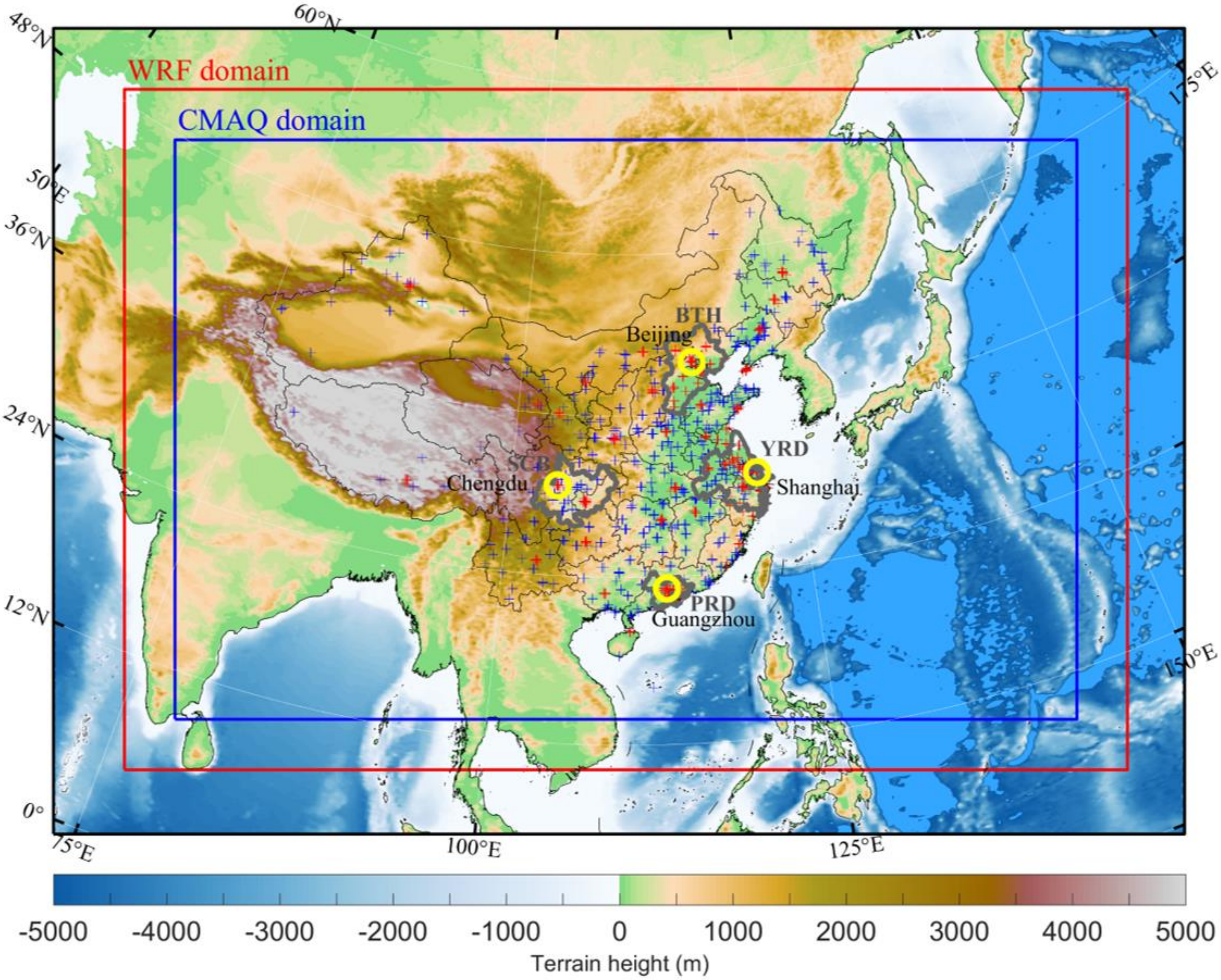

Figure S1 Model domains for the WRF (Red box) and CMAQ (Blue box) model. The red cross "+" represents the locations of environmental monitoring stations in 73 cities since 2013. The blue cross "+" represents the locations of environmental monitoring stations in other cities. The yellow circle denotes the locations of four major cities, Beijing, Shanghai, Guangzhou and Chengdu. The regions with bold gray boundary are the Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin (SCB) regions.

