

Figure S1: Variations in the observed air pollutant concentrations during 2013–2017 averaged in 74 cities in China, including (a) SO_2 , (b) NO_2 , (c) CO, (d) MDA8 O_3 , (e) $\text{PM}_{2.5}$, (f) PM_{10} . Data are obtained from the China National Environmental Monitoring Center (<http://106.37.208.233:20035/>)

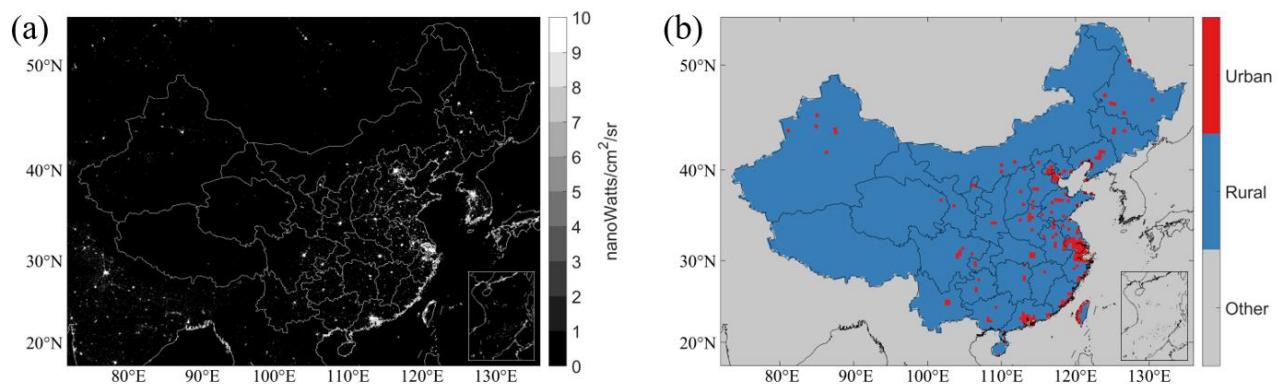


Figure S2: (a) Spatial distribution of nighttime light data in 2015 across China. The global nighttime light data with a horizontal resolution of 15 second is obtained from the Visible Infrared Imaging Radiometer Suite (VIIRS) Day/Night Band (DNB) (https://www.ngdc.noaa.gov/eog/viirs/download_dnb_composites.html).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

