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Worsening urban ozone pollution in China from 2013 to 2017 – Part 2: The effects of emission changes and implications for multi-pollutant control

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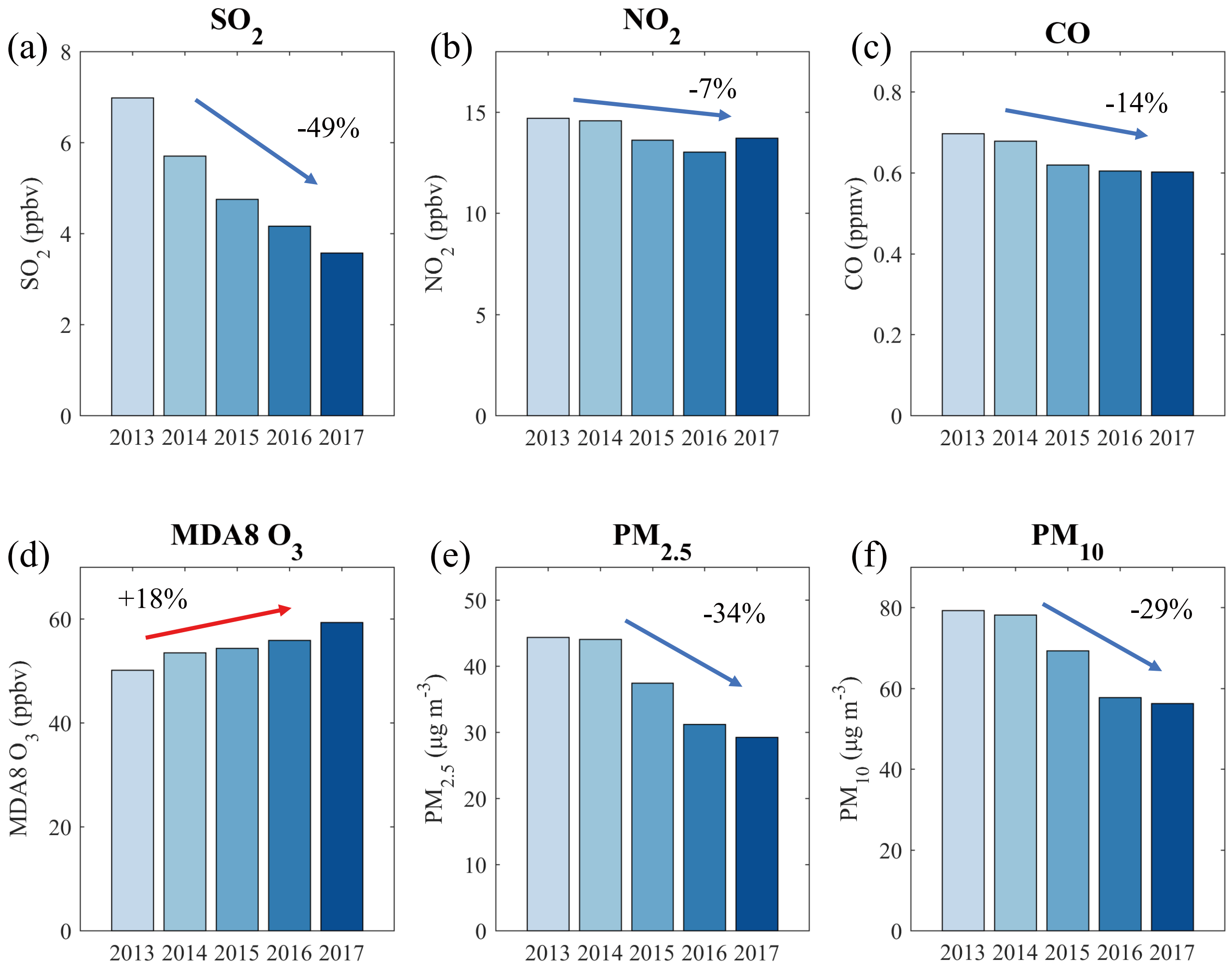

Figure S1: Variations in the observed mixing ratios/concentrations of air pollutants during 2013–2017 averaged at 493 sites in 74 cities of China, including (a) SO_2, (b) NO_2, (c) CO, (d) MDA8 O_3, (e) $PM_{2.5}$, (f) PM_{10}. Data are obtained from the China National Environmental Monitoring Center (http://106.37.208.233:20035/)

