Response to Reviewers

No.: ACP-2022-850

Title: 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

Anonymous referee #1:

In this study, the authors used an up-to-date regional climate-chemistry-ecology model to quantify the effect of anthropogenic emission, meteorological factors, and carbon dioxide variations on O_3 variation across China and highlighted the importance of considering CO_2 variations. I suggest this article be published with some modifications to improve the clarity of some details and ambiguous presentation. My comments are listed below.

Response: We thank referee #1 for careful reading and valuable comments. We have responded to each specific comment in blue below. Please note that the line numbers given below refer to the clean version of the manuscript.

1. The main innovation is the emphasis on the role of CO_2, but this only occupies a small part of this study. Other effects such as meteorology and emissions have been extensively discussed in previous studies and the authors need to elaborate more on the significance of this study.

Response: Thanks. We have added some discussions on this aspect.

Changes in manuscript:

Abstract (L25~28): "Changed CO_2 played a critical role in the variability of O_3 through radiative forcing and isoprene emissions, particularly in southern China, inducing an increase in O_3 on the southeast coast of China (0.28~0.46 ppb) and a decrease in the southwest and central China (-0.51~-0.11 ppb)."

Introduction (L88~96): "Previous studies have mainly focused on the impact of anthropogenic emissions and meteorological factors on the rise of O_3 levels, with limited attention given to the role of CO_2 variations. However, due to the rapid socioeconomic growth in China and the subsequent surge in energy consumption, CO_2 emissions, and concentrations have also increased significantly, particularly in the eastern coastal region (Lv et al., 2020; Ren et al., 2014). Furthermore, given the significant impact of CO_2 on O_3, it is crucial to evaluate the influence of changes in CO_2 concentration on the maximum daily 8-hour average (MDA8) O_3 concentrations at the surface. Thus, a comprehensive assessment of the impact of anthropogenic emissions, meteorological factors, and CO_2 on surface O_3 is imperative."

Section 3.4 (L386~393): "In some years, the impact of changed CO_2 can be as significant as or even surpass that of anthropogenic emissions and meteorology (Figure 10). For example, in 2013, CO_2 caused an increase of 0.95 ppb in MDA8 O_3

in the YRD region, which exceeded that of anthropogenic emissions (0.87 ppb). Similarly, in the PRD region in 2012, the effect of CO_2, anthropogenic emissions, and meteorology was 1.41, 1.77, and 1.95 ppb, respectively. Even in the NCP in 2010, the impact of CO_2 (0.75 ppb) was comparable to that of anthropogenic emissions (1.5 ppb). In summary, CO_2 has a significant impact on surface O_3 concentrations by influencing radiation and isoprene emissions, with more prominent effects in regions with abundant vegetation."

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2. Line 98-100: Why did you choose ERA-Interim data to evaluate meteorological variables simulation instead of using observations?

Response: Thanks. Firstly, observations are considered as the ground truth for meteorological variables and are essential for validating model performance. However, their usefulness in evaluating models is often limited due to their sparse spatial and temporal coverage (Wang et al., 2021). In contrast, reanalysis data, such as ERA-Interim, is a gridded dataset that offers high spatial and temporal resolution with global coverage. It is derived by assimilating observations into a numerical weather prediction model, resulting in a more consistent dataset in both space and time compared to observations (He et al., 2020; Lindsay et al., 2014).

Secondly, reanalysis data can provide a comprehensive set of variables that are not always available from observations. For instance, ERA-Interim includes a wide range of meteorological variables such as wind speed, temperature, precipitation, wind vectors, radiation fields, cloud properties, soil moisture, and relative humidity. These variables are produced by incorporating the observation fields, forecast model, and a four-dimensional variational assimilation system (4D-VAR). Furthermore, ERA-Interim conducts a completely automated bias correction after a series of quality control and blacklist data selection (Balsamo et al., 2015; Nogueira, 2020; Rivas and Stoffelen, 2019).

On the whole, while observations are crucial for model validation, reanalysis data, such as ERA-Interim, provides a more complete and consistent dataset that can be used to evaluate model performance in a variety of contexts. Consequently, the use of reanalysis data to evaluate model performance has become increasingly prevalent in recent years (Pu et al., 2017; Xu et al., 2022; Zhou and Wang, 2016; Liu et al., 2023). In our study, we rely on ERA-Interim data to evaluate meteorological variables simulation as it provides a long-term record (2015-2018) of these variables at various altitudes (1000, 850, and 200 hPa), and it is derived by assimilating observations into a numerical weather prediction model.

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3. Line 134-139: Can you give more descriptions of the model improvement?

Response: Thanks. We added some descriptions of the model improvement.