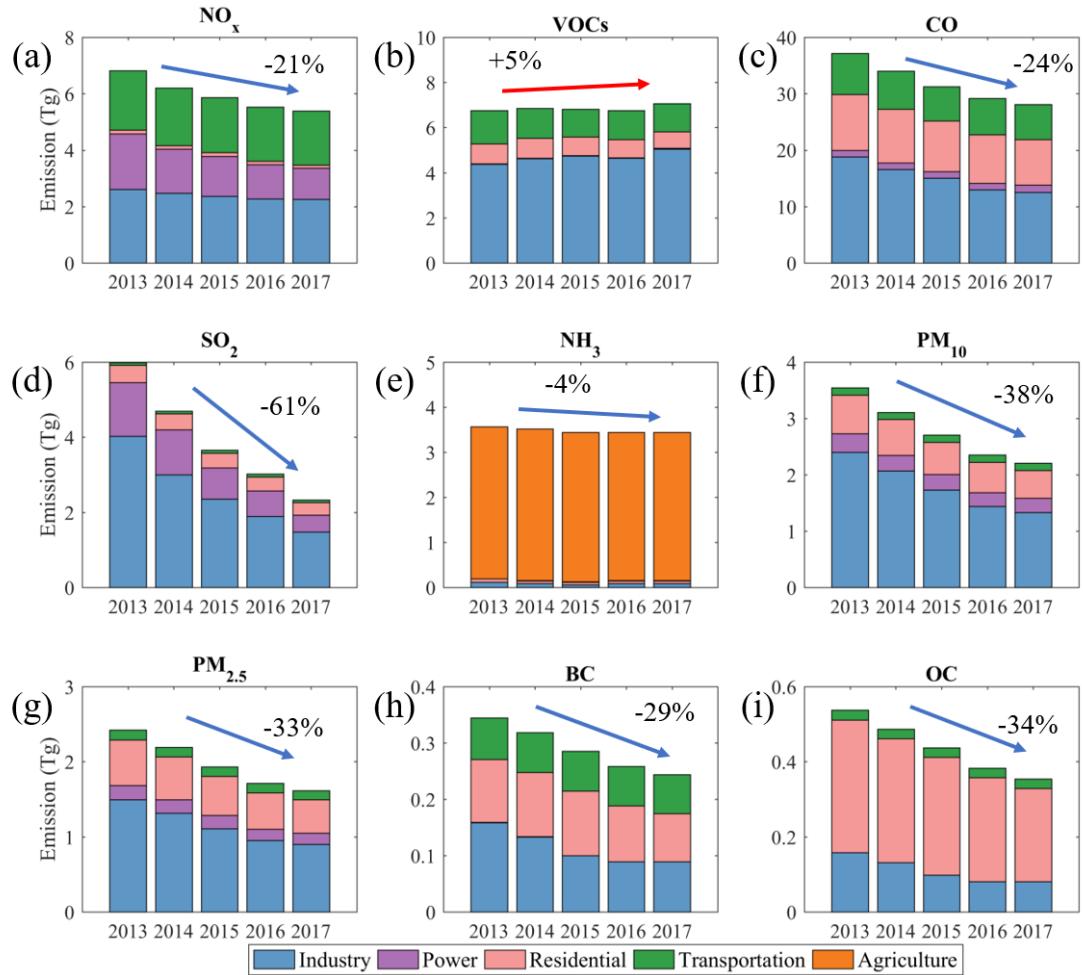


Figure S3: Trends of anthropogenic pollutant emissions in summer in mainland China from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO₂, (e) NH₃, (f) PM₁₀, (g) PM_{2.5}, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; <http://www.meicmodel.org>).