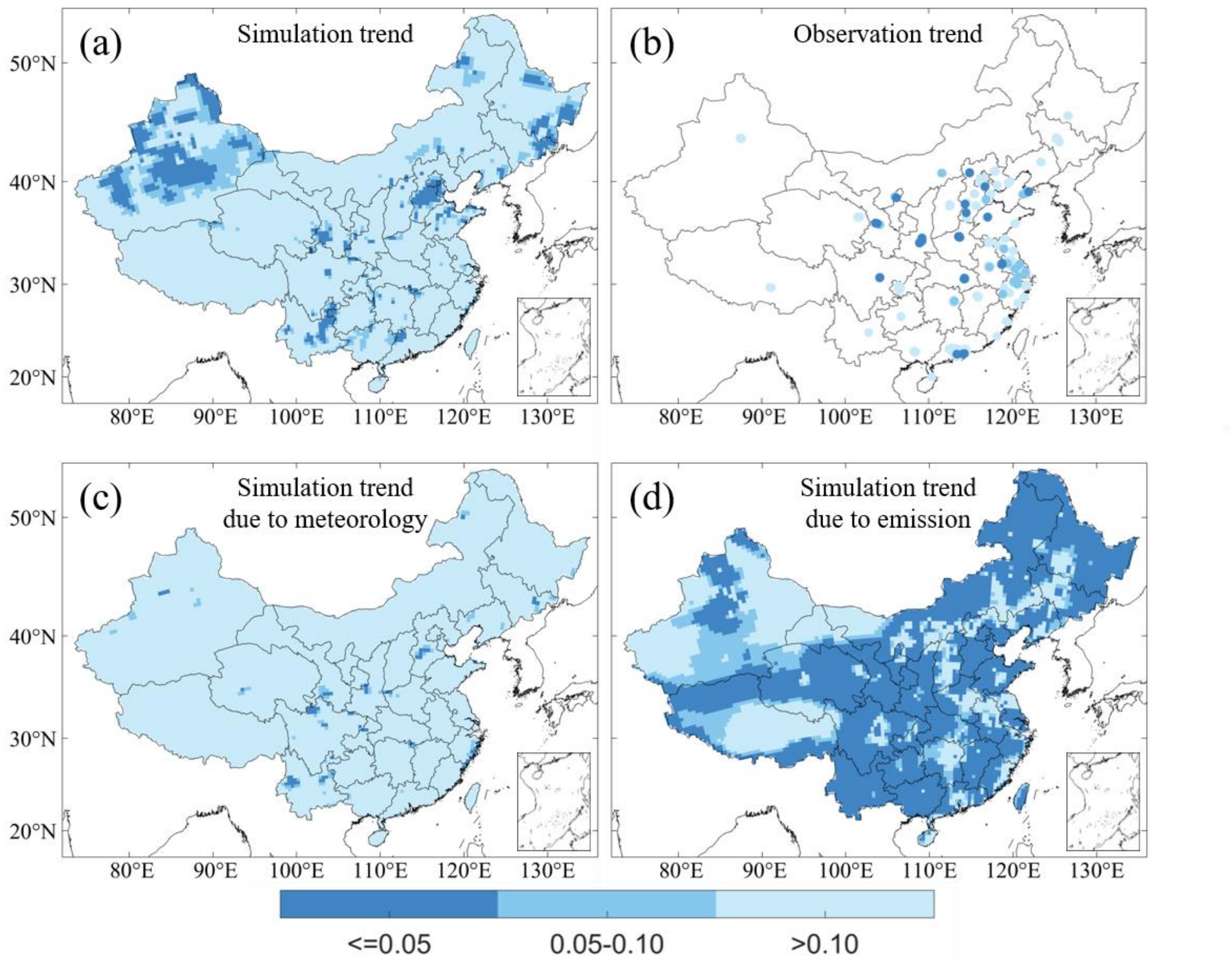

Figure S2 p values of regression for the rates of changes in the simulated (a) and observed (b) summer surface MDA8 O_3 concentrations, and those in the simulated concentrations due to variations in meteorological conditions (c) and anthropogenic emissions (d) across China from 2013 to 2017.

