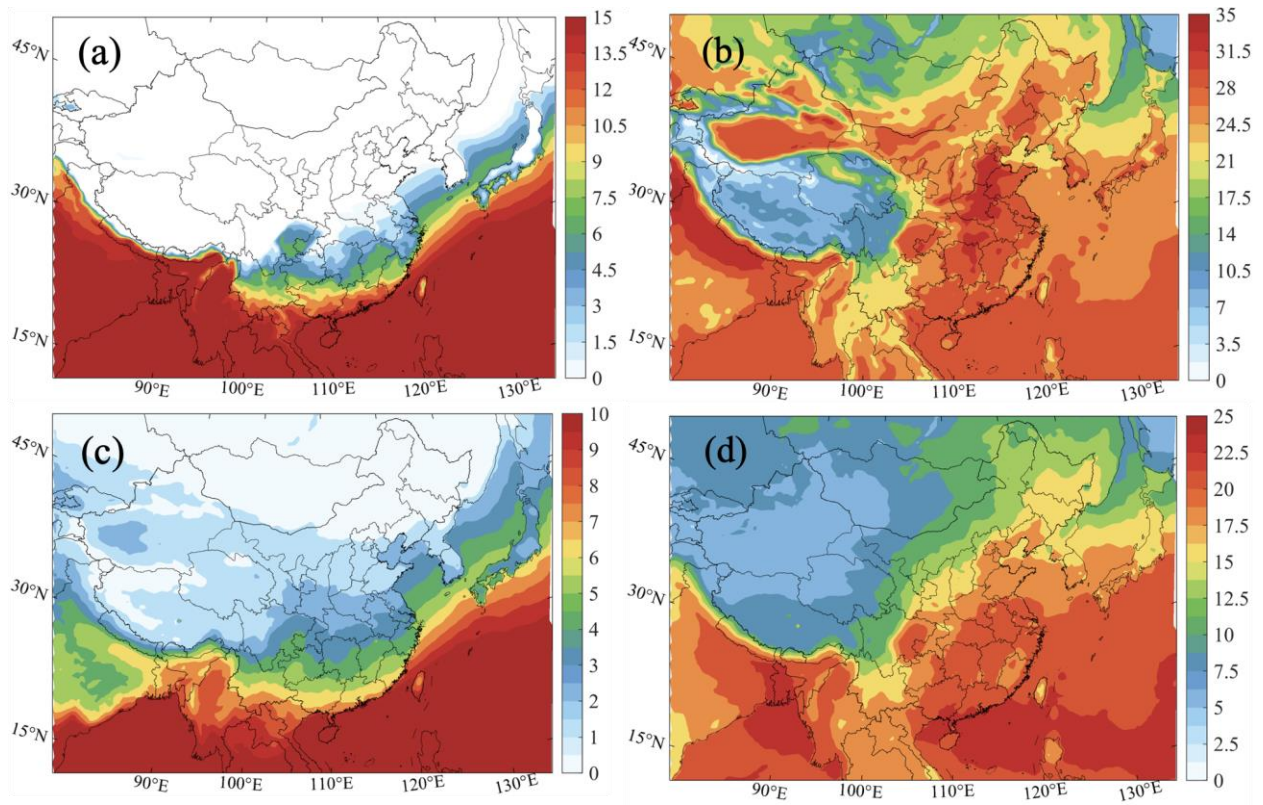
**Figure S12.** Spatial distribution of the monthly average simulated aerosol surface area density [Unit:  $10^{-5} \text{ cm}^2 \text{ cm}^{-3}$ ] average in January (a) and July (b), 2018.



