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The effect of anthropogenic emission, meteorological factors, and carbon dioxide on the surface ozone increase in China from 2008 to 2018 during the East Asia summer monsoon season

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Abstract. Increasing surface ozone (O_3) concentrations have long been a significant environmental issue in China, despite the Clean Air Action Plan launched in 2013 by the government. In this study, we assessed the effect of anthropogenic emissions, meteorological factors, and CO_2 changes on the summer surface O_3 from 2008 to 2018 in China using an improved regional climate-chemistry-ecology model (RegCM-Chem-YIBs). The model was improved regarding the photolysis of O_3 and the radiation effect of CO_2 and O_3. The investigations showed anthropogenic emissions dominated the O_3 increase in China, contributing 4.08–18.51 ppb a^{-1} in the North China Plain. The meteorological conditions decreased O_3 over China and could be more significant than anthropogenic emissions in some regions. In Pearl River Delta, for example, the contributions of meteorological conditions and anthropogenic emissions on O_3 were -1.29 and 0.81 ppb in 2013, respectively. CO_2 was critical in O_3 variations, especially in southern China, inducing an increase in O_3 on the southeast coast of China (0.28–0.46 ppb a^{-1}) and a decrease in the southwest and central China (-0.51–-0.11 ppb a^{-1}). Our study comprehensively analyzed O_3 variation across China from various perspectives and highlighted the importance of considering CO_2 variations when designing long-term O_3 control policies, especially in high vegetation coverage areas.

1 Introduction

O_3 is a strong oxidant detrimental to human health (Lu et al., 2020; Liu et al., 2018a) and the ecosystem (Monks et al., 2015; Wang et al., 2017). It is also an essential active specie of radiation, with up to 0.47 W/m^2 effective radiative forcing in 2019 (Ipcc, 2021). As it is vital to air quality and climate change, tropospheric O_3 has received extensive attention over the past decades (Duan et al., 2017; Li et al., 2019; Ashmore and Bell, 1991; Lu et al., 2018).

With the rapid development in China, O_3 concentrations have increased annually since the beginning of the 20th century (Liu and Wang, 2020a; Ma et al., 2016). Surface O_3 pollution has been a severe air quality issue in China (Verstraeten et al., 2016; Xu et al., 2018). In 2013, the Chinese government performed the Clean Air Action Plan to control air pollution, and the concentration of surface fine particles has reduced significantly (Wang et al., 2021; Zhai et al., 2019). However, surface O_3 concentrations keep increasing continuously in major urban areas of China, including the North China Plain (NCP), Fenwei Plain (FWP), Yangtze River Delta (YRD), Pearl River Delta (PRD), and the Sichuan Basin (SCB) (Wang et al., 2020; Wang et al., 2017; Yin and Ma, 2020; Shen et al., 2019; Zhao et al., 2018; Wang et al., 2009).

Recent studies suggested that regional meteorological conditions affect surface O_3 through different pathways (Jacob and Winner, 2009; Shen et al., 2016; Lin et al., 2008). Modeling studies have examined that O_3 is sensitive to temperature, humidity, wind speed, mixing height, and other meteorological conditions (Pfister et al., 2014; Sanchez-Ccoyllo et al., 2006). The temperature can change the chemical formation rate of O_3 (Lee et al., 2014). Precipitation reduces surface O_3 concentrations through wet removal (Fang et al., 2011). The elevated planetary boundary layer (PBL) height enhances upward movement, thus decreasing surface O_3 concentrations (Haman et al., 2014). Therefore, the long-term modeling of surface O_3 should involve changes in meteorological conditions.

CO_2 is the primary anthropogenic force on the climate system (Gauss et al., 2003; Schimel et al., 2015). CO_2 can adjust regional air temperature and precipitation, thus varying the surface O_3 concentrations (Lu et al., 2013; Yang et al., 2014). On the other hand, the BVOCs are the significant O_3 precursors, and isoprene is the primary specie among BVOCs that vegetation emits (Zheng et al., 2009; Fiore et al., 2011). In most of China, O_3 is VOCs-limited in the summer, especially in industrial cities (Li et al., 2018; Wu et al., 2018). Thus, it plays a significant role in modulating O_3 levels and positively correlates with O_3 concentrations in major urban areas of China.

It is known that CO_2 will enhance vegetation's photosynthesis (Sun et al., 2013; Heald et al., 2009; Tai et al., 2013; Monson and Fall, 1989). The elevated photosynthetic rate may directly increase the isoprene's emission (Rapparini et al., 2004). Based on the observation, Rosenstiel et al. (2003) found that the isoprene emissions of plants increased by about 21% and 41% when CO_2 reached 800 ppm and 1200 ppm, respectively. However, Wilkinson et al. (2009) indicated that different vegetation types show various responses in isoprene emission when CO_2 increases. The isoprene emission was decreased by 30–40% in Populus tremuloides Michx but increased by about 100% in Quercus rubra when CO_2 concentrations were grown (Sharkey et al., 1991). High concentrations of CO_2 may inhibit the emission of isoprene by reducing the activity of BVOCs synthetase or decreasing the synthesis of adenosine triphosphate (Possell et al., 2005). Guenther et al. (1991) indicated that isoprene emissions were significantly reduced when CO_2 was increased from 100 to 600 μmol mol^{-1}. To sum up, the impact of elevated CO_2 on isoprene emission may be positive or negative, mainly related to the relative size of inhibition caused by elevated CO_2 and promotion by enhanced photosynthesis.

Numerous studies have concluded that anthropogenic emissions dominate the surface ozone increases in different regions or years in China. Meanwhile, the effects of meteorological parameters should not be negligible (Wang et al., 2019b; Lu et al., 2019; Dang et al., 2021; Liu and Wang, 2020a). For example, Li et al. (2020) indicated that anthropogenic emissions are the main reason for the surface ozone increase in China from 2013 to 2019. Liu and Wang (2020a) suggested anthropogenic emissions play the dominant role in the ozone increase in China, but the effects of meteorological conditions could be more significant in some regions. Han et al. (2020) analyzed the ozone changes in summer and suggested that meteorology can explain about 43% of that in eastern China.

Most previous research focused on the effects of anthropogenic emission and meteorological factors on O_3 increase, ignoring the contributions of CO_2 variations. And CO_2 emission has kept annual increasing in China (Lv et al., 2020; Ren et al., 2014). Therefore, a comprehensive evaluation of the impact of anthropogenic emission, meteorological factors, and carbon dioxide on surface maximum daily 8 h average (MDA8) O_3 is necessary.

Here, we used an up-to-date regional climate-chemistry-ecology model to quantify the effect of anthropogenic emission, meteorological factors, and carbon dioxide variations during the summer monsoon period (May, June, July, and August), thereby benefitting the development of a comprehensive O_3 improvement strategy. Sect. 2 describes the methods and data, and the results and discussion are given in Sect. 3, finally, the conclusions are shown in Sect. 4.

2 Methods and data

2.1 Measurement data

Simulated regional meteorological factors were compared with the European Centre for Medium-Range Weather Forecasts Interim reanalysis data (ERA-Interim), including temperature, relative humidity, and wind speed, at 37 vertical levels (Balsamo et al., 2015; Hoffmann et al., 2019). The observed surface O_3 was taken from the China National Environmental Monitoring

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Center (CNEMC). There were more than 1400 environmental monitoring stations in 2018 (Wang et al., 2018; Kong et al., 2021; Zheng et al., 2014). The World Data Centre for Greenhouse Gases (WDCGG) data (Liu et al., 2009; Li et al., 2017) was applied to evaluate the simulated surface CO_2 concentrations. The monitoring stations of O_3 and CO_2 are shown in Figure. 1.