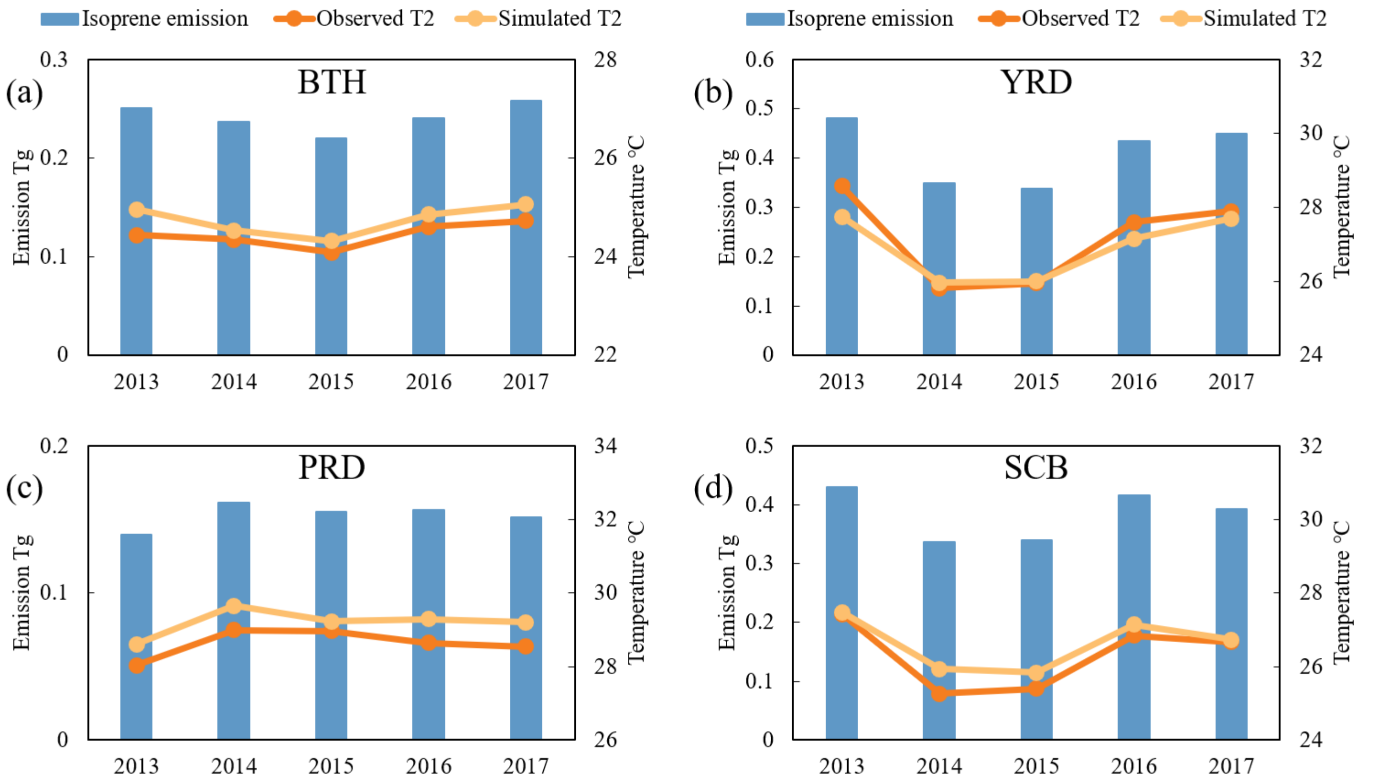

Figure S3 Variations in summer biogenic isoprene emissions, and observed and simulated temperature at a height of 2 m in the (a) Beijing-Tianjin-Hebei (BTH), (b) Yangtze River Delta (YRD), (c) Pearl River Delta (PRD), and (d) Sichuan Basin (SCB) regions from 2013 to 2017. The temperatures are averaged for the weather stations of the corresponding regions.