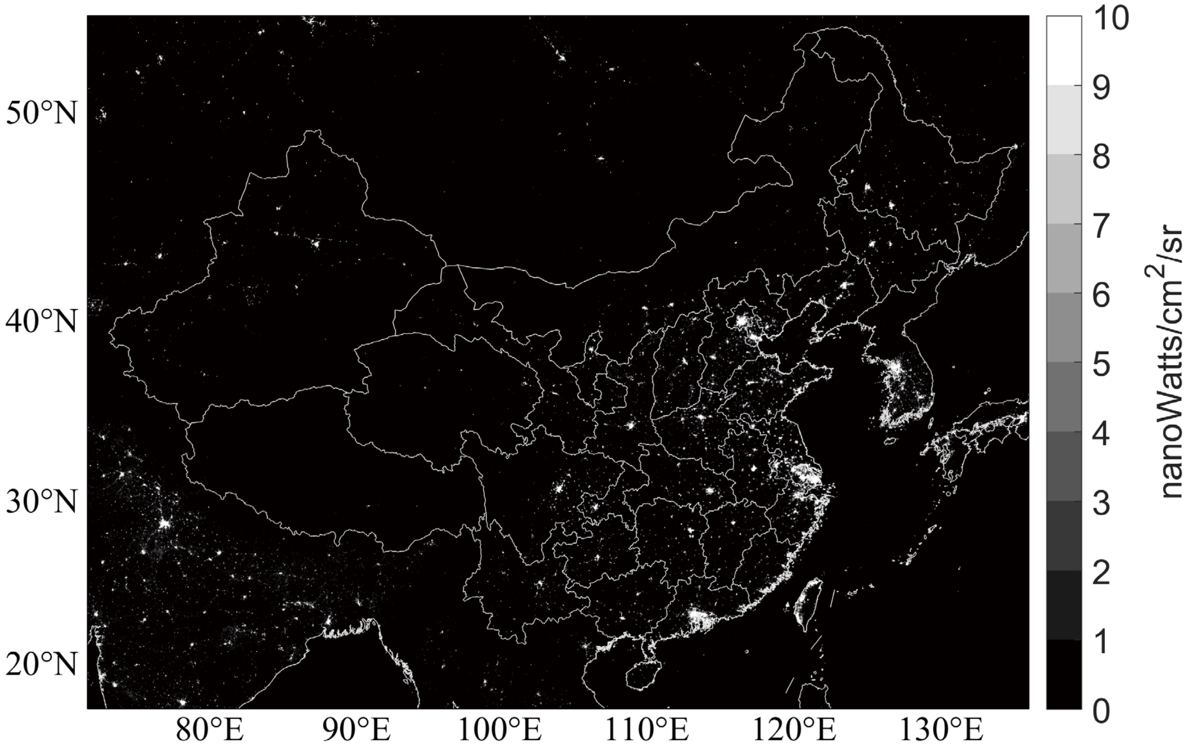

Figure S2: Spatial distribution of nighttime light data in 2015. The global nighttime light data with a horizontal resolution of 15 second is obtained from the Visible Infrared Imaging Radiometer Suite (VIIRS) Day/Night Band (DNB) (https://www.ngdc.noaa.gov/eog/viirs/download_dnb_composites.html).