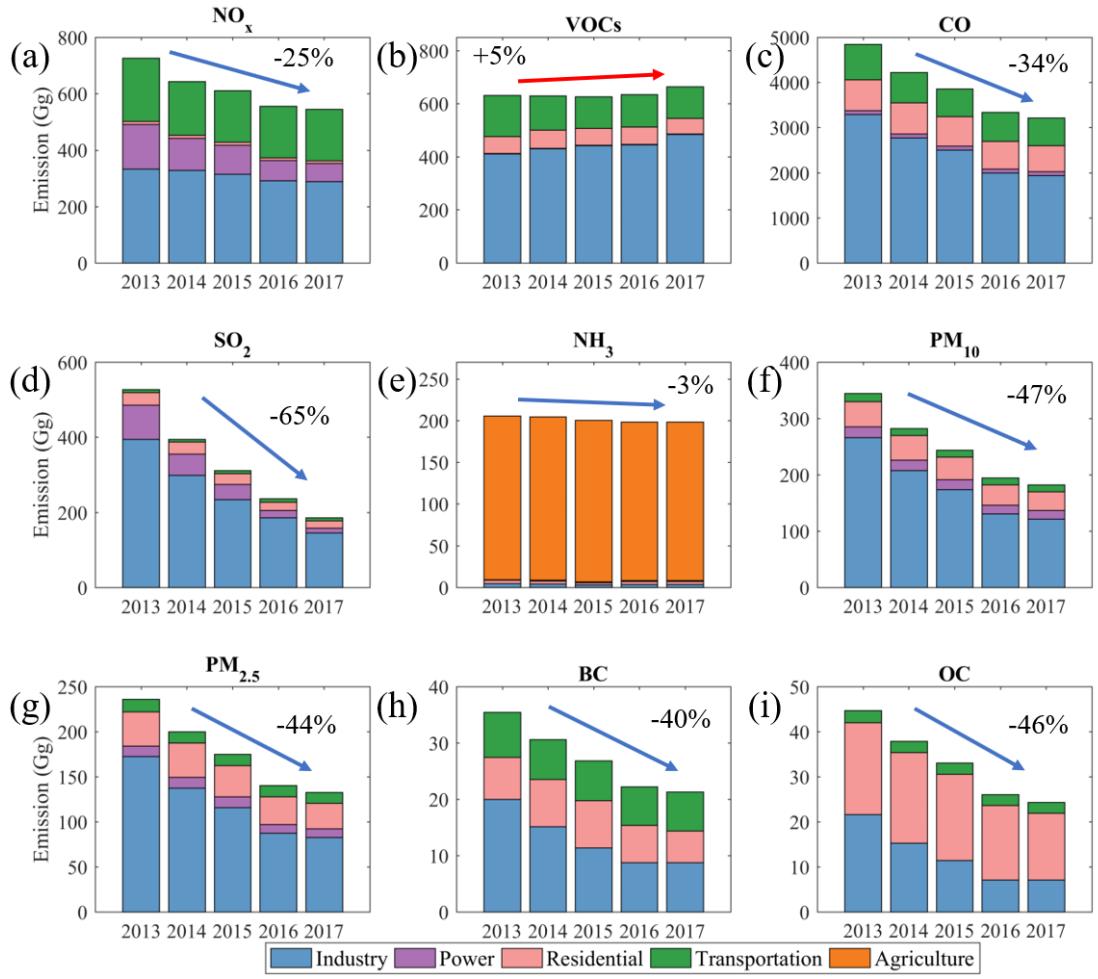


Figure S4: Variations in the anthropogenic pollutant emissions in Beijing-Tianjin-Hebei region in summer from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO₂, (e) NH₃, (f) PM₁₀, (g) PM_{2.5}, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; <http://www.meicmodel.org>).

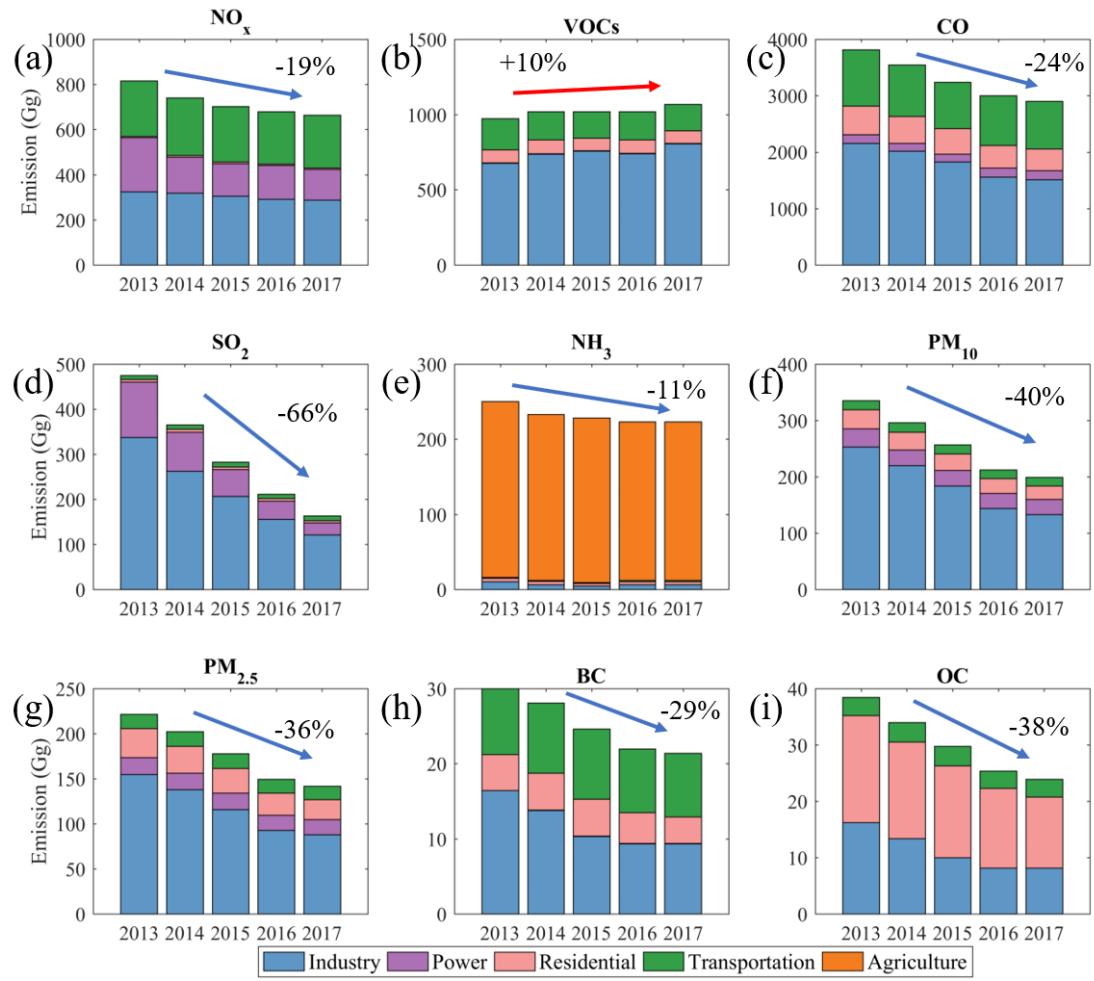


Figure S5: Variations in the anthropogenic pollutant emissions in Yangtze River Delta region in summer from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO₂, (e) NH₃, (f) PM₁₀, (g) PM_{2.5}, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; <http://www.meicmodel.org>).