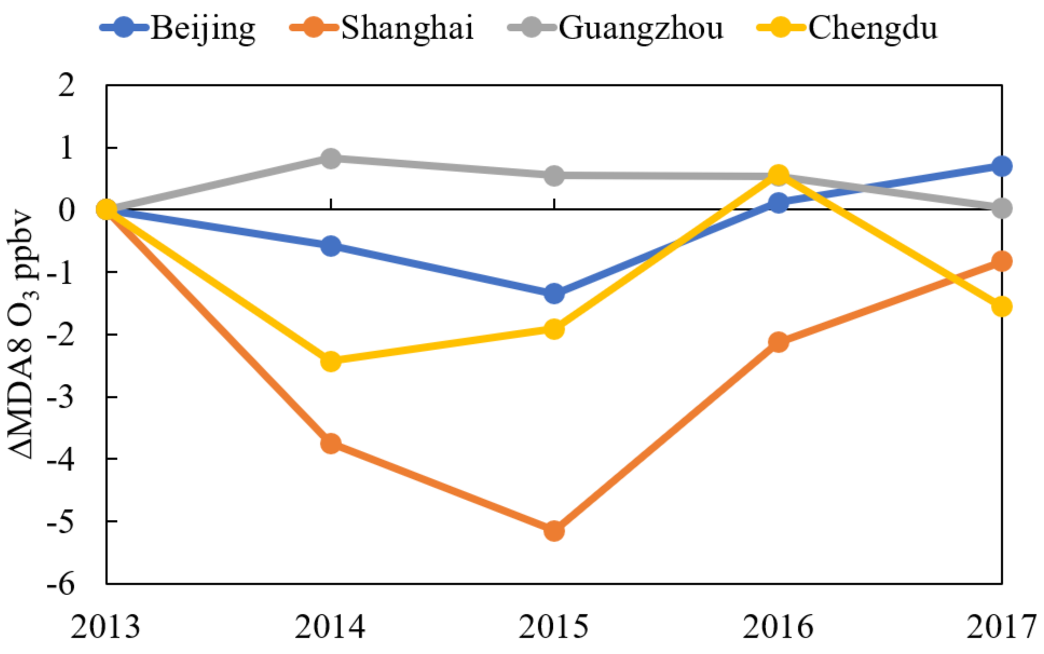

Figure S4 Changes in the simulated summer surface MDA8 O_3 due to variations in biogenic emissions in Beijing, Shanghai, Guangzhou, and Chengdu from 2013 to 2017 relative to 2013.