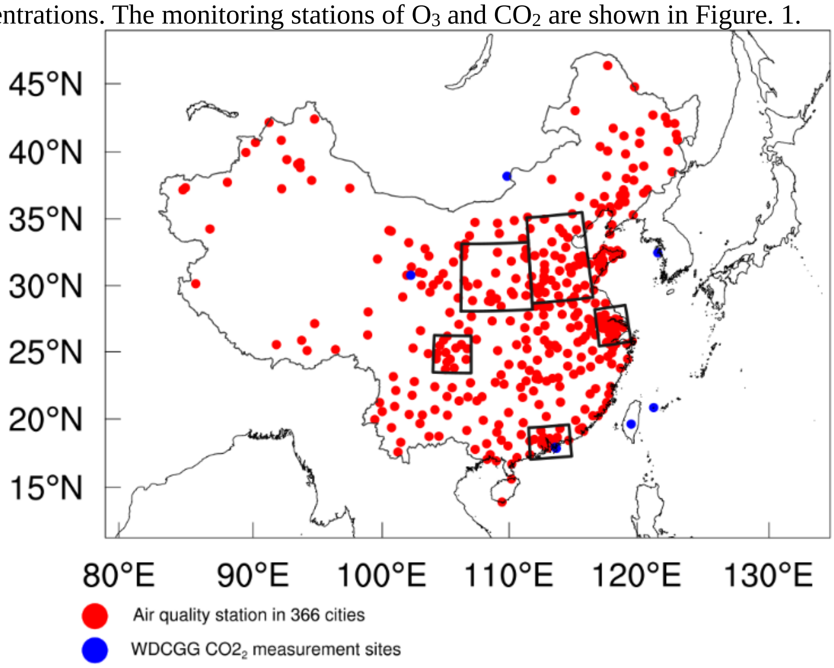

Figure 1. Model domains for the RegCM-Chem-YIBs model. The regions with black boundaries are the North China Plain (34–41◦ N, 113–119◦ E), the Yangtze River Delta (30–33◦ N, 119–122◦ E), the Pearl River Delta (21.5–24◦ N, 112–115.5◦ E), the Sichuan Basin (28.5–31.5◦ N, 103.5–107◦ E), and the Fenwei Plain (33.5–39◦ N, 106–113◦ E) regions.

2.2 Model description

The RegCM-Chem-YIBs is a regional climate-chemistry-ecology model developed by a team of Prof. Wang in the School of Atmospheric Sciences of Nanjing University (Xie et al., 2020; Xie et al., 2019), the International Center for Theoretical Physics (ICTP) in Italy (Giorgi et al., 2012; Giorgi and Mearns, 1999), and a team led by Prof. Yue in Nanjing University of Information Technology (Yue and Unger, 2015).

The ecological model (YIBs) was fully coupled into the regional climate-chemical model (RegCM-Chem) to reproduce the interactions between atmospheric composition and the ecosystem in the authentic atmosphere (Xie et al., 2019). The meteorological factors and the concentrations of air pollutants output by RegCM-Chem are delivered into YIBs to simulate the physiological processes of vegetation and calculate land surface parameters such as carbon dioxide flux, Biogenic volatile organic compound (BVOC) emissions, and stomatal conductance of the terrestrial ecosystem. Conversely, the simulations of the YIBs model are feedback into the RegCM-Chem model to adjust the air qualities, temperature, humidity, circulation, and other meteorological fields. The RegCM-Chem-YIBs has been widely used in surface O_3, $PM_{2.5}$, CO_2, the summer monsoon, and the interactions between air quality and the ecosystem (Zhuang et al., 2018; Pu et al., 2017; Xu et al., 2022b; Xie et al., 2018).

2.3 Model improvements

The previous version RegCM-Chem-YIBs model simulated the radiation effect that considers spatial-temporal variations of PM only. The air CO_2 and O_3 concentrations in the radiation module were constant in the year to calculate the radiation. We have taken simulated CO_2 and O_3 concentrations with a spatiotemporal variation into the radiation module and improved the associated radiation effects to better simulate the regional radiation balance.

Lefer et al. (2003) suggested that better aerosol optical parameter inputs, including aerosol optical depth (AOD) and single scattering albedo (SSA), played a significant role in the photolysis of O_3. We improved the calculation involving the AOD and SSA in the photolysis subroutine so that the extinction effect of the particles can be fed back to the photolysis reaction correctly. These improvements led to more realistic simulations in air components and regional meteorology.

2.4 Emissions and Experiment settings

Anthropogenic emissions from 2008 to 2018 were taken from the Multi-resolution Emission Inventory for China (MEIC), which was compiled and maintained by Tsinghua University since 2010 (Zheng et al., 2018; Wang et al., 2014). CO_2 emissions were derived from the NOAA CarbonTracker CT2019 dataset (Jacobs et al., 2021). The initial meteorological boundary data are obtained from the ERA-Interim reanalysis dataset (Liu et al., 2018b). The weekly mean Sea Surface Temperature dataset was supplied by the National Ocean and Atmosphere Administration (NOAA) (Reynolds et al., 2002). The Model of Ozone and Related Chemical Tracers (MOZART) model was chosen to drive climatological chemical boundary conditions. The boundary layer scheme was Holtslag PBL (Khayatianyazdi et al., 2021). The Grell cumulus convection scheme was employed (Grell, 1993). The CCM3 radiation scheme and CLM3.5 land surface module were used in the model (Collins et al., 2006; Giorgi and Mearns, 1999; Decker and Zeng, 2009).

The simulated domain is shown in Figure 1. The target region was centered at 36°N, 107°E, and the 60 km by 60 km grid resolution was applied. 18 vertical levels were set from the surface to 50 hPa.

The interannual changing anthropogenic emissions, meteorological fields, and CO_2 emissions were applied in the base experiment during the summer monsoon period from 2008 to 2018. The meteorological conditions were maintained at 2008 in all ten years, namely the $SIM_{i,m=2008}$ experiment.

The changes in O_3 concentrations from 2009 to 2018 relative to 2008 were obtained by comparing the simulations of different years with 2008 in the base experiment (Eq. (1)). The effect of meteorology in the O_3 was obtained by comparing the results between SIMi,m=2008 with the base experiment in the same year (Eq. (2)). Similarly, the effects of CO_2 emissions were derived (Eq. (3)). Finally, the influence of anthropogenic emissions was calculated by excluding the impact of meteorological factors and CO_2 from the changes in O_3 concentrations (Eq. (4)). The numerical experiments are shown in table 1.

$$\Delta O_i = SIM_i - SIM_{2008} \tag{1}$$

$$\Delta M_i = SIM_i - SIM_{i,M=2008} \tag{2}$$

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$$\Delta C_i = SIM_i - SIM_{i,C=2008} \quad (3)$$

$$\Delta E_i = \Delta O_i - \Delta M_i - \Delta C_i \quad (4)$$

ΔO_i : The changes of O_3 concentrations in the year i relative to 2008.
SIM_i : The simulated O_3 concentrations in the year i.
ΔM_i : The effect of meteorological conditions variations on O_3 in the year i relative to 2008.
$SIM_{i,M=2008}$: The meteorological conditions remained at 2008.
ΔC_i : The effect of CO_2 variations on O_3 in the year i relative to 2008.
$SIM_{i,C=2008}$: The CO_2 emissions remained at 2008.
ΔE_i : The effect of anthropogenic emissions changes on O_3 in the year i relative to 2008.