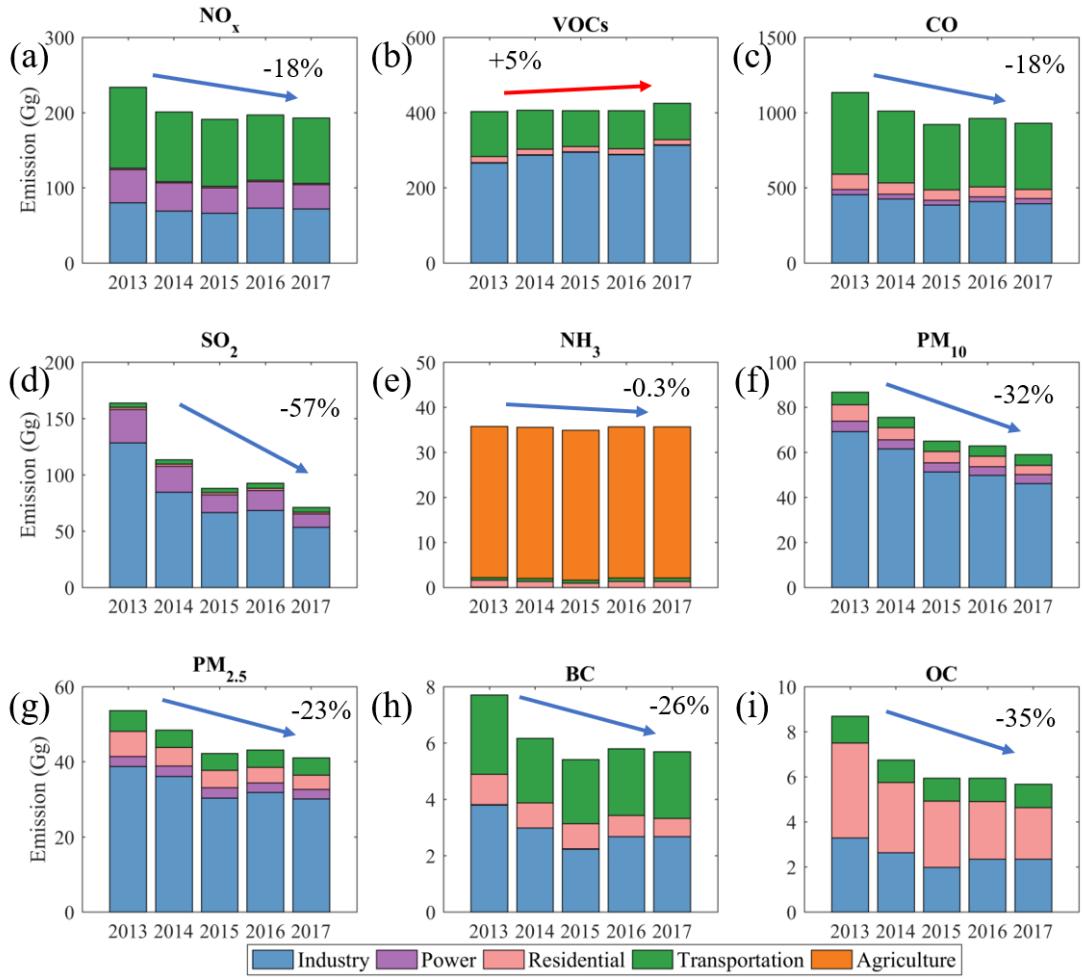


Figure S6: Variations in the anthropogenic pollutant emissions in Pearl River Delta region in summer from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO₂, (e) NH₃, (f) PM₁₀, (g) PM_{2.5}, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; <http://www.meicmodel.org>).