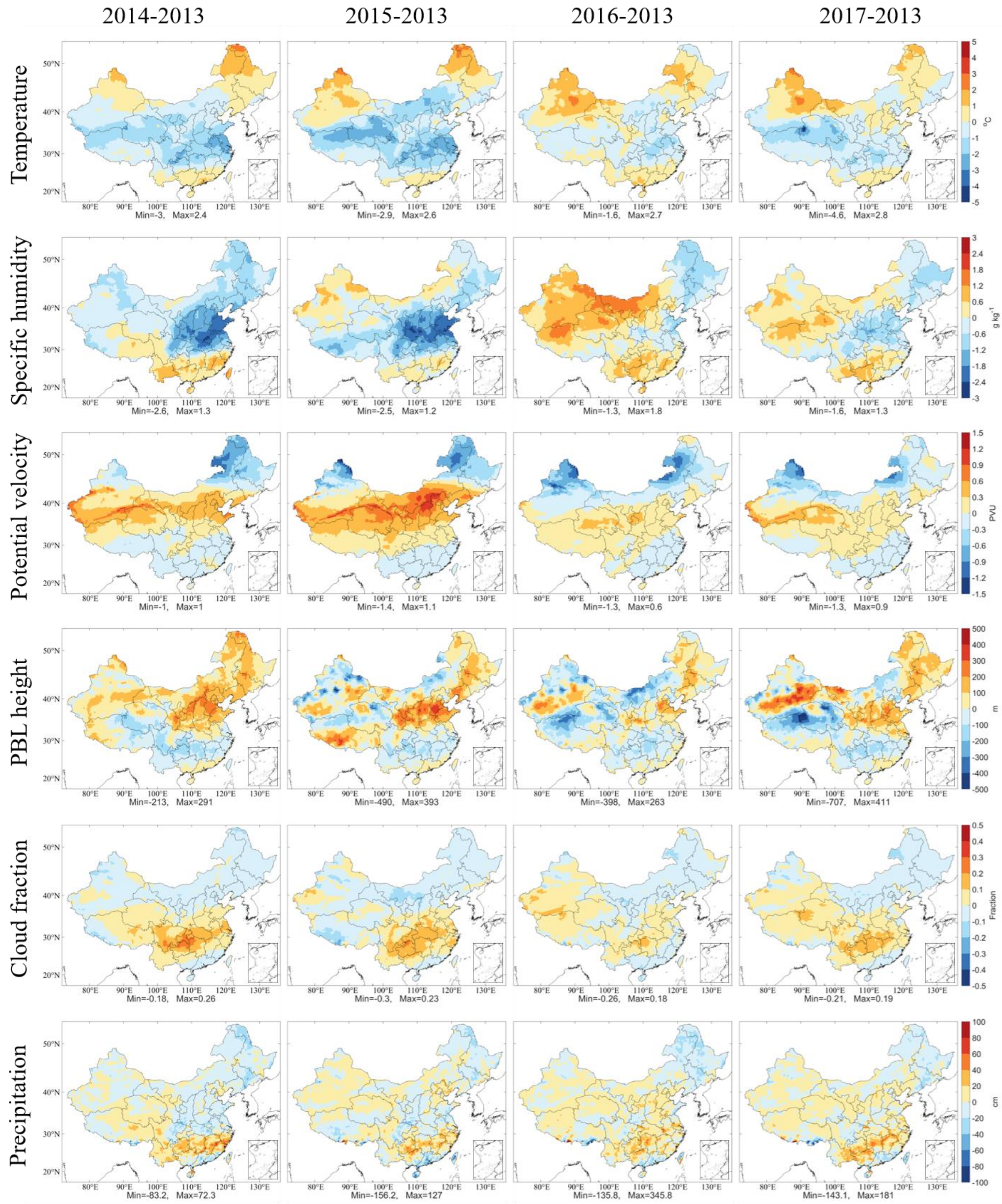

Figure S5 Changes in averaged temperature at a height of 2 m, specific humidity at a height of 2 m, potential velocity at a height of ~300 hPa (PVU: potential vorticity unit; 1 PVU=10^{-6} km^2 kg^{-1} s^{-1}), planetary boundary layer height, total clouds fraction, and accumulated precipitation in the daytime in summer from 2014 to 2017 relative 2013.