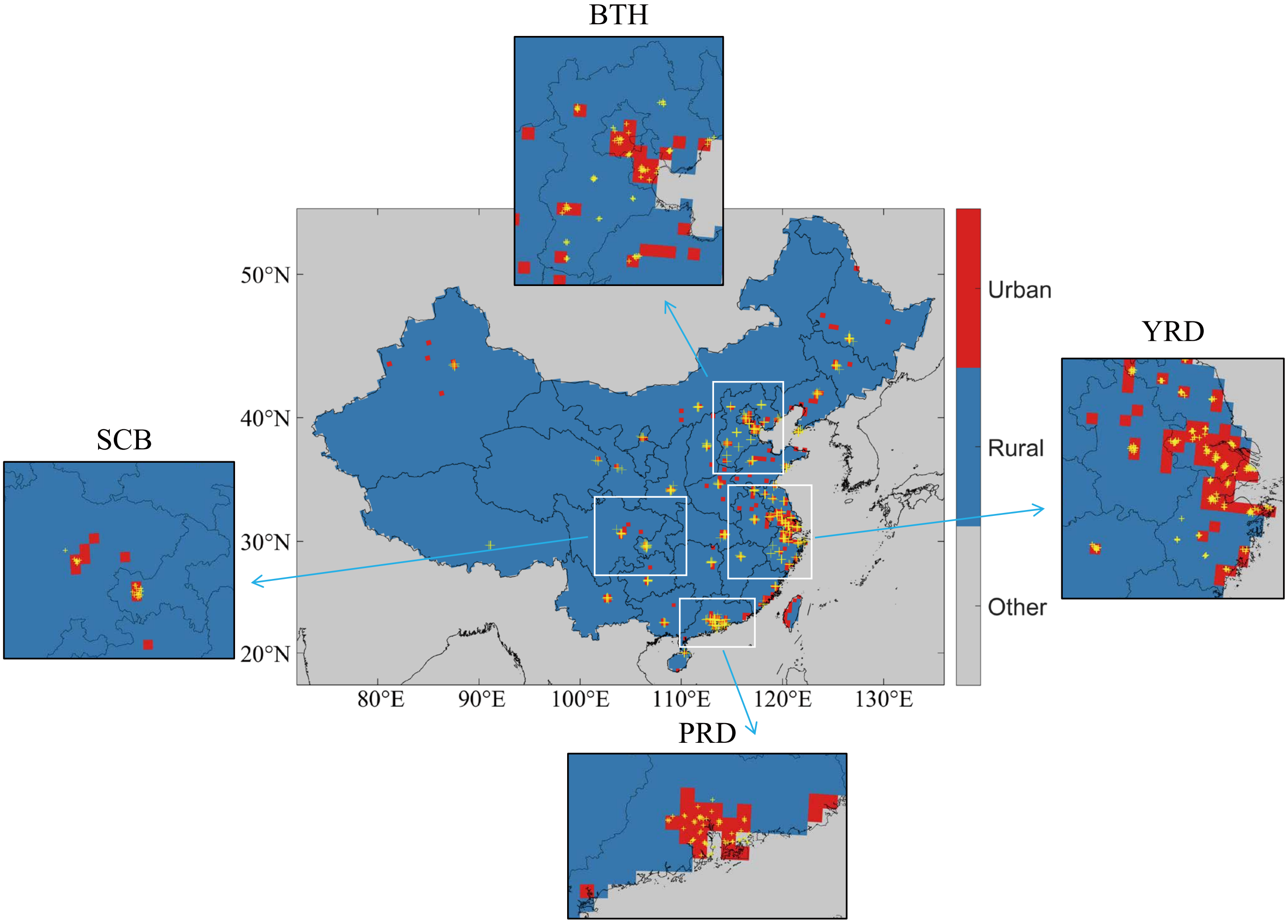

Figure S3: Spatial distribution of the urban and rural areas in land areas of China identified by using the nighttime light data. The yellow cross "+" represents the locations of 493 environmental monitoring stations in 74 cities since 2013. BTH, YRD, PRD, SCB are the Beijing-Tianjin-Hebei, Yangtze River Delta, Pearl River Delta, and Sichuan Basin regions, respectively.

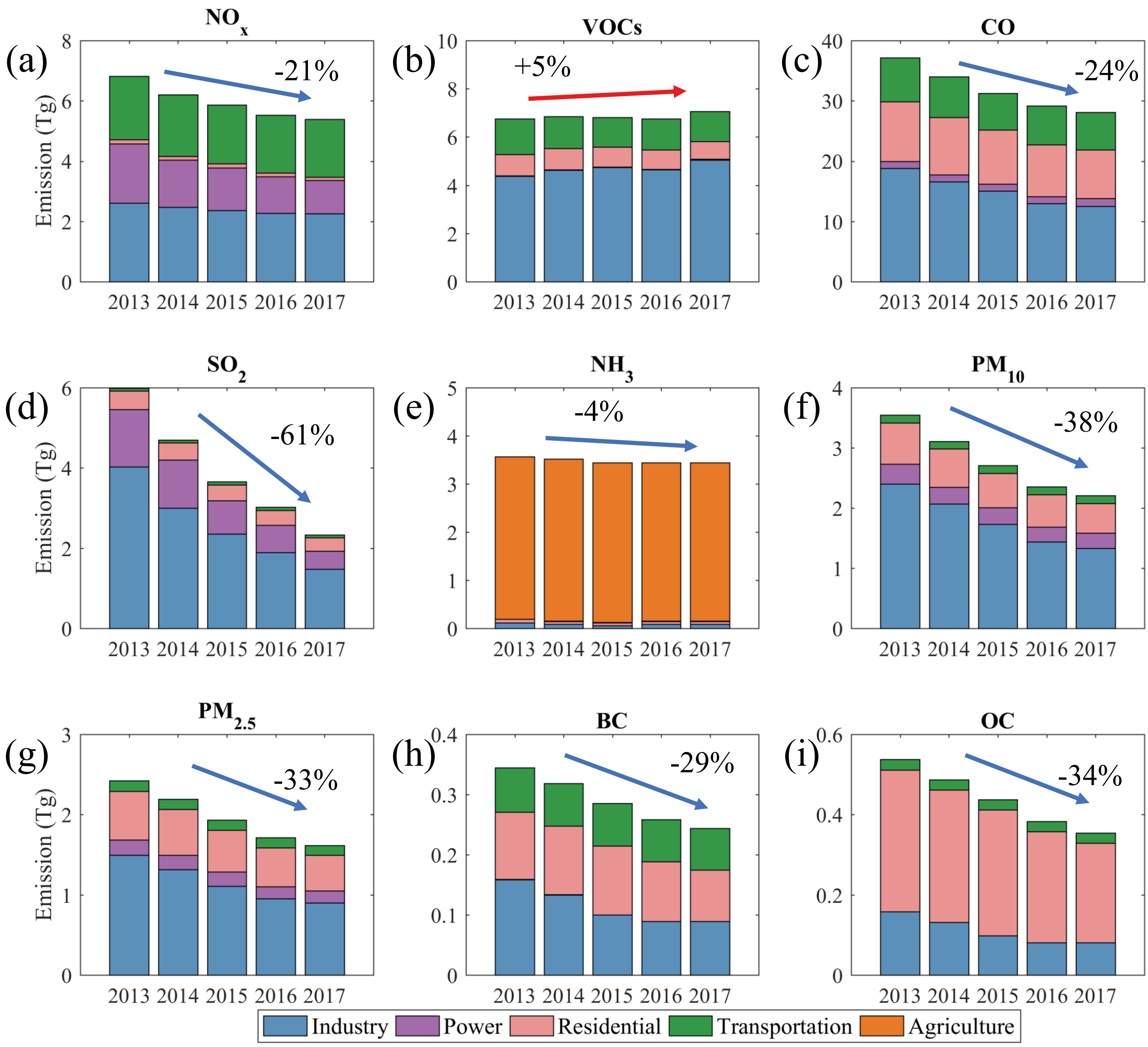

Figure S4: Trends of anthropogenic pollutant emissions in summer in mainland China from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO_2, (e) NH_3, (f) PM_{10}, (g) $PM_{2.5}$, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; http://www.meicmodel.org).