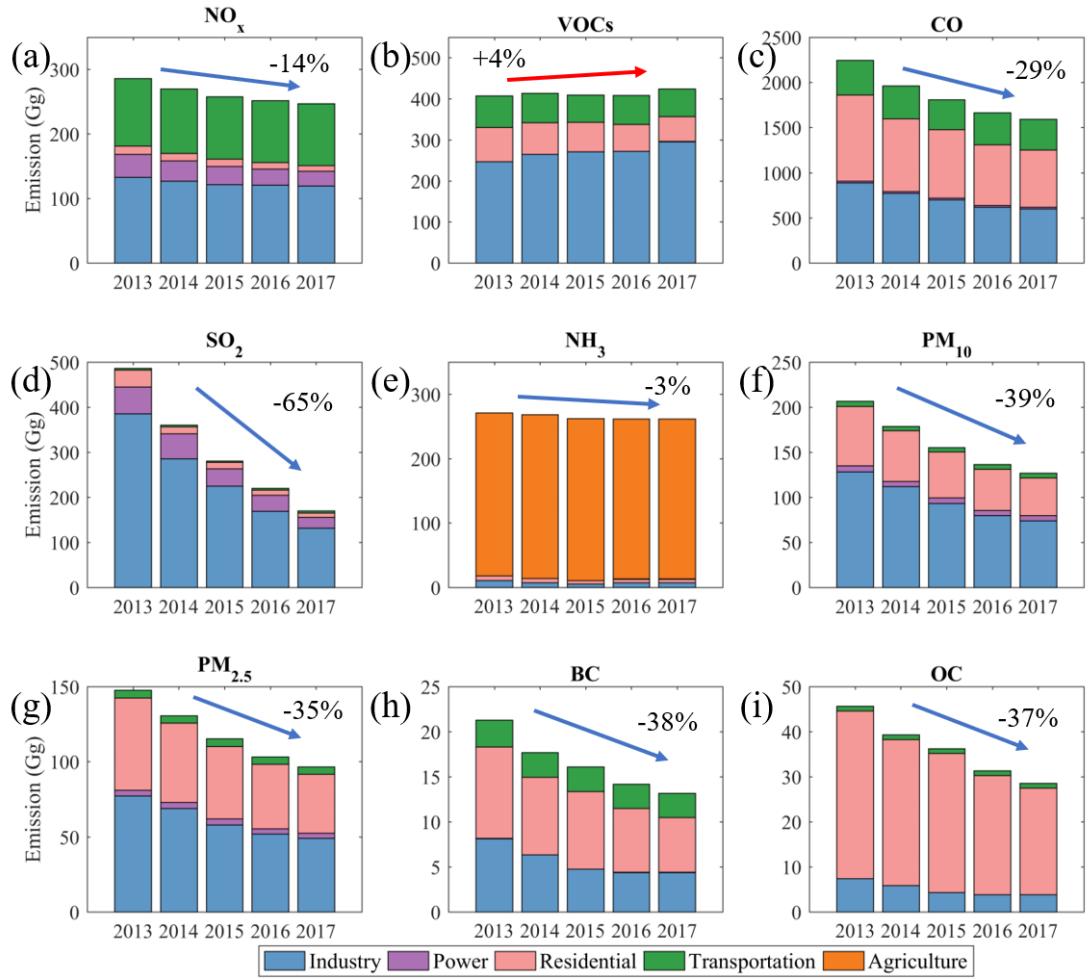


Figure S7: Variations in the anthropogenic pollutant emissions in Sichuan Basin in summer from 2013 to 2017, including (a) NO_x, (b) VOCs, (c) CO, (d) SO₂, (e) NH₃, (f) PM₁₀, (g) PM_{2.5}, (h) BC, and (i) OC. Emission data are obtained from Multi-resolution Emission Inventory for China (MEIC; <http://www.meicmodel.org>).

Table S1: Comparison of simulated summer MDA8 O₃ concentration in summer 2013 before and after the inclusion of heterogeneous reactions on aerosol surfaces.