Table 1. The Numerical experimental in this study.

Experiment	Time	Description
Base	2008-2018	Interannual changing anthropogenic emissions, meteorological fields, and CO_2 emissions
SIM_MET08	2009-2018	Meteorological conditions remained at 2008
SIM_CO$_2$08		CO_2 emissions remained at 2008

3 Results and discussion

3.1 Model evaluation

The ability of RegCM to reproduce East Asian climate and air quality has been widely evaluated in recent years. Overall, the RegCM can reasonably simulate the essential characteristic and interannual variations of air components and meteorological fields in East Asia. (Xu et al., 2022a; Xie et al., 2019; Zhuang et al., 2018; Tesfaye et al., 2015; Pu et al., 2017; Li et al., 2021). Here, for more confidence, the meteorological fields, O_3 and CO_2 are compared between simulations and observations in 2018.

Figure 2 shows the RegCM-Chem-YIBs model has well captured the spatial distribution and magnitude of temperature, humidity, and wind over East Asia at 850 hPa and 200 hPa. The model underestimated the temperatures and wind speed slightly at 850 hPa. In contrast, the relative humidity was overpredicted by about 10% at 850 hPa. Due to the influences of complex terrain on the lower atmosphere, most models show better results at higher levels (Zhuang et al., 2018; Anwar et al., 2019; Xie et al., 2019). Therefore, the simulations at 200 hPa are closer to the reanalysis data.

The evaluations of surface O_3 and CO_2 in East Asia are shown in Table 2. The RegCM-Chem-YIBs model reality reproduced surface O_3 and CO_2 concentrations during the East Asia summer monsoon season, with high correlation coefficients (0.73 for O_3 and 0.41 for CO_2). The overpredicted MDA8 O_3 (1.73%) and CO_2 (6.57%) were mainly driven by the uncertainty of the emissions inventory (Wang et al., 2014; Hong et al., 2017; Zhang et al., 2014). The normalized mean bias was 6.63% and 1.73%, respectively. Therefore, the simulated meteorological factors and surface O_3 and CO_2 were acceptable.

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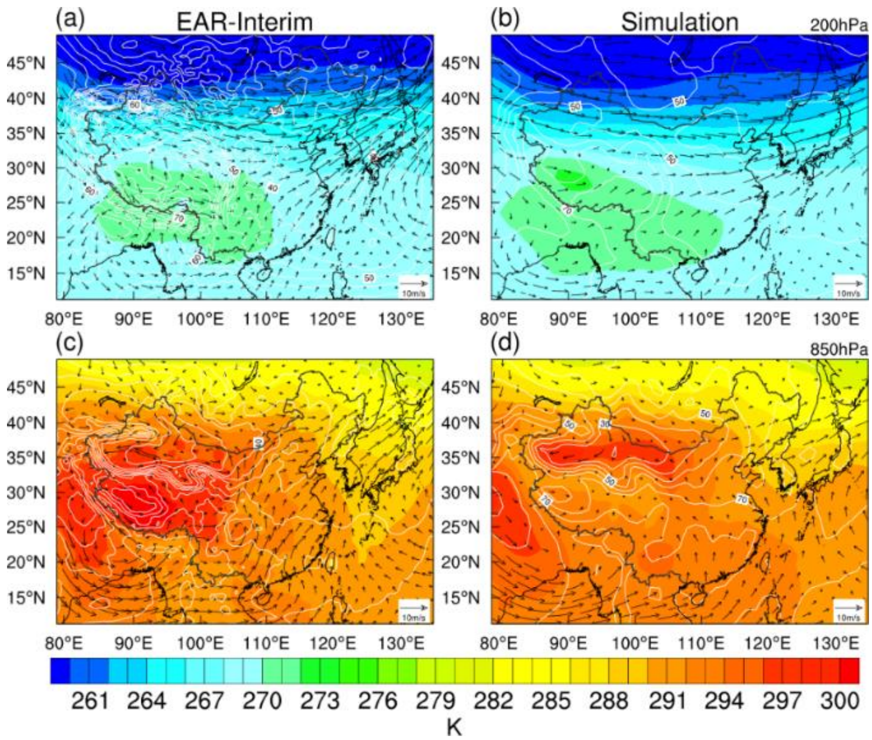

Figure 2. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 200 hPa (a, b) and 850 hPa (c, d).

Table 2. Evaluations of the surface CO_2 and MDA8 O_3 in East Asia.

Species	OBS	SIM	MB	NMB (%)	RMSE	R
CO_2（ppm）	409.61	416.68	7.07	1.73	11.32	0.41
MDA8O_3（ppb）	52.08	55.53	3.42	6.63	24.78	0.73

OBS: observation; SIM: simulation; MB: bias; NMB: normalized mean bias; RMSE: root mean square error; R: correlation coefficient.

3.2 Ozone variation from 2008 to 2018

Figure 3 shows the seasonal mean MDA8 O_3 concentrations over East Asia in the summer monsoon period from 2008 to 2018. High O_3 concentrations appeared in eastern China, related to elevated emissions and intense radiation in the region (Gao et al., 2020; Mousavinezhad et al., 2021). The surface O_3 increased annually in most of China from 2008 to 2018, especially in megacity clusters.

We analyzed the surface O_3 increase in five target regions: the NCP, the YRD, the PRD, the SCB, and the FWP. The surface MDA8 O_3 concentrations averaged 74 ppb across the NCP

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region in 2018 and were lower in the other areas (ranging from 42 to 67 ppb in FWP, YRD, PRD, and SCB). Less anthropogenic emissions induced lower surface O_3 levels in SCB and FWP regions. The YRD and PRD regions were more susceptible to meteorological factors. The East Asian summer monsoon (EASM) brings cleaner air and precipitations from the sea to the areas, inducing lower air pollution concentrations (He et al., 2012). The spatial distribution and increasing trend of the surface MDA8 O_3 in China were consistent with Li et al. (2020) and Shen et al. (2022).

Figure 4 shows the changes in surface MDA8 O_3 from 2009 to 2018 relative to 2008, which were calculated by comparing the simulations in different years with 2008 in the base experiment. The surface MDA8 O_3 has increased drastically in China in the past decade, especially in 2017 and 2018. Based on the Clean Air Action Plan performed in 2013, we divide 2009 to 2018 into two phases: the pre-governance period (PreG, 2009-2013) and the post-governance period (PostG, 2014-2018). Table 5 shows the surface MDA8 O_3 concentration increased significantly in NCP (18.42 ppb a^{-1}), followed by SCB (11.21 ppb a^{-1}), FWP (10.9 ppb a^{-1}), and the YRD (10.07 ppb a^{-1}), while increased slightly in PRD (4.94 ppb a^{-1}), in PosG relative to 2008. Our results agree with Lu et al. (2020), Ma et al. (2016), and Mousavinezhad et al. (2021).