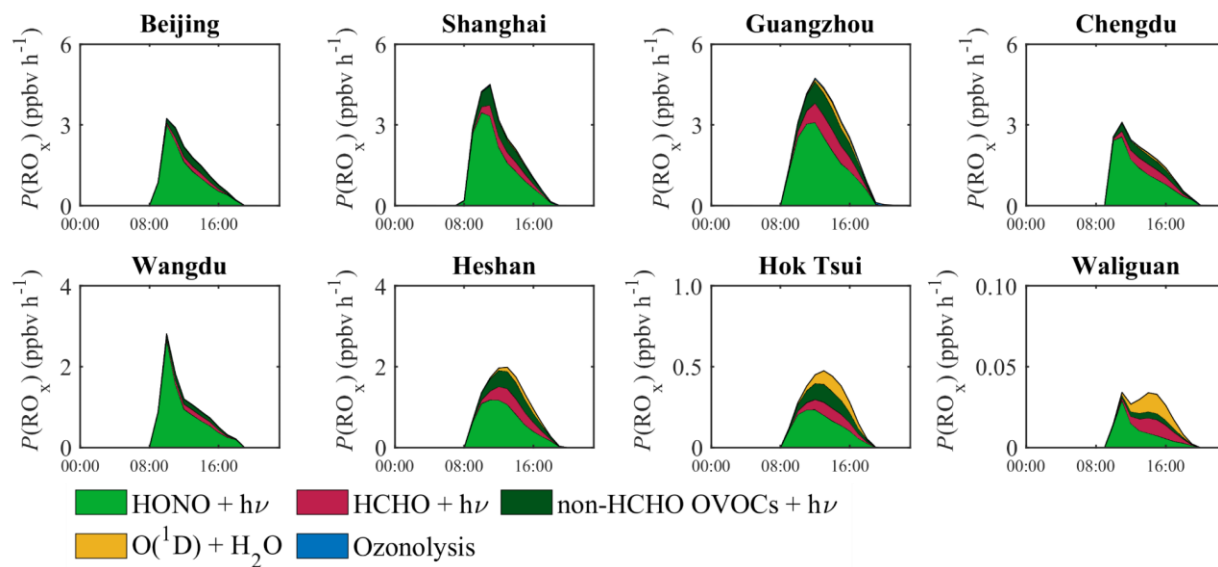
**Figure S13.** Diurnal variation of observed (OBS; in red) and simulated (SIM; in blue) mixing ratio [Unit: ppbv] of Ethene (a), Propene (b), Isoprene (c), Ethane (d), Propane (e), Benzene (f), Toluene (g) and Xylene (h) at the Hok Tsui site in January of 2018. Error bars represent the standard deviation.



**Figure S14.** Spatial distribution of production rate of  $RO_x$  ( $RO_2+HO_2+OH$ ) [ $P(RO_x)$ , Unit:  $ppbv\ h^{-1}$ ] (*Het-All* case) from the ozonolysis (Reactions between  $O_3$  and alkene) in the daytime (08:00-19:00 Local Standard Time (LST)) of January (a) and July (b) 2018.

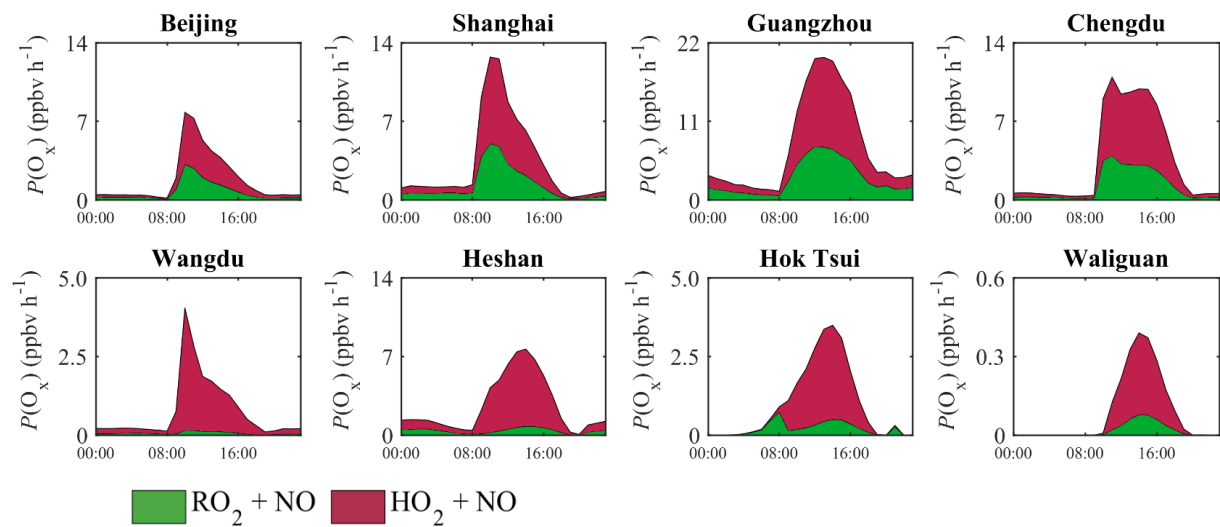


**Figure S15.** Spatial distribution of surface temperature [Unit: °C; a, b] and water vapor [Unit: g kg<sup>-1</sup>; c, d] in the daytime of January (a, c) and July (b, d) from the *Het-All* case.