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 ₂ O 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)
CINO 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)×10 ⁸	(1-6)×10 ⁸	(3-14)×10 ⁸	Tan et al. (2017)
	Yufa, Beijing	18-31 Aug 2006	(1-10)×10 ⁸	(1-9)×10 ⁸	(2-24)×10 ⁸	Lu et al. (2013)
	Backgarden, Guangzhou	3-30 Jul 2006	(1-12)×10 ⁸	(1-10)×10 ⁸	(3-25)×10 ⁸	Lu et al. (2012)
OH (daily maxima, cm ⁻³)	Wangdu	7 Jun - 8 Jul 2014	(1-20)×10 ⁶	(1-19)×10 ⁶	(5-15)×10 ⁶	Tan et al. (2017)
	Yufa, Beijing	18-31 Aug 2006	(1-19)×10 ⁶	(3-18)×10 ⁶	(4-17)×10 ⁶	Lu et al. (2013)
	Backgarden, Guangzhou	3-30 Jul 2006	(2-20)×10 ⁶	(3-20)×10 ⁶	(15-26)×10 ⁶	Lu et al. (2012)
MDA8 O ₃ (mean, ppbv)	371 sites in China	Jun - Aug 2013	62.3	57.7	50.9	
H ₂ O 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: Settings 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: Settings 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 ₂ O 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 ₂ O 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 ₂ O 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 ₂ O ₂ on aerosol surface

Table S4: Settings of sensitivity experiments for assessing the response of MDA8 O₃ concentrations 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

Reference:

- Li, S. W., Liu, W. Q., Xie, P. H., Qin, M., and Yang, Y. J.: Observation of Nitrate Radical in the Nocturnal Boundary Layer During a Summer Field Campaign in Pearl River Delta, China, *Terr Atmos Ocean Sci*, 23, 39–48, 10.3319/Tao.2011.07.26.01(a), 2012a.
- Li, X., Brauers, T., Häseler, R., Bohn, B., Fuchs, H., Hofzumahaus, A., Holland, F., Lou, S., Lu, K. D., Rohrer, F., Hu, M., Zeng, L. M., Zhang, Y. H., Garland, R. M., Su, H., Nowak, A., Wiedensohler, A., Takegawa, N., Shao, M., and Wahner, A.: Exploring the atmospheric chemistry of nitrous acid (HONO) at a rural site in Southern China, *Atmos. Chem. Phys.*, 12, 1497–1513, 10.5194/acp-12-1497-2012, 2012b.
- Li, Z. Y., Xue, L. K., Yang, X., Zha, Q. Z., Tham, Y. J., Yan, C., Louie, P. K. K., Luk, C. W. Y., Wang, T., and Wang, W. X.: Oxidizing capacity of the rural atmosphere in Hong Kong, Southern China, *Sci. Total Environ.*, 612, 1114–1122, 10.1016/j.scitotenv.2017.08.310, 2018.
- Liang, H., Chen, Z. M., Huang, D., Zhao, Y., and Li, Z. Y.: Impacts of aerosols on the chemistry of atmospheric trace gases: a case study of peroxides and HO₂ radicals, *Atmos. Chem. Phys.*, 13, 11259–11276, 10.5194/acp-13-11259-2013, 2013.
- Lu, K. D., Rohrer, F., Holland, F., Fuchs, H., Bohn, B., Brauers, T., Chang, C. C., Häseler, R., Hu, M., Kita, K., Kondo, Y., Li, X., Lou, S. R., Nehr, S., Shao, M., Zeng, L. M., Wahner, A., Zhang, Y. H., and Hofzumahaus, A.: Observation and modelling of OH and HO₂ concentrations in the Pearl River Delta 2006: a missing OH source in a VOC rich atmosphere, *Atmos. Chem. Phys.*, 12, 1541–1569, 10.5194/acp-12-1541-2012, 2012.
- Lu, K. D., Hofzumahaus, A., Holland, F., Bohn, B., Brauers, T., Fuchs, H., Hu, M., Häseler, R., Kita, K., Kondo, Y., Li, X., Lou, S. R., Oebel, A., Shao, M., Zeng, L. M., Wahner, A., Zhu, T., Zhang, Y. H., and Rohrer, F.: Missing OH source in a suburban environment near Beijing: observed and modelled OH and HO₂ concentrations in summer 2006, *Atmos. Chem. Phys.*, 13, 1057–1080, 10.5194/acp-13-1057-2013, 2013.
- Qin, M., Xie, P. H., Su, H., Gu, J. W., Peng, F. M., Li, S. W., Zeng, L. M., Liu, J. G., Liu, W. Q., and Zhang, Y. H.: An observational study of the HONO-NO₂ coupling at an urban site in Guangzhou City, South China, *Atmos. Environ.*, 43, 5731–5742, 10.1016/j.atmosenv.2009.08.017, 2009.
- Qin, M. R., Chen, Z. M., Shen, H. Q., Li, H., Wu, H. H., and Wang, Y.: Impacts of heterogeneous reactions to atmospheric peroxides: Observations and budget analysis study, *Atmos. Environ.*, 183, 144–153, 10.1016/j.atmosenv.2018.04.005, 2018.
- Tan, Z., Fuchs, H., Lu, K., Hofzumahaus, A., Bohn, B., Broch, S., Dong, H., Gomm, S., Häseler, R., He, L., Holland, F., Li, X., Liu, Y., Lu, S., Rohrer, F., Shao, M., Wang, B., Wang, M., Wu, Y., Zeng, L., Zhang, Y., Wahner, A., and Zhang, Y.: Radical chemistry at a rural site (Wangdu) in the North China Plain: observation and model calculations of OH, HO₂ and RO₂ radicals, *Atmos. Chem. Phys.*, 17, 663–690, 10.5194/acp-17-663-2017, 2017.
- Tham, Y. J., Yan, C., Xue, L. K., Zha, Q. Z., Wang, X. F., and Wang, T.: Presence of high nitril chloride in Asian coastal environment and its impact on atmospheric photochemistry, *Chinese Sci Bull*, 59, 356–359, 10.1007/s11434-013-0063-y, 2014.
- Tham, Y. J., Wang, Z., Li, Q., Yun, H., Wang, W., Wang, X., Xue, L., Lu, K., Ma, N., Bohn, B., Li, X., Kecorius, S., Größ, J., Shao, M., Wiedensohler, A., Zhang, Y., and Wang, T.: Significant concentrations of nitril chloride sustained in the morning: investigations of the causes and impacts on ozone production in a polluted region of northern China, *Atmos. Chem. Phys.*, 16, 14959–14977, 10.5194/acp-16-14959-2016, 2016.
- Wang, J. Q., Zhang, X. S., Guo, J., Wang, Z. W., and Zhang, M. G.: Observation of nitrous acid (HONO) in Beijing, China: Seasonal variation, nocturnal formation and daytime budget, *Sci. Total Environ.*, 587, 350–359, 10.1016/j.scitotenv.2017.02.159, 2017a.
- Wang, S. S., Shi, C. Z., Zhou, B., Zhao, H., Wang, Z. R., Yang, S. N., and Chen, L. M.: Observation of NO₃ radicals over Shanghai, China, *Atmos. Environ.*, 70, 401–409, 10.1016/j.atmosenv.2013.01.022, 2013.
- Wang, Y., Chen, Z., Wu, Q., Liang, H., Huang, L., Li, H., Lu, K., Wu, Y., Dong, H., Zeng, L., and Zhang, Y.: Observation of atmospheric peroxides during Wangdu Campaign 2014 at a rural site in the North China Plain, *Atmos. Chem.*