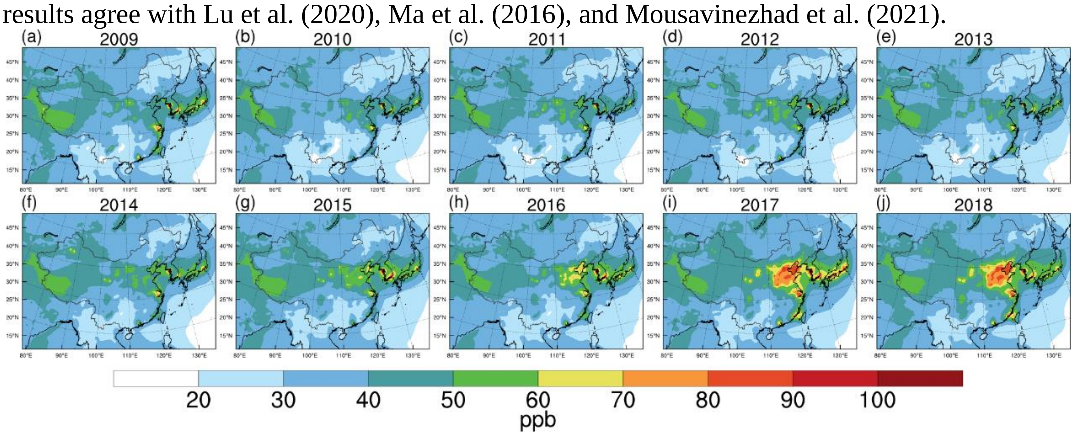

Figure 3. Simulated surface MDA8 O_3 concentrations in the summer monsoon period of 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j).

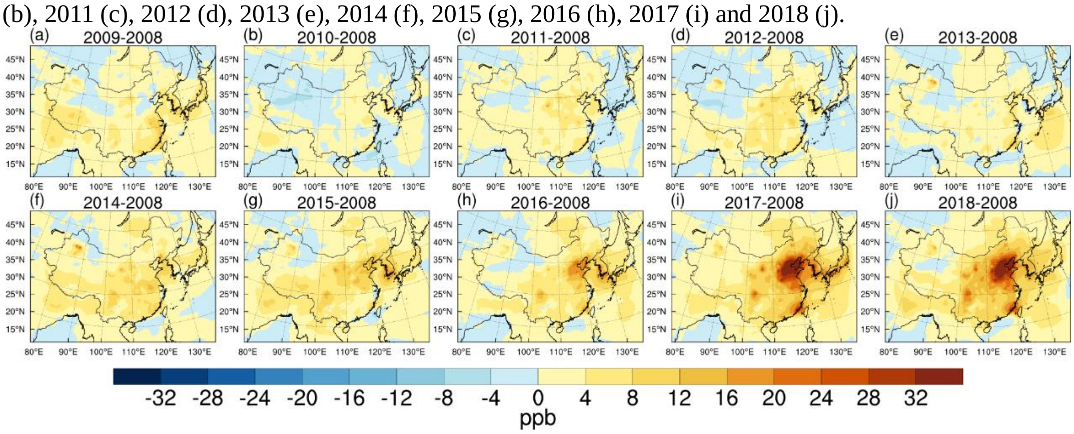

Figure 4. Changes in the surface MDA8 O_3 concentrations from 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

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3.3 The effect of meteorology in 2008–2018 ozone increase

The meteorological factors were generally unfavorable to O_3 formation during the study period (Fig. 5).

In the NCP and FWP regions, the effect of meteorological conditions on surface MDA8 O_3 decreased sharply in PostG (-0.09–-0.04 ppb a^{-1}) compared with PreG (-1.41–-0.88 ppb a^{-1}). In the SCB region, the influence of meteorological fields was weak due to the basin topography and stable atmospheric conditions (-0.41–0.71 ppb a^{-1}). But in the east and southeast coastal areas of China, due to the significant influence of the EASM, the impact of meteorological conditions may be more critical than anthropogenic emission. For example, in the YRD and PRD regions, the meteorological conditions significantly changed O_3 (-1.29–1.3 ppb a^{-1}) than that of anthropogenic emission (0.81–0.87 ppb a^{-1}) in 2013. This reflected the significant influence of meteorological conditions on surface O_3.

Our results agree with Liu and Wang (2020a), who show that meteorological variations induced the decrease of O_3 in Shanghai from 2013 to 2017. Chen et al. (2019) and Liu and Wang (2020a) indicated meteorology factors are unfavorable to O_3 formation in NCP and FWP, and the influence of meteorology on surface O_3 decreased in PostG. Cheng et al. (2019) indicated that meteorological conditions' effects on long-term O_3 variations were lower than 3 %, similar to our study.

The individual variations of MDA8 O_3, precipitation, clouds, shortwave flux (SWF), wind speed, temperature, and PBL height are shown in Figure 6. The increased SWF can accelerate ozone formation via photochemistry (Jiang et al., 2012; Lelieveld and Crutzen, 1990). Therefore, the increased cloud fraction reduced surface ozone by decreasing shortwave radiation, especially in NCP, FWP, YRD, and SCB in the PreG period (-10.63–-1.6 W/m^2). In addition, the precipitation was enhanced in NCP, FWP, YRD, and SCB (0.37–1.81 mm/day) and then decreased surface O_3 levels significantly. The significant increase in wind speed (0.17–0.26 m/s) also reduced surface O_3 in NCP.

Another important reason, the warmer surface (0–5 K) strengthened the upward movement and increased the PBL height (0–500 m) in most of East Asia. Therefore, the higher temperature and PBL height can dilute surface O_3.

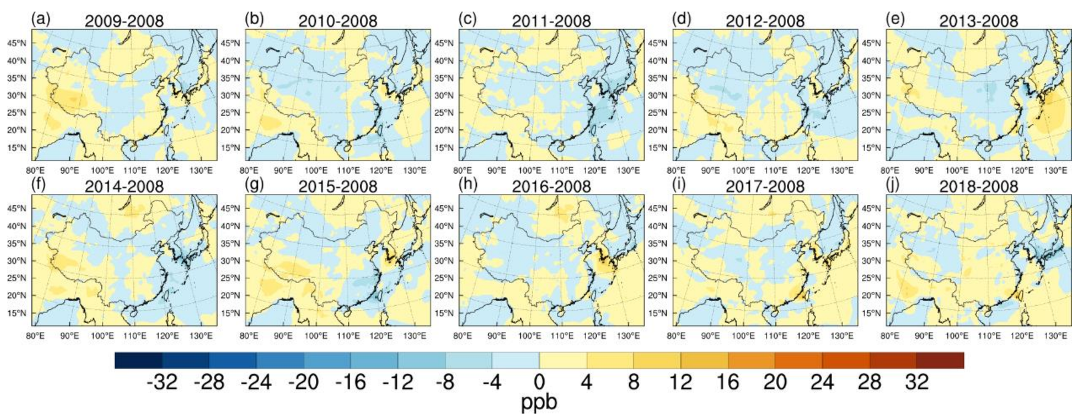

Figure 5. The responds of the surface MDA8 O_3 mixing ratios to variations in meteorological conditions