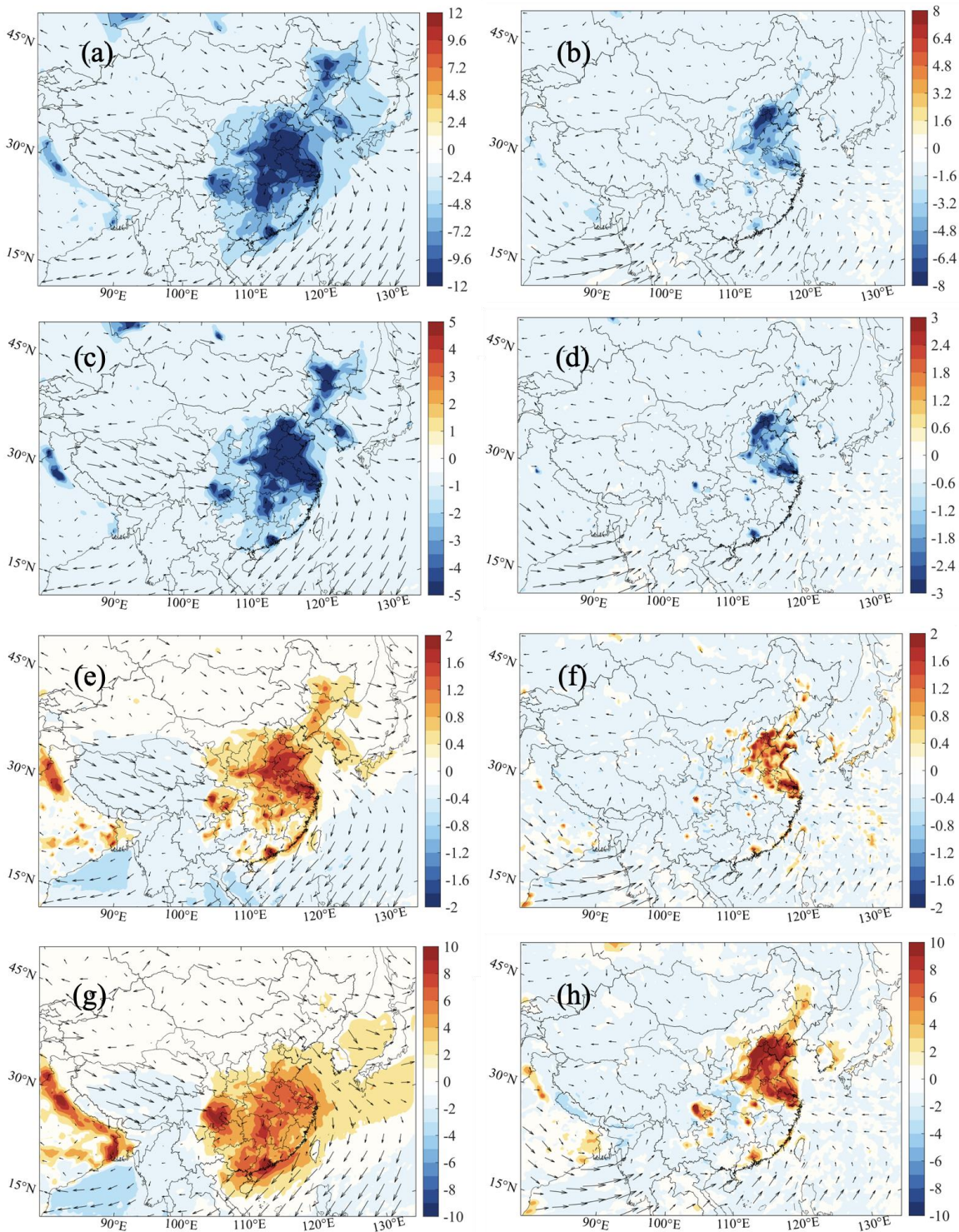


**Figure S16.** Diurnal variation of simulated production of  $RO_x$  (RO<sub>2</sub>+HO<sub>2</sub>+OH) [Unit: ppbv h<sup>-1</sup>] in cities and remote sites in January.