Changes in manuscript:

2.3.1 Radiation (L156~168)

"In the previous version of the RegCM-Chem-YIBs model, radiative calculations only accounted for changes in the spatiotemporal distribution of particulate matter. To simplify the radiation calculations, the atmospheric CO_2 and O_3 concentrations were assumed to be constant throughout the year. However, atmospheric CO_2 and O_3 are subject to modulation by various sources, sinks, physical processes, and chemical processes (Ballantyne et al., 2012; Wang et al., 2019a). Additionally, rapid urbanization in China has led to an annual increase in CO_2 and O_3 concentrations (Guan et al., 2021; Wei et al., 2022), with elevated concentrations and growth rates primarily distributed in the eastern regions where urbanization is most prominent (Shi et al., 2016; Wang et al., 2017b). To more accurately simulate the atmospheric radiation balance and East Asian monsoon climate, it is necessary to incorporate spatiotemporal variations of CO_2 and O_3 concentrations into the radiation module. Therefore, we included the varying CO_2 and O_3 concentrations simulated by the model in the radiation module to calculate the corresponding radiative forcing."

2.3.2 Photolysis (L170~181)

"The photolysis process was simulated using the Tropospheric Ultraviolet and Visible (TUV) model, which is commonly used to compute photolysis rates in various models (Tie et al., 2003; Shetter et al., 2002; Borg et al., 2011). The TUV model employs input parameters such as zenith angle, altitude, ozone column, SO_2 column, NO_2 column, aerosol optical depth (AOD), single scattering albedo (SSA), and albedo, among others, to calculate photolysis rates (Singh and Singh, 2004). However, in the TUV module of the RegCM-Chem-YIBs model, AOD and SSA were held constant. This is problematic as accurate aerosol optical parameters, such as AOD and SSA, play a crucial role in the photolysis of O_3 (Lefer et al., 2003). To address this issue, we incorporated temporally and spatially varying AOD and SSA simulated by the RegCM-Chem-YIBs model into the photolysis rate calculations in the TUV module. This enabled us to accurately incorporate the extinction effect of the varying particles into the photolysis reaction, leading to more realistic simulations of air components and regional meteorology."

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4. Line 162: “i,m=2008” should be the subscript.
Response: Thanks. Sorry for the mistake. We have revised.

5. Section 3.1: Why did you only compare simulations and observations in 2018? Did the model perform well in other years? I suggest you evaluate simulated surface meteorological variables because they significantly affect surface air pollutants. I also recommend you assess the spatial distribution of surface O_3 and CO_2. For example, you can additionally evaluate model performance in key regions like NCP, YRD, and PRD apart from the whole domain.
Response: Thanks for your suggestion. We added the evaluations of meteorological

fields, O_3, and CO_2 from 2015 to 2018.

Changes in manuscript (L238~261):

"Given that the monitoring of near-surface O_3 levels by CNEMC was initiated only in late 2013, the monitoring sites in 2013 and 2014 were limited, and the monitoring period was disjointed. As a result, in this study, we compared the simulated meteorological fields, O_3, and CO_2 levels with observations only from 2015 to 2018.

Figures S1~4 demonstrated that the RegCM-Chem-YIBs model effectively captured the spatial distribution and magnitude of temperature, humidity, and wind over East Asia at 500 hPa, 850 hPa, and 1000 hPa between 2015 and 2018. However, due to the complex terrain's influence on the lower atmosphere, most models show better results at higher levels (Zhuang et al., 2018; Anwar et al., 2019; Xie et al., 2019). Thus, the simulations at 500 hPa were more consistent with the reanalysis data. At 1000 hPa, the simulated wind speed was slightly higher than the reanalysis data in eastern China. This difference may be due to common deficiencies in meteorological models, such as insufficient horizontal resolution, initial and boundary conditions, and physical parameterizations (Cassola and Burlando, 2012; Accadia et al., 2007), particularly in areas with low wind speeds (Carvalho et al., 2012).

Figures S5 and S6 demonstrated that the model accurately reproduced the observed increase in surface CO_2 and O_3 from 2015 to 2018, with high correlation coefficients ranging from 0.39 to 0.74 (Table 2). The model effectively captured the high concentrations of O_3 in major urban areas such as the NCP, the YRD, the PRD, the SCB, and the FWP, while also successfully reproducing the gradient in CO_2 concentrations between eastern and western China. However, the model slightly underpredicted MDA8 O_3 concentrations (-4.02 to -3.21 ppb) and overestimated CO_2 levels (3.32~7.07 ppm). These discrepancies are mainly attributed to uncertainties in the emissions inventory (Hong et al., 2017). Overall, the simulated meteorological factors and surface CO_2 and O_3 concentrations were deemed acceptable."