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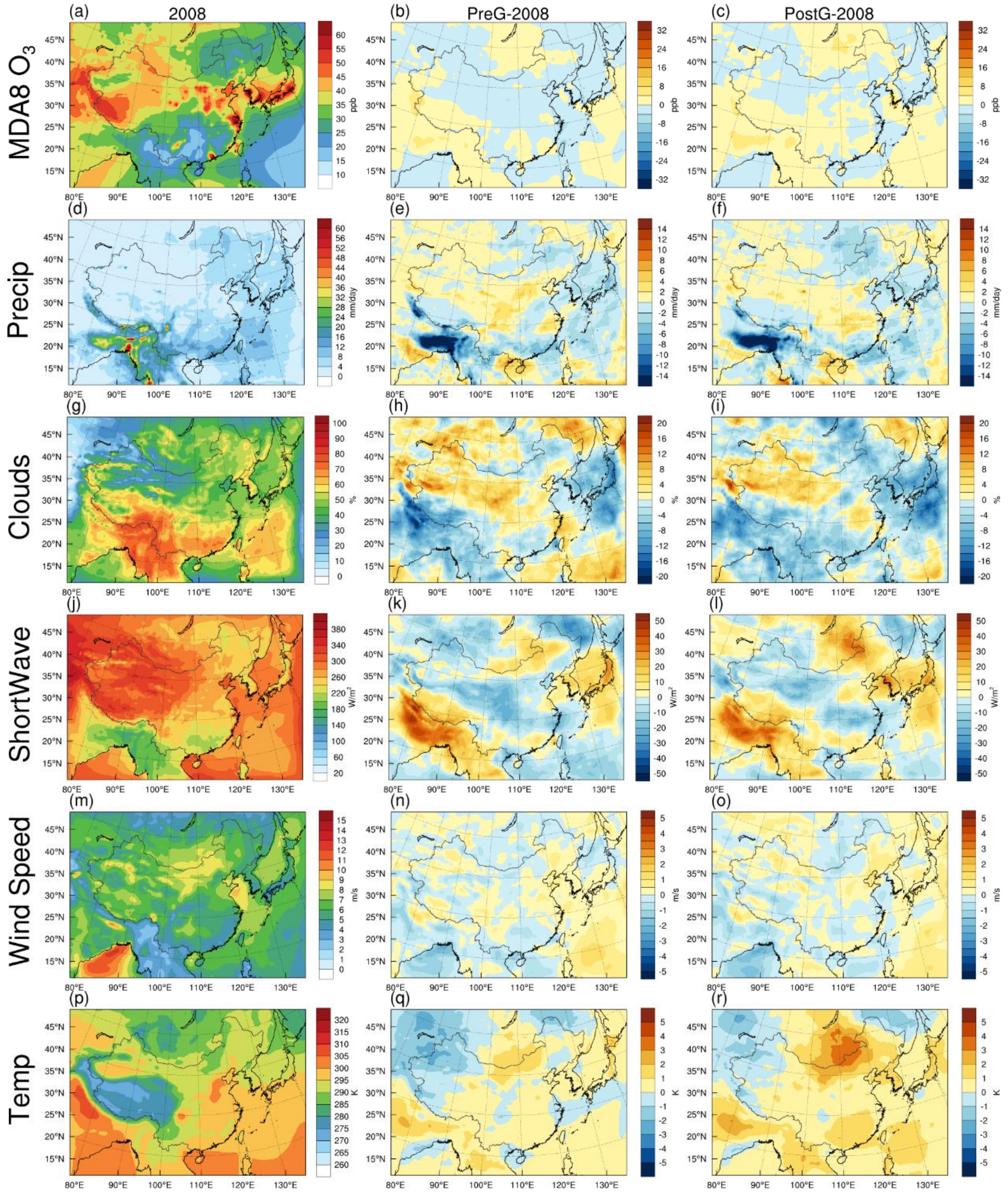
in 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.



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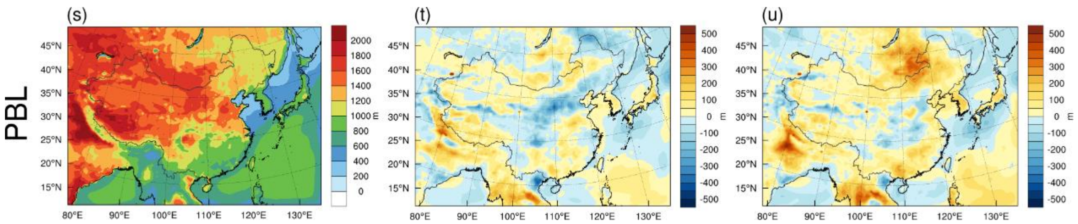

Figure 6. The MDA8 O_3 (a–c), precipitation (d-f), clouds (g-i), shortwave flux (j-l), wind speed (m-o), temperature (p-r), and planetary boundary layer (s–u) in 2008 from the base simulations (the left column) and their responses due to variations in meteorological conditions in PreG (the central column) and PostG (the right column) relative to 2008.

Table 3. Response of the MDA8 O_3 mixing ratios, precipitations, clouds, shortwave flux, wind speed, temperature, and planetary boundary layer to the changes in meteorological conditions over North China Plain, Fenwei Plain, Yangtze River Delta, Pearl River Delta, and Sichuan Basin in PreG and PostG relative to 2008.

Regions	Period	MDA8 O3	Precip	Clouds	SWF	Wind Speed	Temp	PBL
NCP	PreG	-0.88	0.58	1.33	-3.04	0.17	0.32	-46.8
	PostG	-0.04	0.6	-0.93	3.06	0.26	0.6	-14.5
FWP	PreG	-1.41	1.68	2.86	-10.63	-0.06	0.1	-108.5
	PostG	-0.09	0.81	-0.94	-0.81	0.05	0.46	-15.3
YRD	PreG	-1.03	1.02	1.07	-1.6	0.18	-0.29	-33.9
	PostG	-0.96	0.48	-1.18	-4.85	-0.08	0.45	21.9
PRD	PreG	-0.23	-2.39	-1.93	2.24	-0.02	0.36	29.6
	PostG	-1.08	-3.24	-3.98	5.37	0.18	1.00	52.2
SCB	PreG	-0.41	1.81	0.59	-8.8	0.13	-0.58	-136.5
	PostG	0.71	0.37	-2.23	-3.2	-0.03	-0.14	-76

3.4 The effect of CO_2 in 2008–2018 ozone increase

CO_2 is a significant driver of climate and biogenic emissions. The effect of CO_2 on O_3 was 0.5–2 ppb on the southeast coast of China and -0.5–-2 ppb in southwest and central China (Figures 8 b and c).

Figure 8 e and f show the increasing trend of CO_2 over East Asia, especially in eastern China, which was related to intensive human activities. The CO_2 significantly modulated the surface ozone in southern China (YRD, PRD, and SCB regions), with high precipitations, temperatures, and vegetation covers.

In YRD, the surface MDA8 O_3 was reduced by 0.09–0.14 ppb a^{-1} via the decreased isoprene. In contrast, the isoprene was increased by the elevated CO_2 concentrations on the southeast coast of China and then promoted the formation of surface O_3 (1–3 ppb) (Figure 8 b and c). In PRD, for example, the increased isoprene (0.31–0.92 $\mu g/m^3$ a^{-1}) resulted in MDA8 O_3 enhanced by 0.28 to 0.46 ppb a^{-1}. Due to the lower temperature and vegetation cover, surface O_3 was insensitive to air CO_2 in the NCP.

As the foremost anthropogenic forcing of radiation, the increased CO_2 modified other aspects of meteorology, including temperature, cloud fraction, and precipitations (Bala et al., 2010; Cao et al., 2012). And that was going to vary the surface O_3 concentrations. For instance, precipitation increased by 0.06 to 0.64 mm/day in the NCP, FWP, YRD, and SCB, where surface

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O_3 was reduced. In contrast, on the southeast coast of China (PRD), reduced precipitation accelerated the accumulation of MDA8 O_3 (0.28 to 0.46 ppb a^{-1}).