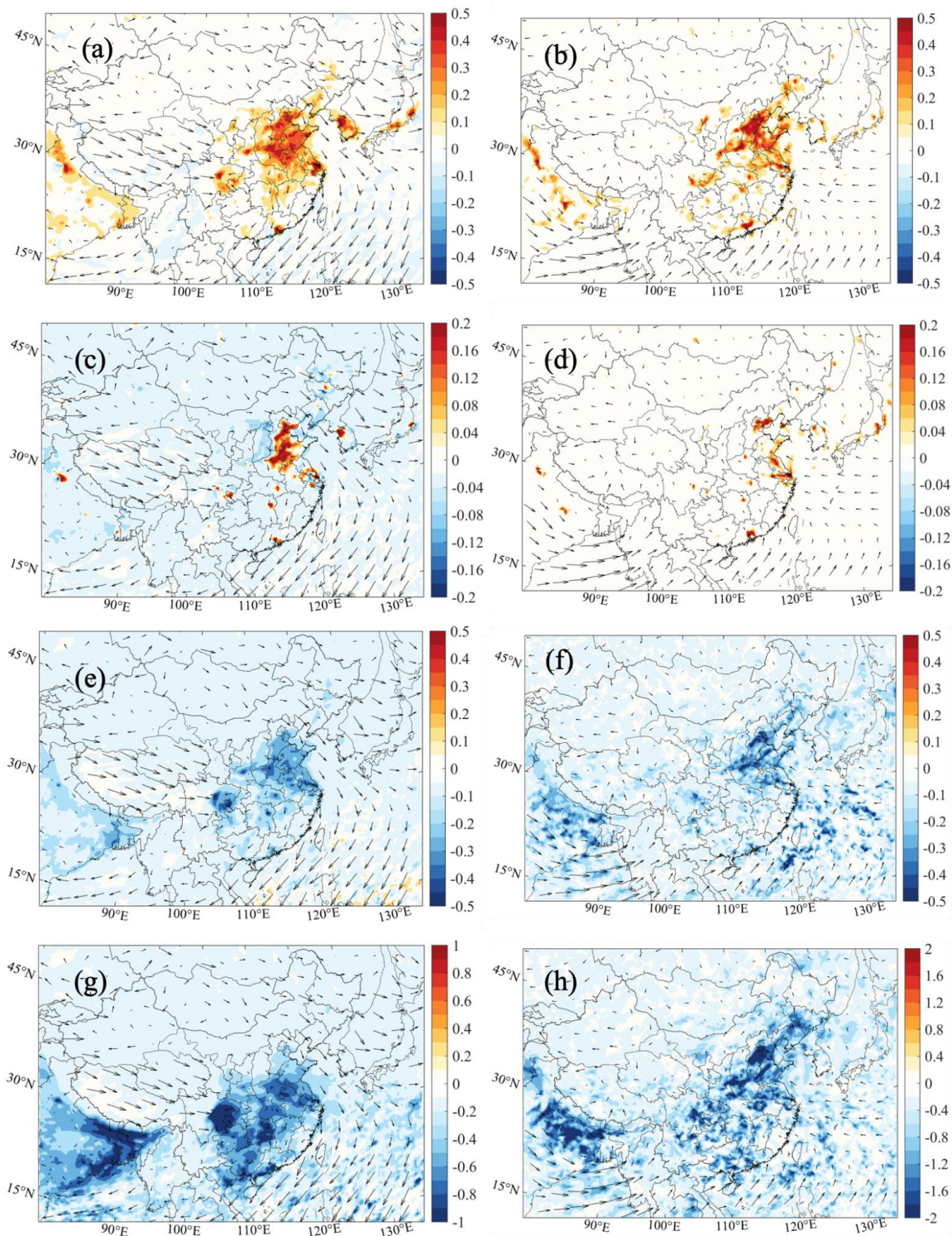




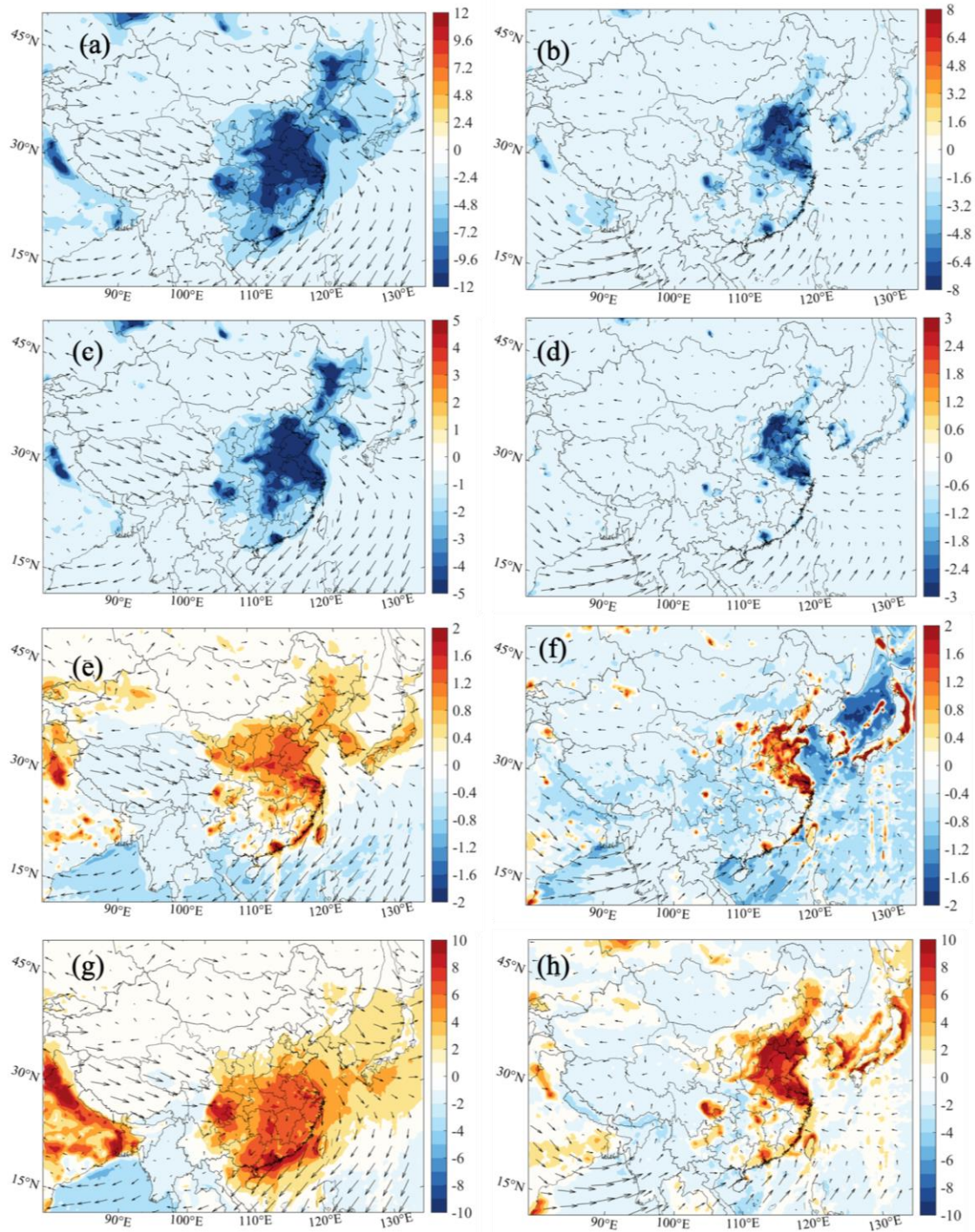
**Figure S17.** Diurnal variation of simulated  $O_x$  production rate [Unit:  $\text{ppbv h}^{-1}$ ] in different regions in January 2018.