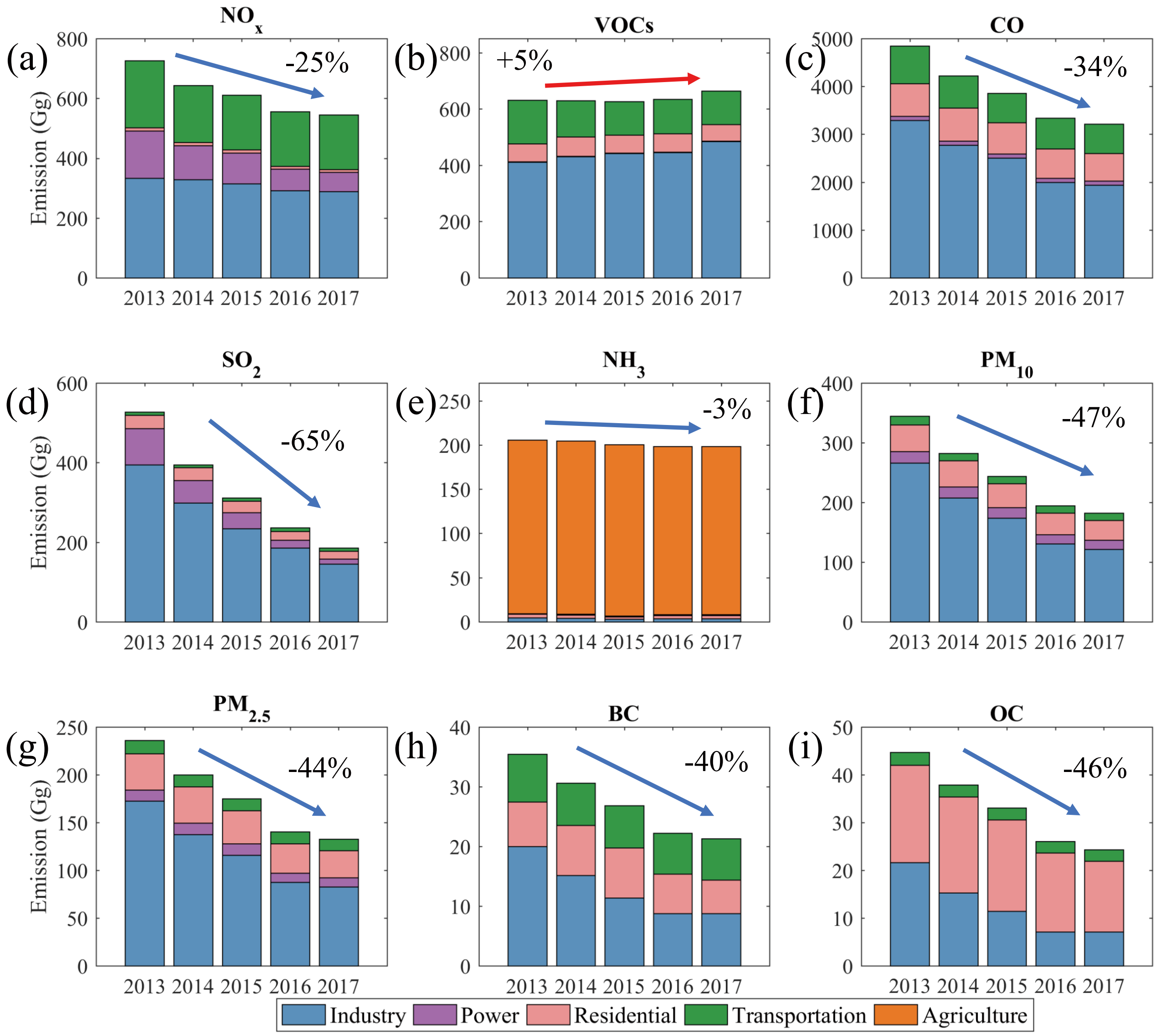

Figure S5: Variations in the anthropogenic pollutant emissions in Beijing-Tianjin-Hebei region in summer from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO_2, (e) NH_3, (f) PM_{10}, (g) $PM_{2.5}$, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; http://www.meicmodel.org).