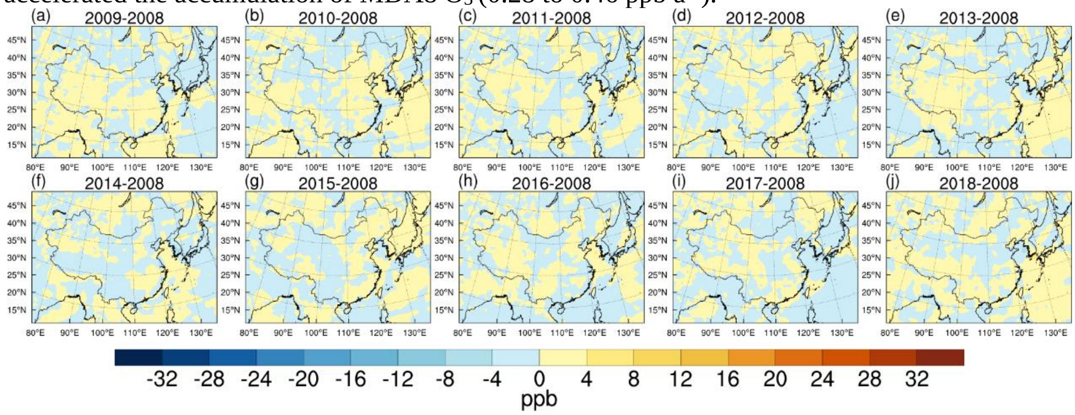

Figure 7. Simulated responses of surface MDA8 O_3 mixing ratios to the variations in CO_2 emissions in 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

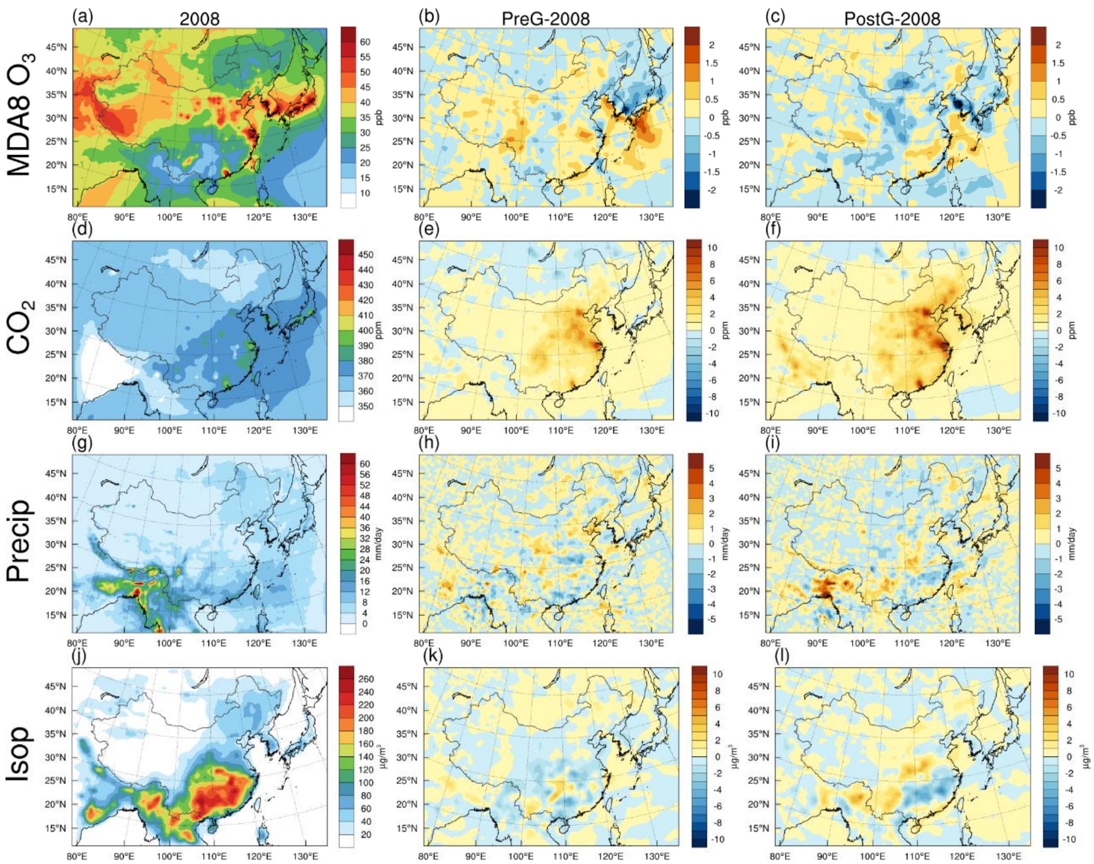

Figure 8. The simulated averaged MDA8 O_3 (a–c), CO_2 (d-f), precipitation (g-i), and isoprene mixing

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ratios (j-l) in 2008 from the base simulations (the left column) and their changes due to variations in CO_2 emissions in PreG (the central column) and PostG (the right column) relative to 2008.

Table 4. Simulated responses of MDA8 O_3 mixing ratios, CO_2 mixing ratios, precipitations, and isoprene mixing ratios to the changes in CO_2 emissions over North China Plain, Fenwei Plain, Yangtze River Delta, Pearl River Delta, and Sichuan Basin in PreG and PostG relative to 2008.

Regions	Period	MDA8 O_3	CO_2	Precipitation	Isoprene
NCP	PreG	0.07	3.19	0.27	-0.1
	PostG	-0.05	4.24	0.13	0.26
FWP	PreG	-0.11	1.70	0.21	-0.16
	PostG	-0.51	2.05	0.06	0.33
YRD	PreG	-0.09	4.1	0.13	-0.32
	PostG	-0.14	6.2	0.09	-0.58
PRD	PreG	0.46	1.97	-1.02	0.31
	PostG	0.28	3.20	-0.33	0.92
SCB	PreG	-0.30	2.80	0.64	-0.78
	PostG	-0.30	2.78	0.21	0.69

3.5 The effect of anthropogenic emission in 2008–2018 ozone increase

Finally, we calculated anthropogenic emissions' effect on the 2008–2018 ozone increase. Figure 9 shows that anthropogenic emissions induced a significant surface O_3 increase in most of China, especially in megacity clusters. The effect of anthropogenic emissions on O_3 was 2.33 to 18.51 ppb a^{-1} in five target regions.

It can be seen in Figure 10 that the O_3 variations caused by anthropogenic emissions show the same magnitude and spatial distribution as the changes in the base experiment. The contributions of meteorological factors and CO_2 were relatively low (-4–8 ppb). The effect of CO_2 could be compared with that of meteorology in magnitude. Therefore, anthropogenic emissions dominated the increase of surface O_3 in China from 2008 to 2018 (Figure 10), and CO_2 played a significant role in O_3 changes.

A high-impact center of anthropogenic emissions was found in North China (Figure 10). Surface O_3 showed a significant increase in the NCP region (4.08–18.51 ppb a^{-1}), followed by the FWP, YRD, and SCB regions (4.10–11.5 ppb a^{-1}). The O_3 was enhanced by 2.33–5.74 ppb a^{-1} in the PRD region due to the slight increase in air O_3 concentration. The role of anthropogenic emissions increased linearly from 2008 to 2018, despite the Clean Air Action Plan being performed in 2013 (Figure 11). For example, anthropogenic emissions significantly increased surface MDA8 O_3 in NCP (4.08 ppb a^{-1} in PreG and 18.51 ppb a^{-1} in PostG). Similarly, FWP experienced 5.15 and 11.5 ppb a^{-1} increases in PreG and PostG periods, respectively. In the SCB region, the surface MDA8 O_3 was mainly affected by anthropogenic emissions variations due to high anthropogenic emissions in the complex basin topography. In YRD and PRD, the anthropogenic emissions resulted in changes to O_3 of 2.56–10.07 ppb a^{-1}. The reasons for this characteristic are multiplied. After aerosol reduction, the elevated photochemical formation rate significantly elevated surface O_3 level, especially in the PostG period (Bian et al., 2007). The reduced NO emission weakened the titration effect, thus increasing surface O_3 (Li et al., 2022).