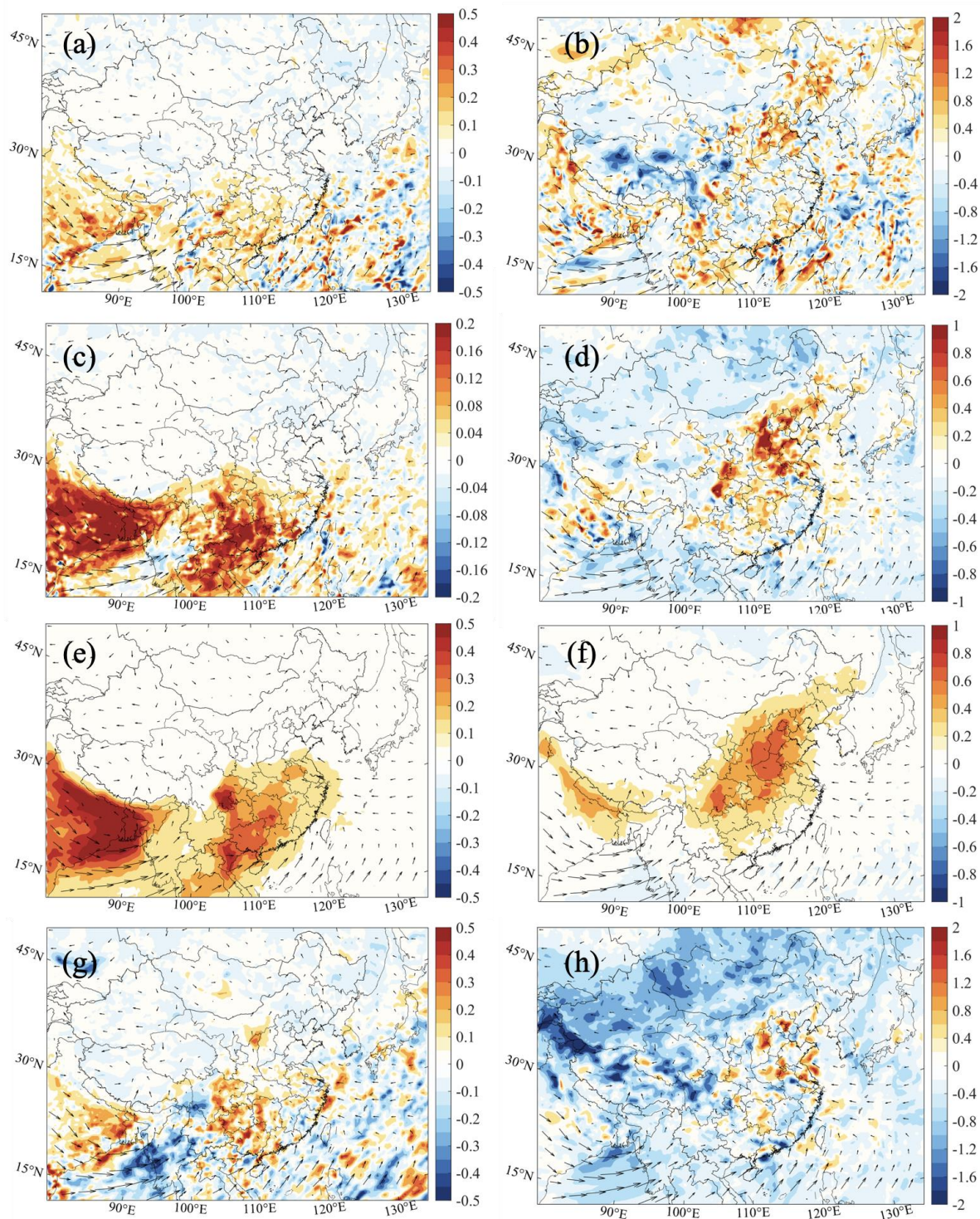
**Figure S18.** Changes in the surface mixing ratio of NO<sub>2</sub> (a, b) [Unit: ppbv], NO (c, d) [Unit: ppbv], OH (e, f) [Unit: 0.1 pptv], and HO<sub>2</sub> (g, h) [Unit: pptv] resulting from the introduction in the model of on heterogeneous chemical processes on aerosol particles in the daytime (06:00-19:00 LST) of January and July.



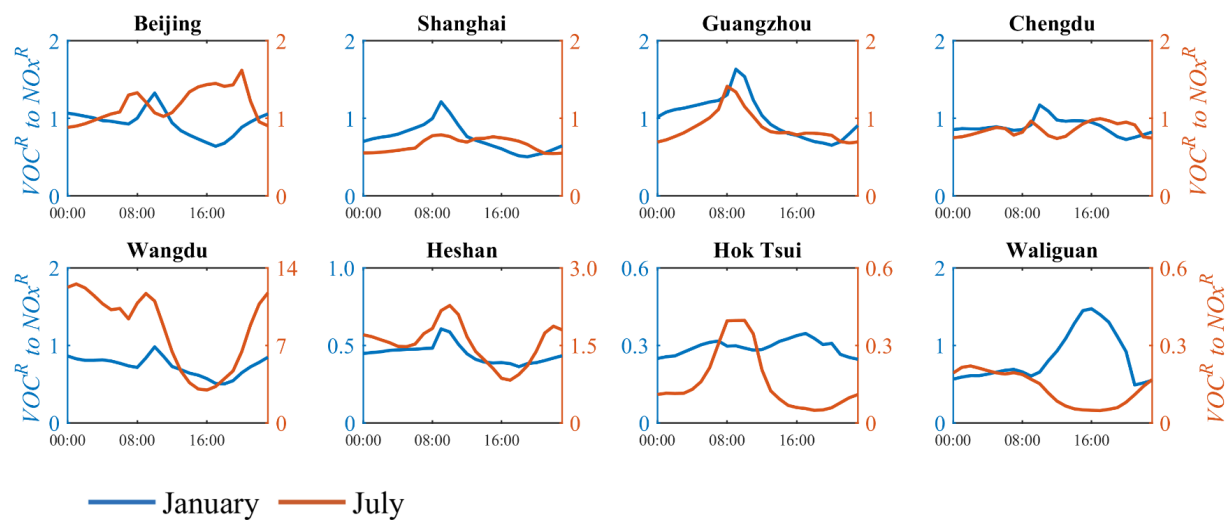
**Figure S19.** Changes in the surface mixing ratio of NO<sub>2</sub> (a, b) [Unit: ppbv], NO (c, d) [Unit: ppbv], OH (e, f) [Unit: 0.1 pptv] and HO<sub>2</sub> (g, h) [Unit: pptv] resulting from the introduction in the model of aerosol effects on photolysis (increased extinction of solar light) in the daytime (06:00-19:00 LST) of January and July.