- Phys., 16, 10985-11000, 10.5194/acp-16-10985-2016, 2016.
- Wang, Z., Wang, W., Tham, Y. J., Li, Q., Wang, H., Wen, L., Wang, X., and Wang, T.: Fast heterogeneous N₂O₅ uptake and C₁N₂O₂ production in power plant and industrial plumes observed in the nocturnal residual layer over the North China Plain, *Atmos. Chem. Phys.*, 17, 12361-12378, 10.5194/acp-17-12361-2017, 2017b.
- Xu, Z., Wang, T., Wu, J. Q., Xue, L. K., Chan, J., Zha, Q. Z., Zhou, S. Z., Louie, P. K. K., and Luk, C. W. Y.: Nitrous acid (HONO) in a polluted subtropical atmosphere: Seasonal variability, direct vehicle emissions and heterogeneous production at ground surface, *Atmos. Environ.*, 106, 100-109, 10.1016/j.atmosenv.2015.01.061, 2015.
- Yan, C., Tham, Y. J., Zha, Q. Z., Wang, X. F., Xue, L. K., Dai, J. N., Wang, Z., and Wang, T.: Fast heterogeneous loss of N₂O₅ leads to significant nighttime NO_x removal and nitrate aerosol formation at a coastal background environment of southern China, *Sci. Total Environ.*, 677, 637-647, 10.1016/j.scitotenv.2019.04.389, 2019.