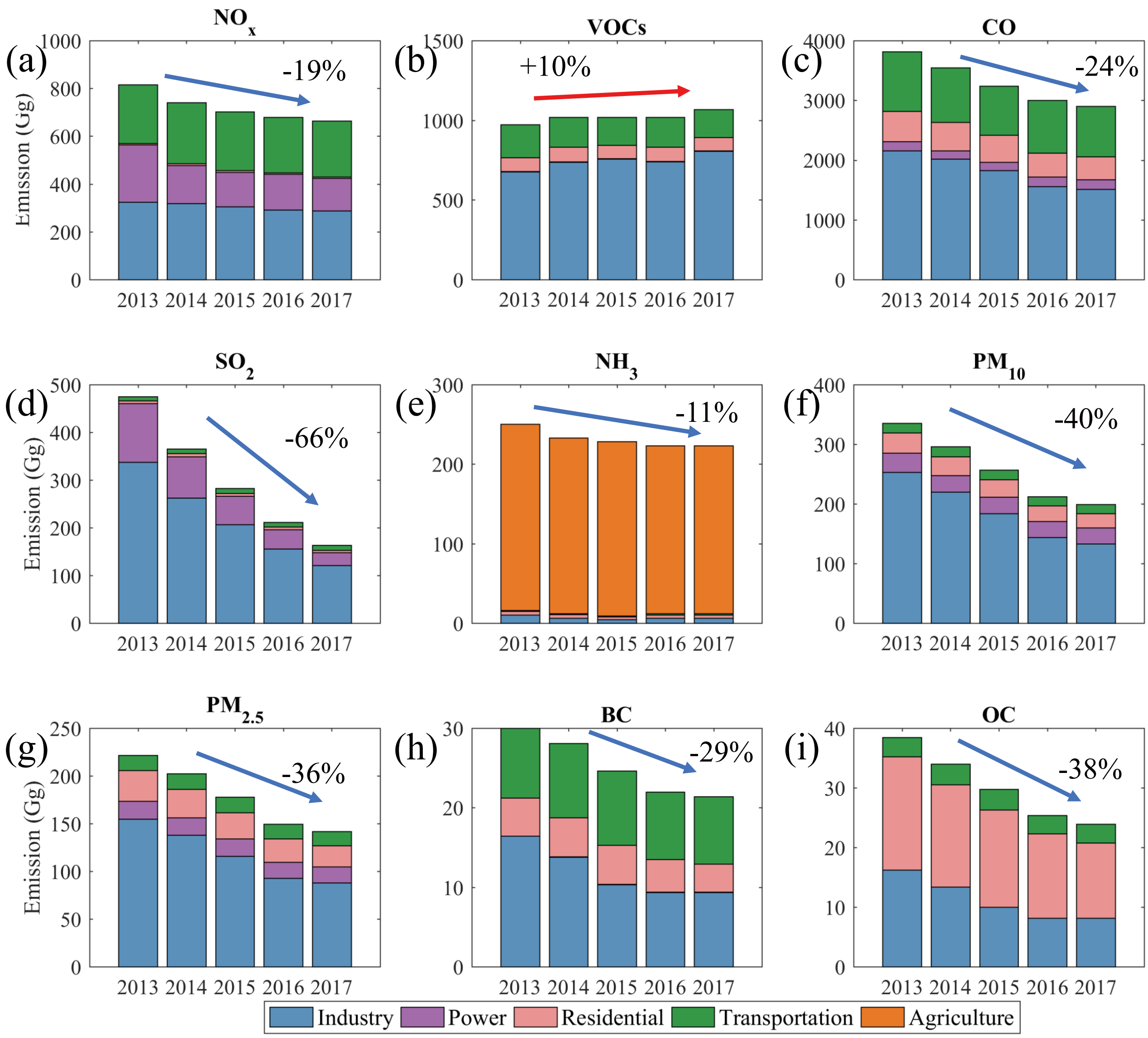

Figure S6: Variations in the anthropogenic pollutant emissions in Yangtze River Delta region in summer from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO_2, (e) NH_3, (f) PM_{10}, (g) $PM_{2.5}$, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; http://www.meicmodel.org).