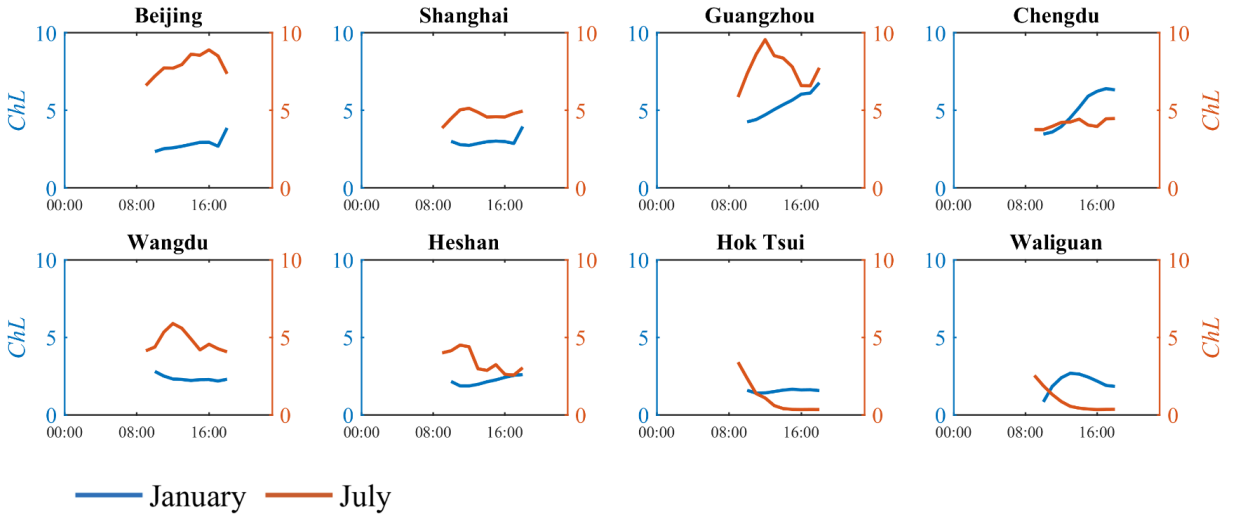
**Figure S20.** Changes in the surface mixing ratio of NO<sub>2</sub> (a, b) [Unit: ppbv], NO (c, d) [Unit: ppbv], OH (e, f) [Unit: 0.1pptv], and HO<sub>2</sub> (g, h) [Unit: pptv] resulting from the introduction in the model of combined aerosol effects (effect on radiation/photolysis and uptake on particles) in the daytime (06:00-19:00 LST) of January and July.