Our results agree with Wang et al. (2019a) and Liu and Wang (2020b), indicating the dominant and almost linear increasing role of anthropogenic emission in the ozone increase from

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2013 to 2015 in four cities (Beijing, Shanghai, Guangzhou, and Chengdu).

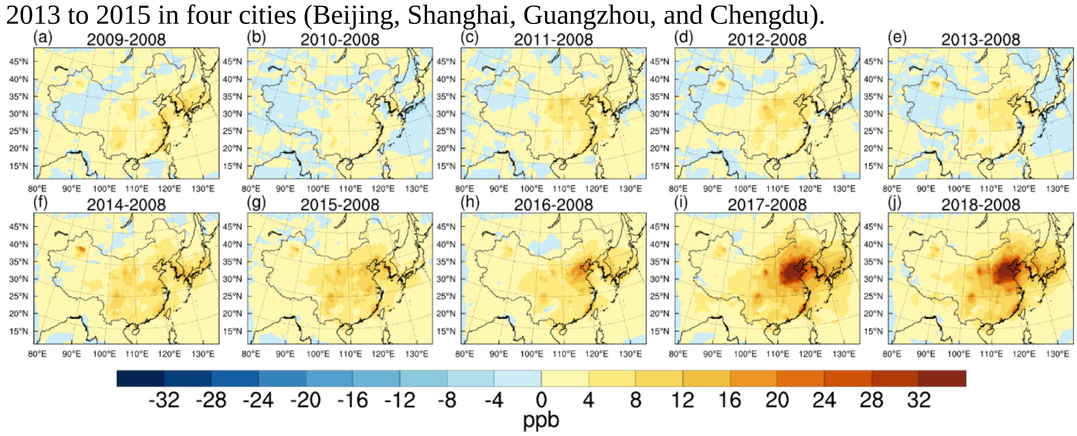

Figure 9. Simulated responses of the surface MDA8 O_3 mixing ratios to variations in anthropogenic emissions in 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

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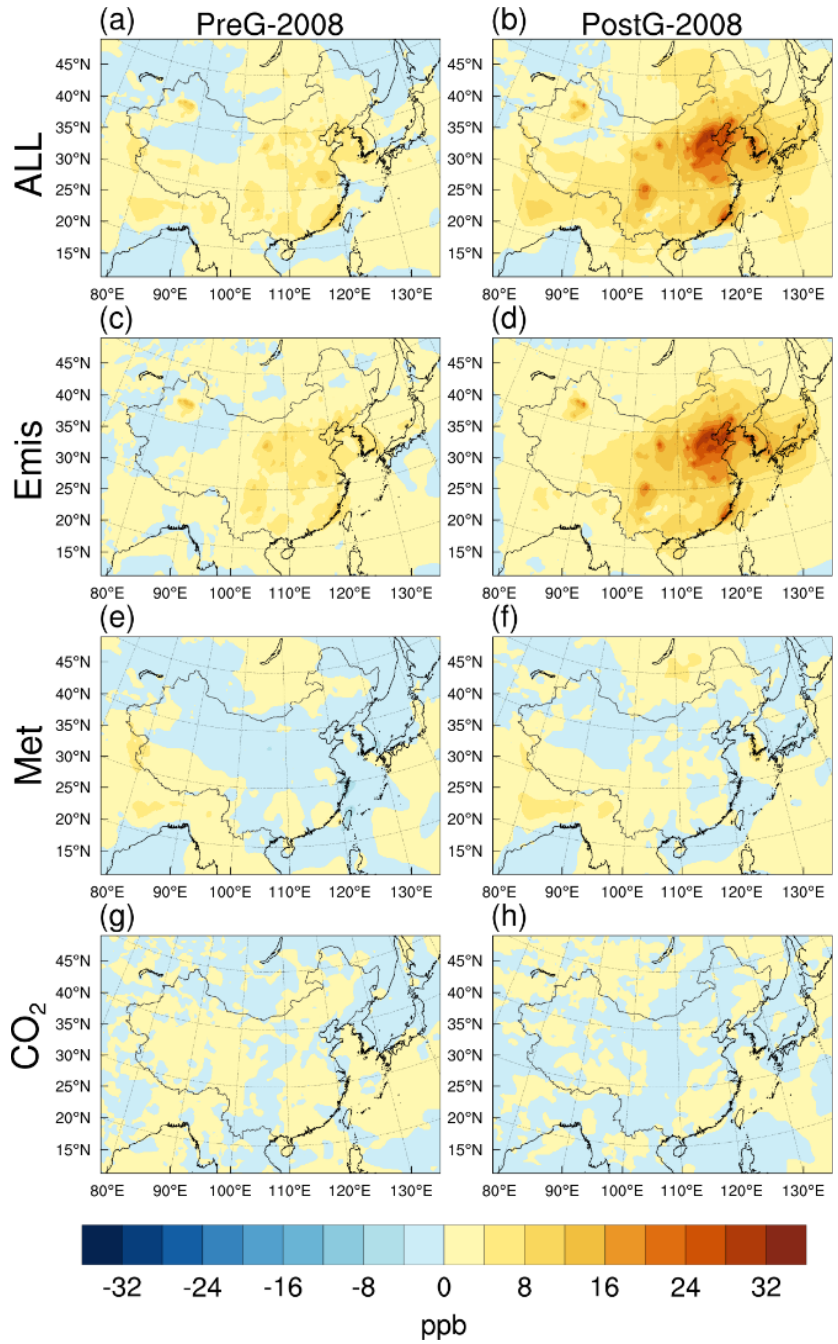

Figure 10. Changes in the simulated surface MDA8 O_3 mixing ratios from the base simulation (All, a,b); those due to variations in anthropogenic emissions (Emis, c,d), meteorological conditions (Met, e,f), and CO_2 emissions (CO_2, g,h) in PreG (the left column) and PostG (the right column) relative to 2008.

Table 5. Simulated response of the MDA8 O_3 mixing ratios to the changes in anthropogenic emissions

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(Emis), meteorological conditions (Met), and CO_2 emissions (CO_2) over North China Plain, Fenwei Plain, Yangtze River Delta, Pearl River Delta, and Sichuan Basin in PreG and PostG relative to 2008.