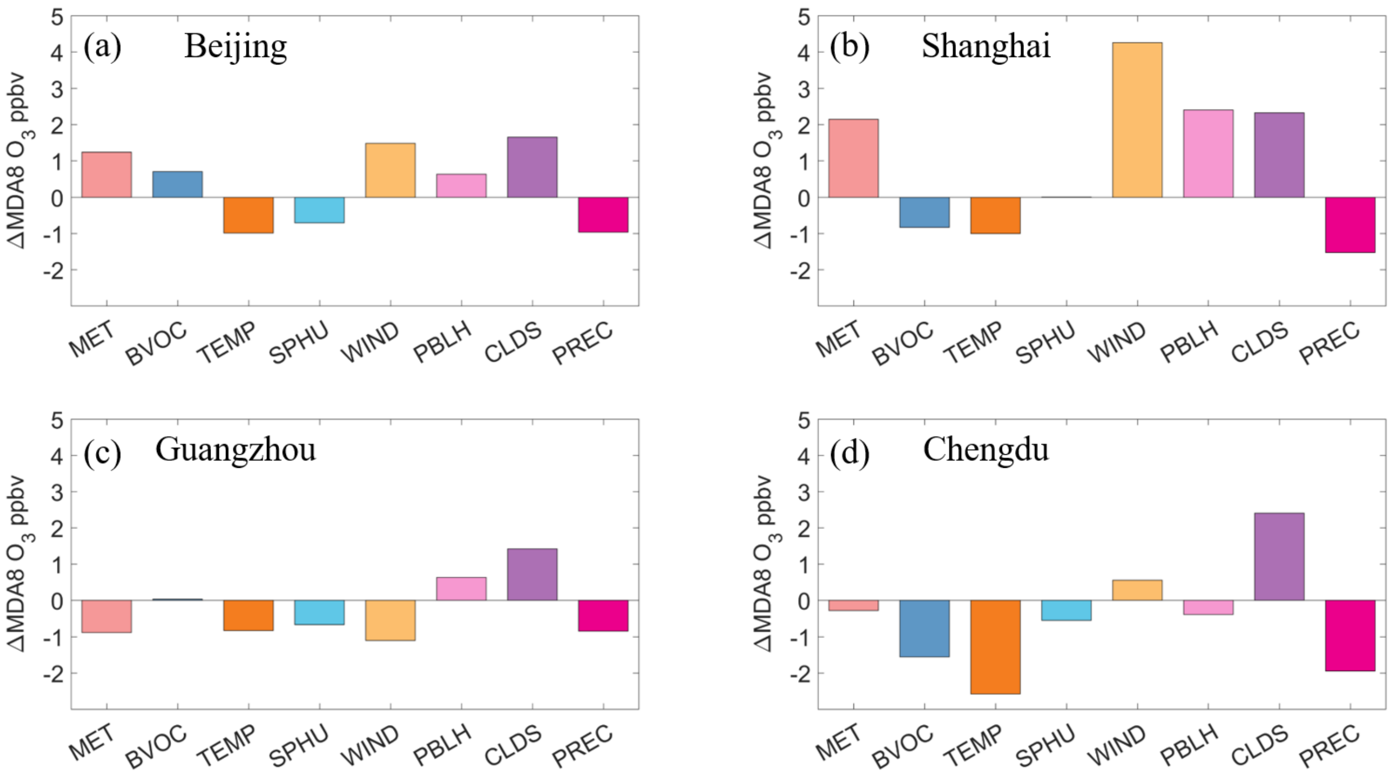

Figure S6 Changes in the simulated summer surface MDA8 O_3 concentrations in (a) Beijing, (b) Shanghai, (c) Guangzhou, and (d) Chengdu due to variations in various meteorological factors in 2017 relative to 2013, including meteorology (MET), temperature-dependent biogenic emissions (BVOC), temperature via changes in reaction rates (TEMP), specific humidity (SPHU), wind fields (WIND), planetary boundary layer height (PBLH), clouds (CLDS), and precipitation (PREC).

Table S1. Settings of physical parameterization schemes for the WRF model

Item	Option
Microphysics scheme	Lin et al. scheme
Longwave radiation scheme	rrtm scheme
Shortwave radiation scheme	Goddard short wave
Surface-layer scheme	Revised MM5 Monin-Obukhov scheme
Land-surface scheme	Unified Noah land-surface model
Boundary-layer scheme	YSU scheme
Cumulus scheme	Kain-Fritsch (new Eta) scheme
Nudging options	Grid four-dimensional data analysis (FDDA)

Table S2. Heterogeneous reactions included in the CMAQ model and their uptake coefficients.

No.	Heterogeneous reactions	Uptake coefficient (γ) or reaction rate (k)	References
Original CMAQ			
OR1	$N_2O_5(g) + H_2O(cd) + \varphi^a Cl^-(cd) \rightarrow$ $\varphi^a(HNO_3(g) + ClNO_2(g)) + 2(1-\varphi^a)HNO_3(g)$	$\gamma_{N_2O_5} = 3.2 \times 10^{-8} k' \left(1 - \frac{1}{\left(\frac{0.06[H_2O(l)]}{[NO_3^-]}\right) + 1 + \left(\frac{29[Cl^-]}{[NO_3^-]}\right)}\right)$ [b] $k' = 1.15 \times 10^6 - 1.15 \times 10^6 e^{[-0.13 \times [H_2O(l)]]}$ [b]	Bertram and Thornton (2009)
OR2	$NO_2(g) \rightarrow 0.5HONO(g) + 0.5HNO_3(g)$	$k_{NO_2} = 5 \times 10^{-5} \times (S/V)$ S/V = surface to volume ratio	Kurtenbach et al. (2001)
OR3	$NO_3(g) \rightarrow HNO_3(g)$	1.0×10^{-4}	Mao et al. (2013)
Newly updated CMAQ			
NR1	$N_2O_5(g) + H_2O(cd) + \varphi^a Cl^-(cd) \rightarrow$ $\varphi^a(HNO_3(g) + ClNO_2(g)) + 2(1-\varphi^a)HNO_3(g)$	$\gamma_{N_2O_5} = 3.2 \times 10^{-8} k \left(1 - \frac{1}{\left(\frac{0.06[H_2O(l)]}{[NO_3^-]}\right) + 1 + \left(\frac{29[Cl^-]}{[NO_3^-]}\right)}\right)$ [b] $k = 1.15 \times 10^6 - 1.15 \times 10^6 e^{[-0.13 \times [H_2O(l)]]}$ [b]	Bertram and Thornton (2009)
NR2	$NO_2(g) \rightarrow 0.5HONO(g) + 0.5HNO_3(g)$	$k_{NO_2} = 5 \times 10^{-5} \times f_{RH} \times (S/V)$ $f_{RH} = \begin{cases} RH/50 \ (RH < 50) \\ RH/10 - 4 \ (50 \le RH < 80) \\ 4 \ (RH \ge 80) \end{cases}$ $k_{NO_2} = 1 \times 10^{-3} \times \frac{Light\ intensity}{400} \times (S/V)$ RH = relative humidity S/V = surface to volume ratio	Fu et al. (2019)
NR3	$NO_3(g) \rightarrow HNO_3(g)$	1.0×10^{-3}	Jacob (2000)
NR4	$HO_2(g) \rightarrow 0.5H_2O_2(g)$	2.0×10^{-1}	Jacob (2000)
NR5	$OH(g) \rightarrow$ Products	1.0×10^{-1}	DeMore et al. (1997)
NR6	$O_3(g) \rightarrow$ Products	1.0×10^{-5}	Bauer et al. (2004)
NR7	$H_2O_2(g) \rightarrow$ Products	2.0×10^{-3}	de Reus et al. (2005)