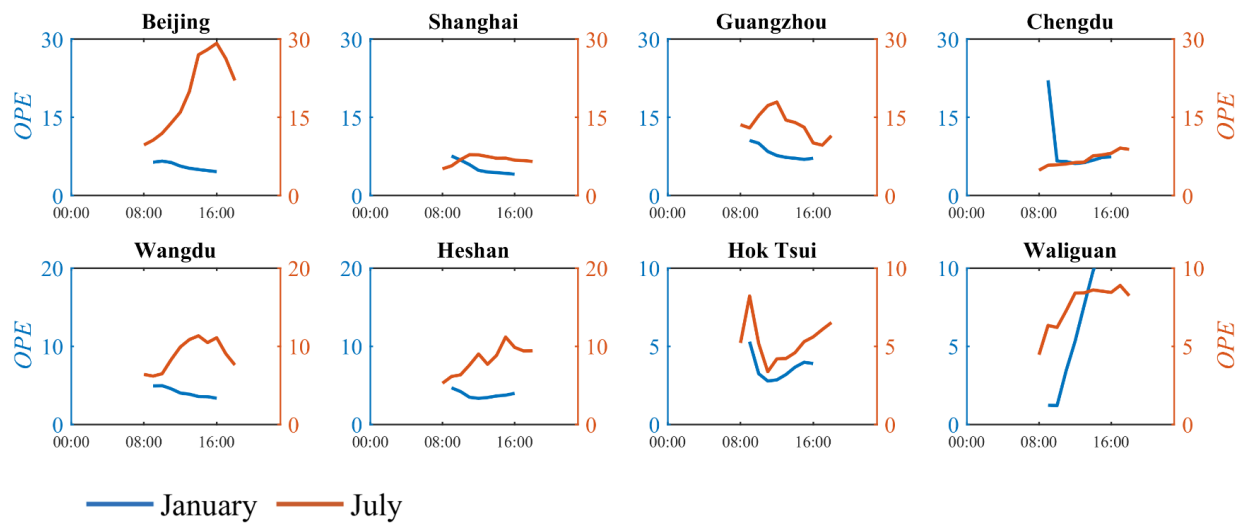
**Figure S21.** Changes in the surface mixing ratio of OH (a, b) [Unit: 0.1 pptv], HO<sub>2</sub> (c, d) [Unit: pptv], H<sub>2</sub>O<sub>2</sub> (e, f) [Unit: ppbv], and O<sub>3</sub> (g, h) [Unit: ppbv] resulting from the difference between the HO<sub>2</sub> uptake on aerosol producing H<sub>2</sub>O<sub>2</sub> and H<sub>2</sub>O in the daytime (06:00-19:00 LST) of January (left panels a, c, e, g) and July (right panels b, d, f, h) 2018.



**Figure S22.** Diurnal variation of the simulated value of the ratio of  $VOC^R$  and  $NO_x^R$  in different sites in January (in blue, left axis) and July (in orange, right axis) of 2018.

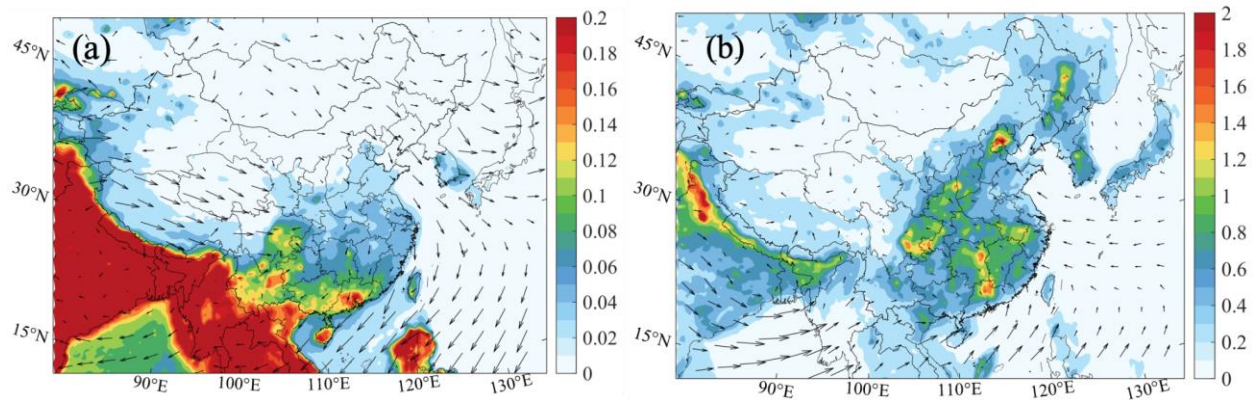


**Figure S23.** Diurnal variation of  $ChL$  in different sites in January (in blue, left axis) and July (in orange, right axis) of 2018.



**Figure S24.** Diurnal variation of  $OPE$  in different sites in January (in blue, left axis) and July (in orange, right axis) of 2018.





**Figure S25.** Spatial distribution of nighttime atmospheric oxidation capacity ( $AOC$  [Unit:  $10^7$  molecular  $\text{cm}^{-3} \text{s}^{-1}$ ]) in January (a) and July(b) of 2018 extracted from the *Het-All* case.

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