Regions	Period	ALL	Emis	Met	CO_2
NCP	PreG	3.27	4.08	-0.88	0.07
	PostG	18.42	18.51	-0.04	-0.05
FWP	PreG	3.63	5.15	-1.41	-0.11
	PostG	10.9	11.5	-0.09	-0.51
YRD	PreG	2.98	4.10	-1.03	-0.09
	PostG	10.07	11.17	-0.96	-0.14
PRD	PreG	2.56	2.33	-0.23	0.46
	PostG	4.94	5.74	-1.08	0.28
SCB	PreG	3.67	4.38	-0.41	-0.30
	PostG	11.21	10.80	0.71	-0.30

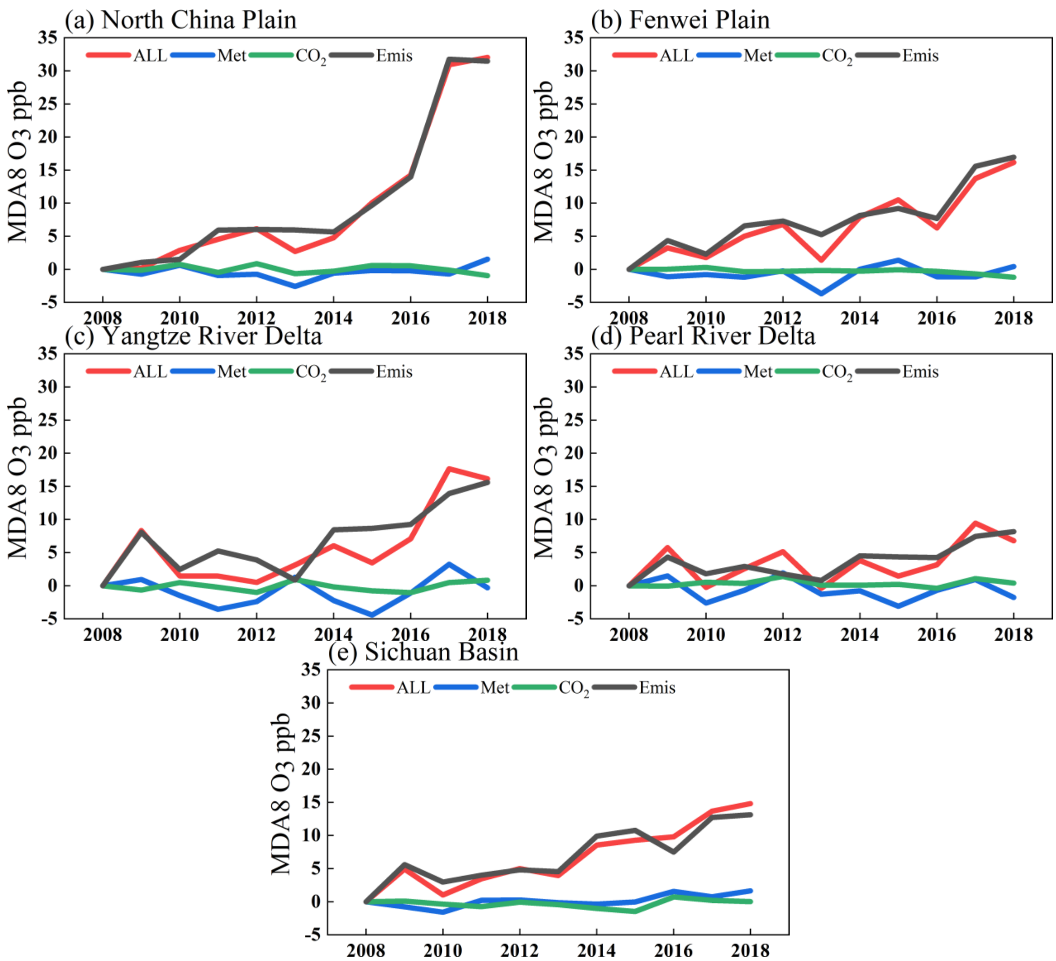

Figure 11. Interannual variations of the surface MDA8 O_3 mixing ratios in the summer monsoon period (ALL) and the responses of variations in anthropogenic emissions (Emis), meteorological conditions

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(Met), and CO_2 emissions (CO_2) in (a) North China Plain, (b) Fenwei Plain, (c) Yangtze River Delta, (d) Pearl River Delta, and (e) Sichuan Basin in 2008–2018 relative to 2008.

3.6 Uncertainty analysis

In this work, the boundary conditions are kept consistent in base and sensitivity studies. We ignore the influence of boundary conditions on ozone due to the following reasons. First, in general, the regional model was coupled with the global model to get a more realistic influence from the boundary. However, for long-term climate-chemistry modeling, such coupling means a large computing resource. Second, the boundary conditions were derived from global models (Liu et al., 2017; Ban et al., 2014) and have to be prescribed in sensitive experiments. Third, fixed boundary conditions are widely used in some O_3 studies in China (Liu and Wang, 2020a, b; Wang et al., 2019a). Regional emissions are the major contributors of surface O_3 in China, accounting for 80% from May to August (Lu et al., 2019). Therefore, the uncertainty of fixed boundary conditions can be ignored at the current stage.

4 Conclusions

In this study, we estimated the effects of anthropogenic emission, meteorological factors, and CO_2 on the surface ozone increase in China from 2008 to 2018 during the EASM season. Overall, anthropogenic emissions played the dominant role in surface O_3 increasing from 2008 to 2018. The meteorological factors induced the decrease of surface O_3 concentrations and could be more significant than anthropogenic emissions in some regions. CO_2 was critical in O_3 variations, especially in high vegetation coverage areas.

In the NCP and FWP regions. The increased surface O_3 (4.08–5.15 ppb a^{-1} in PreG and 11.5–18.51 ppb a^{-1} in PostG) was primarily attributed to the changes in anthropogenic emissions. And the impact of anthropogenic emissions has increased linearly despite the Clean Air Action Plan being performed in 2013. By contrast, the effect of meteorological factors and CO_2 in O_3 was weak during the study period.

In the YRD and PRD regions. Ignoring the principal contributions of anthropogenic emissions, CO_2 significantly impacted the O_3 variations (-0.14–0.46 ppb a^{-1}). The varied CO_2 led to surface MDA8 O_3 changes of -0.09–-0.14 ppb a^{-1} in the YRD and 0.28–0.46 ppb a^{-1} in the PRD by modulating the isoprene emissions and precipitations. On the other hand, the meteorological conditions played a more significant role in surface O_3 than in NCP, FWP, and SCB regions, resulting in a decrease in MDA8 O_3 from 2008 to 2018 (-4.42–3.25 ppb a^{-1}).

In the SCB region. The variations of anthropogenic emissions dominated the increase of surface MDA8 O_3 likewise from 2008 to 2018. The effect of meteorological conditions was weak in PreG (-0.41 ppb a^{-1}) and PostG (0.71 ppb a^{-1}) periods due to the high emission and basin topography. However, the changes in CO_2 significantly affected surface O_3 levels and were unfavorable to O_3 formation during the study period (-3.0 ppb a^{-1} in PreG and PostG).

In summary, anthropogenic emissions were dominant in the O_3 increase in China from 2008 to 2018, and the effects of meteorological conditions on surface O_3 could be more significant in some regions. Meanwhile, we highlight the critical contributions of CO_2 emissions, especially in southern China. Therefore, it is necessary to consider CO_2 variability when predicting future O_3 concentrations. Such consideration would be helpful for designing long-term O_3 control policies.

Data availability

ERA-Interim data are available at https://apps.ecmwf.int/datasets/data/interim-full-daily/. MEICv1.3 data are available at http://meicmodel.org/?page_id=560. CarbonTracker data are available at https://gml.noaa.gov/aftp/products/carbontracker/co2/CT2019/. OISST data are available at https://downloads.psl.noaa.gov/Datasets/noaa.oisst.v2/. WDCGG CO_2 data are available at https://gaw.kishou.go.jp/search/gas_species/co2/latest/. CNEMC data are available at http://www.cnemc.cn/. only available in Chinese, last access 1 May 2022.

Author contributions

DM: performed experiments; **TW:** designed the overall research; **HW, YQ, JL, JaL, and SL** reviewed and edited the manuscript; **BL, ML, and MX** contributed to the development of the RegCM-Chem-YIBs model.

Competing interests:

The contact author has declared that none of the authors has any competing interests.

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