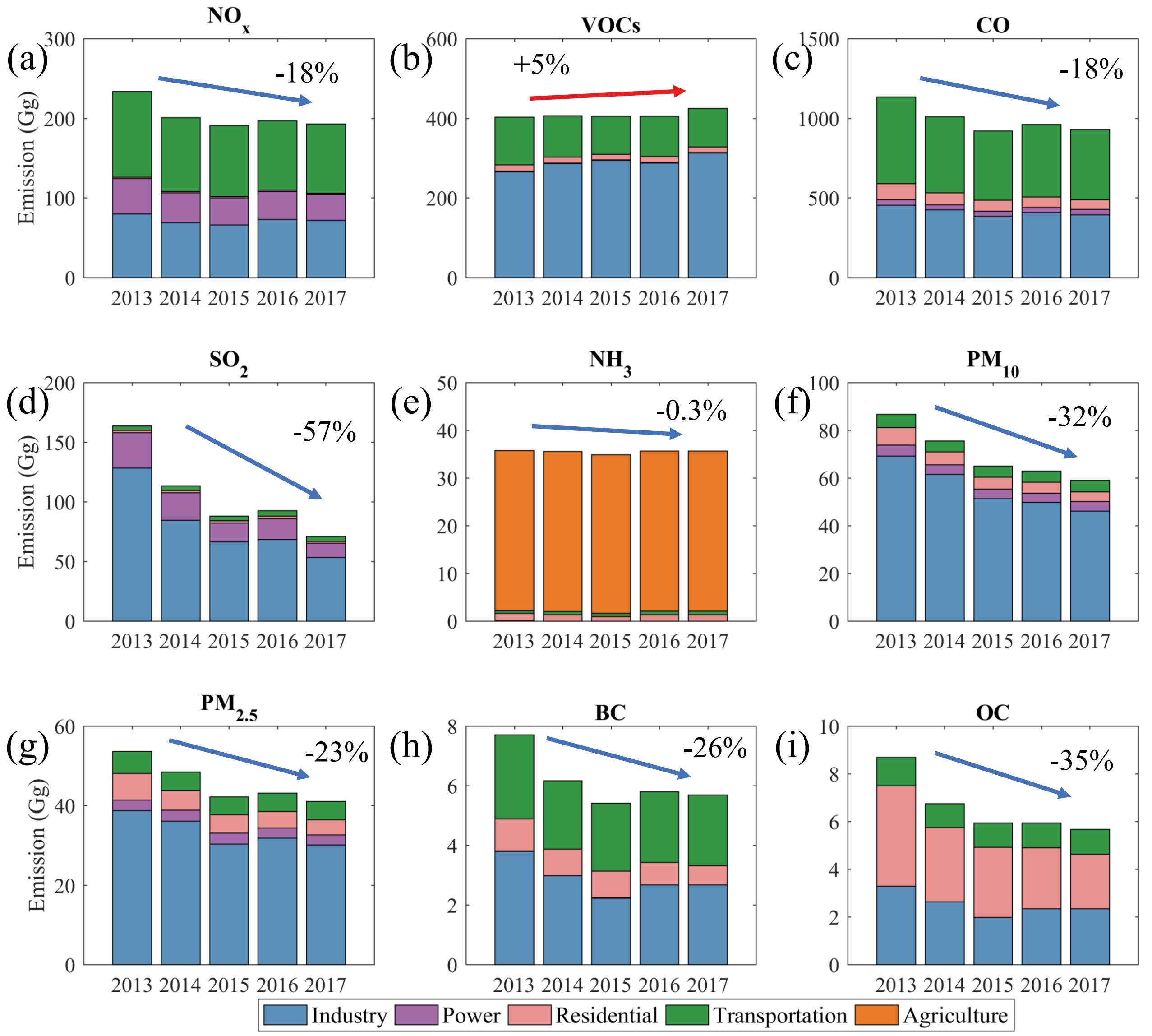

Figure S7: Variations in the anthropogenic pollutant emissions in Pearl River Delta region in summer from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO_2, (e) NH_3, (f) PM_{10}, (g) $PM_{2.5}$, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; http://www.meicmodel.org).