[a] The yield of $ClNO_2$.

[b] $[H_2O(l)]$, $[NO_3^-]$, $[Cl^-]$ are the concentrations of particle liquid water, particulate nitrate, and particulate chloride, respectively.

Table S3. Settings of sensitivity experiments for investigating the effects of changes in meteorological conditions and anthropogenic emissions on ozone variations.

Experiment	Description
M13E13	Modeling the summer of 2013 with MEIC 2013 anthropogenic emission
M14E14	Modeling the summer of 2014 with MEIC 2014 anthropogenic emission
M15E15	Modeling the summer of 2015 with MEIC 2015 anthropogenic emission
M16E16	Modeling the summer of 2016 with MEIC 2016 anthropogenic emission
M17E17	Modeling the summer of 2017 with MEIC 2017 anthropogenic emission
M14E13_BC13	Modeling the summer of 2014 with MEIC 2013 anthropogenic emission and 2013 chemical boundary condition from MOZART
M15E13_BC13	Modeling the summer of 2015 with MEIC 2013 anthropogenic emission and 2013 chemical boundary condition from MOZART
M16E13_BC13	Modeling the summer of 2016 with MEIC 2013 anthropogenic emission and 2013 chemical boundary condition from MOZART
M17E13_BC13	Modeling the summer of 2017 with MEIC 2013 anthropogenic emission and 2013 chemical boundary condition from MOZART
M13E14	Modeling the summer of 2013 with MEIC 2014 anthropogenic emission
M13E15	Modeling the summer of 2013 with MEIC 2015 anthropogenic emission
M13E16	Modeling the summer of 2013 with MEIC 2016 anthropogenic emission
M13E17	Modeling the summer of 2013 with MEIC 2017 anthropogenic emission

Table S4. Settings of sensitivity experiments for investigating the impact of changes in biogenic emissions on ozone variations.

Experiment	Description
M13E13	Modeling the summer of 2013 with MEIC 2013 anthropogenic emission
M13E13_BIO14	M13E13 but with 2014 biogenic emission
M13E13_BIO15	M13E13 but with 2015 biogenic emission
M13E13_BIO16	M13E13 but with 2016 biogenic emission
M13E13_BIO17	M13E13 but with 2017 biogenic emission

Table S5. Settings of sensitivity experiments for investigating the impact of changes of temperature, specific humidity, wind field, planetary boundary layer height, cloud, and precipitation on ozone variations.

Experiment	Description
M13E13	Modeling the summer of 2013 with MEIC 2013 anthropogenic emission
M13E13_TEMP17	M13E13 but with 2017 temperature
M13E13_SPHU17	M13E13 but with 2017 specific humidity
M13E13_WIND17	M13E13 but with 2017 wind field
M13E13_PBLH17	M13E13 but with 2017 planetary boundary layer height
M13E13_CLDF17	M13E13 but with 2017 cloud
M13E13_PREC17	M13E13 but with 2017 precipitation

Table S6. Settings of sensitivity experiments for investigating the impact of changes in long-range transport on ozone variations.

Experiment	Description
M13E13	Modeling the summer of 2013 with MEIC 2013 anthropogenic emission
M13E13_BC14	M13E13 but with 2014 chemical boundary condition
M13E13_BC15	M13E13 but with 2015 chemical boundary condition
M13E13_BC16	M13E13 but with 2016 chemical boundary condition
M13E13_BC17	M13E13 but with 2017 chemical boundary condition

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