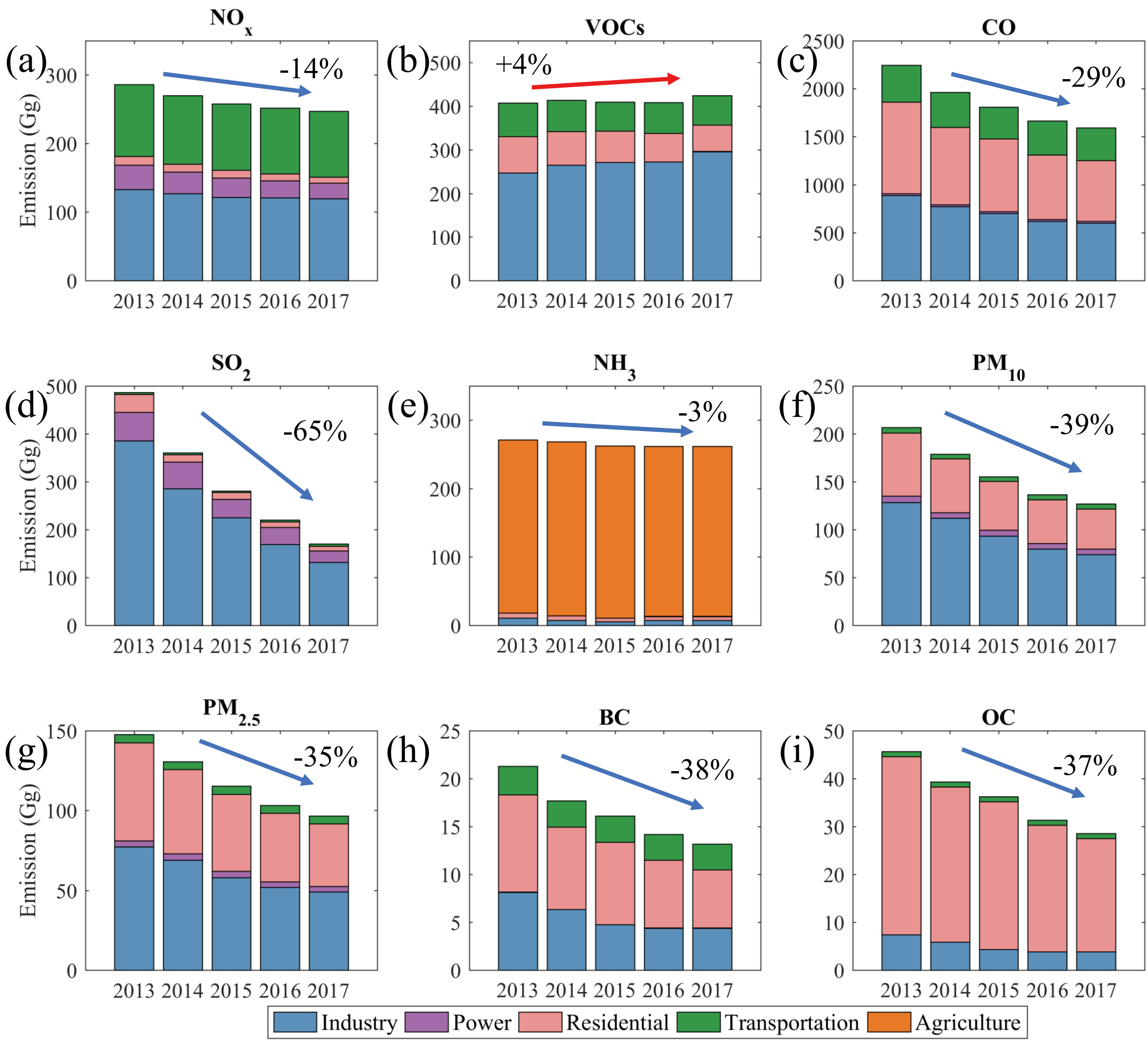

Figure S8: Variations in the anthropogenic pollutant emissions in Sichuan Basin in summer from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO_2, (e) NH_3, (f) PM_{10}, (g) $PM_{2.5}$, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; http://www.meicmodel.org).

Table S1: Comparison of simulated mixing ratios of MDA8 O_3 and other reactive gases in summer 2013 before and after the inclusion of heterogeneous reactions on aerosol surfaces with observed values.

Pollutant	Site	Period	Before	After	Observation	Reference
NO_2 (mean, ppbv)	430 sites in China	Jun - Aug 2013	19.2	16.6	15.1	
HONO (mean ppbv)	Beijing	20 Jun - 25 Jul 2016	0.74	1.37	1.38	Wang et al. (2017a)
	Guangzhou	3-24 Jul 2006	0.59	0.82	~2.8	Qin et al. (2009)
	Backgarden, Guangzhou	3-31 Jul 2006	0.56	1.48	1.04	Li et al. (2012b)
	Hok Tsui, Hong Kong	23-28 Aug 2012	0.03	0.04	0.54	Li et al. (2018)
	Tung Chung, Hong Kong	3 Aug - 7 Sep 2011	0.04	0.06	0.65	Xu et al. (2015)
NO_3 (mean pptv)	Fudan University, Shanghai	15 Aug – 7 Oct 2011	20	19	16	Wang et al. (2013)
	Backgarden, Guangzhou	4 -11 Jul 2006	9	8	22	Li et al. (2012a)
N_2O_5 (mean, pptv)	Wangdu	20 Jun - 9 July 2014	<228	<114	<80	Tham et al. (2016)
	Mount Tai	24 Jul - 27 Aug 2014	44	23	6.8	Wang et al. (2017b)
			<474	<281	<167	
	Hok Tsui, Hong Kong	23-26 Aug 2012	10	8	49	Yan et al. (2019)
$ClNO_2$ (mean, pptv)	Wangdu	20 Jun – 9 July 2014	<383	<318	<550	Tham et al. (2016)
			1733 (Maxima)	1695 (Maxima)	2070 (Maxima)	
	Mount Tai	24 Jul - 27 Aug 2014	174	144	54	Wang et al. (2017b)
			1631 (Maxima)	1366 (Maxima)	2065 (Maxima)	
	Hok Tsui, Hong Kong	23 Aug - 1 Sep 2012	18	20	148	Tham et al. (2014)
			1588 (Maxima)	1561 (Maxima)	1997 (Maxima)	
HO_2 (daily maxima, cm^{-3})	Wangdu	7 Jun - 8 Jul 2014	$(1–9)\times10^8$	$(1–6)\times10^8$	$(3–14)\times10^8$	Tan et al. (2017)
	Yufa, Beijing	18-31 Aug 2006	$(1–10)\times10^8$	$(1–9)\times10^8$	$(2–24)\times10^8$	Lu et al. (2013)
	Backgarden, Guangzhou	3-30 Jul 2006	$(1–12)\times10^8$	$(1–10)\times10^8$	$(3–25)\times10^8$	Lu et al. (2012)
OH (daily maxima, cm^{-3})	Wangdu	7 Jun - 8 Jul 2014	$(1–20)\times10^6$	$(1–19)\times10^6$	$(5–15)\times10^6$	Tan et al. (2017)
	Yufa, Beijing	18-31 Aug 2006	$(1–19)\times10^6$	$(3–18)\times10^6$	$(4–17)\times10^6$	Lu et al. (2013)
	Backgarden, Guangzhou	3-30 Jul 2006	$(2–20)\times10^6$	$(3–20)\times10^6$	$(15–26)\times10^6$	Lu et al. (2012)
MDA8 O_3 (mean, ppbv)	371 sites in China	Jun - Aug 2013	62.3	57.7	50.9	
H_2O_2 (mean, ppbv)	Wangdu	4 Jun - 7 Jul 2014	1.04	0.19	0.51	Wang et al. (2016)
	Peking University, Beijing	11-29 Aug 2015	0.78	0.19	0.27	Qin et al. (2018)
	Peking University, Beijing	7-15 Aug 2011	0.78	0.19	0.1	Liang et al. (2013)
	Peking University, Beijing	10-18 Aug 2010	0.78	0.19	0.2	Liang et al. (2013)

Table S2: Set-up of sensitivity experiments to investigate the impact of changed individual anthropogenic species emissions on ozone trend.

Experiment	Description
M13E13	Modeling the summer of 2013 with MEIC 2013 anthropogenic emission
M13E13_NOx17	M13E13 but with 2017 anthropogenic NO_x emission
M13E13_VOCs17	M13E13 but with 2017 anthropogenic VOCs emission
M13E13_SO217	M13E13 but with 2017 anthropogenic SO_2 emission
M13E13_CO17	M13E13 but with 2017 anthropogenic CO emission
M13E13_NH317	M13E13 but with 2017 anthropogenic NH_3 emission
M13E13_PM17	M13E13 but with 2017 anthropogenic PM emission
M13E13_EC17	M13E13 but with 2017 anthropogenic EC emission
M13E13_OC17	M13E13 but with 2017 anthropogenic OC emission
M13E13_NOxVOCs17	M13E13 but with 2017 anthropogenic NO_x and VOCs emission

Table S3: Set-up of sensitivity experiments to investigate the aerosol impact through the changes of photolysis rates and heterogeneous uptakes on ozone trend.

Experiment	Description
M13E13	Modeling the summer of 2013 with MEIC 2013 anthropogenic emission
M13E13_nophot	M13E13 but without aerosol impact through the change of photolysis rate
M13E13_nohet	M13E13 but without all heterogeneous reactions on aerosol surface
M13E13_nohetNO2	M13E13 but without the uptake of NO_2 on aerosol surface
M13E13_nohetNO3	M13E13 but without the uptake of NO_3 on aerosol surface
M13E13_nohetN2O5	M13E13 but without the uptake of N_2O_5 on aerosol surface
M13E13_nohetHO2	M13E13 but without the uptake of HO_2 on aerosol surface
M13E13_nohetOH	M13E13 but without the uptake of OH on aerosol surface
M13E13_nohetO3	M13E13 but without the uptake of O_3 on aerosol surface
M13E13_nohetH2O2	M13E13 but without the uptake of H_2O_2 on aerosol surface
M13E17	Modeling the summer of 2013 with MEIC 2017 anthropogenic emission
M13E17_nophot	M13E17 but without aerosol impact through the change of photolysis rate
M13E17_nohet	M13E17 but without all heterogeneous reactions on aerosol surface
M13E17_nohetNO2	M13E17 but without the uptake of NO_2 on aerosol surface
M13E17_nohetNO3	M13E17 but without the uptake of NO_3 on aerosol surface
M13E17_nohetN2O5	M13E17 but without the uptake of N_2O_5 on aerosol surface
M13E17_nohetHO2	M13E17 but without the uptake of HO_2 on aerosol surface
M13E17_nohetOH	M13E17 but without the uptake of OH on aerosol surface
M13E17_nohetO3	M13E17 but without the uptake of O_3 on aerosol surface
M13E17_nohetH2O2	M13E17 but without the uptake of H_2O_2 on aerosol surface

Table S4: Set-up of sensitivity experiments for assessing the response of MDA8 O_3 to anthropogenic VOCs emission reduction.

Experiment	Description
M13E13	Modeling the summer of 2013 with MEIC 2013 anthropogenic emission
M13E17_VOCs13	Same as M13E13 but with 2017 anthropogenic emissions except for VOCs
M13E17_VOCs13_10%	Same as M13E17_VOCs13 but with 10% reduction of 2013 anthropogenic VOCs emission
M13E17_VOCs13_20%	Same as M13E17_VOCs13 but with 20% reduction of 2013 anthropogenic VOCs emission
M13E17_VOCs13_30%	Same as M13E17_VOCs13 but with 30% reduction of 2013 anthropogenic VOCs emission
M13E17_VOCs13_40%	Same as M13E17_VOCs13 but with 40% reduction of 2013 anthropogenic VOCs emission
M13E17_VOCs13_50%	Same as M13E17_VOCs13 but with 50% reduction of 2013 anthropogenic VOCs emission

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