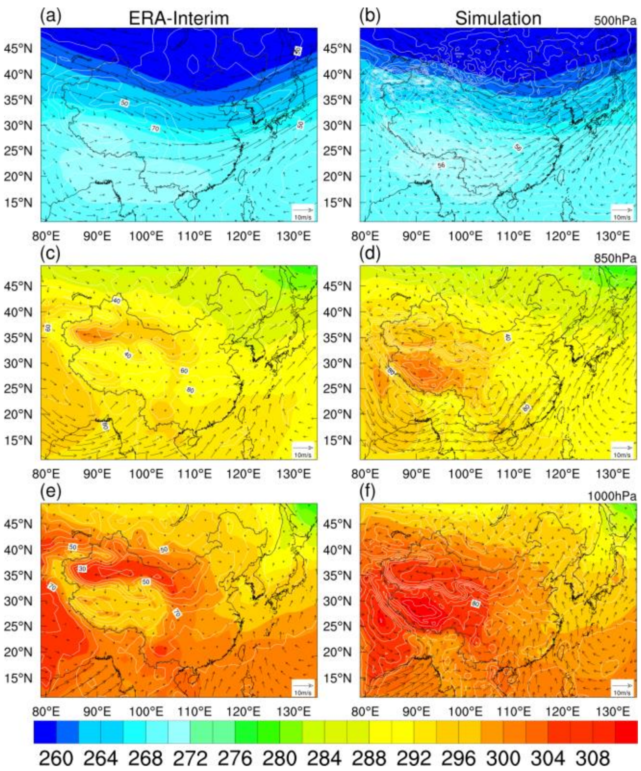

Figure S1. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) in 2015.

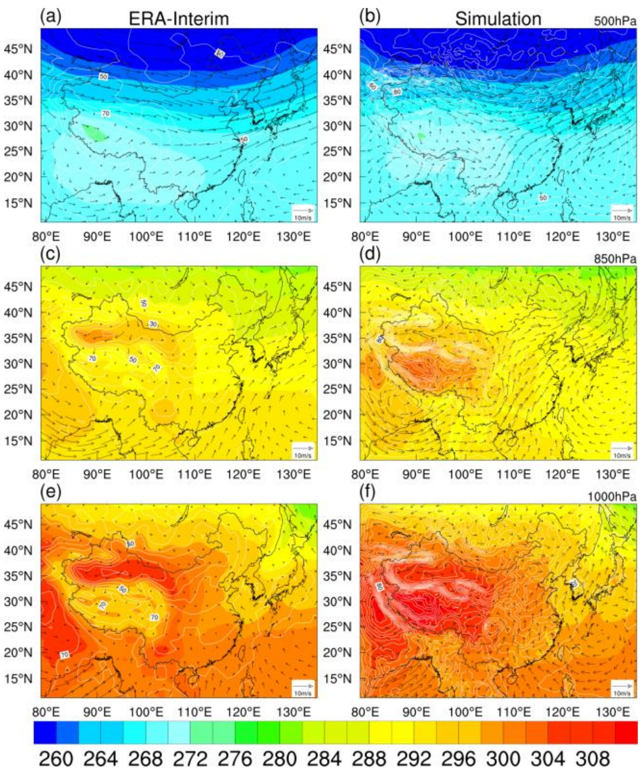

Figure S2. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) in 2016.

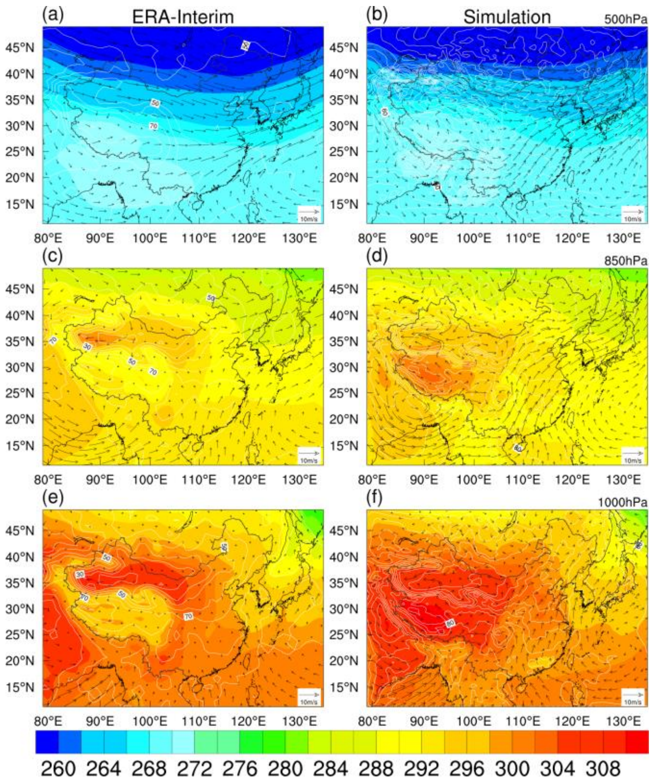

Figure S3. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) in 2017.

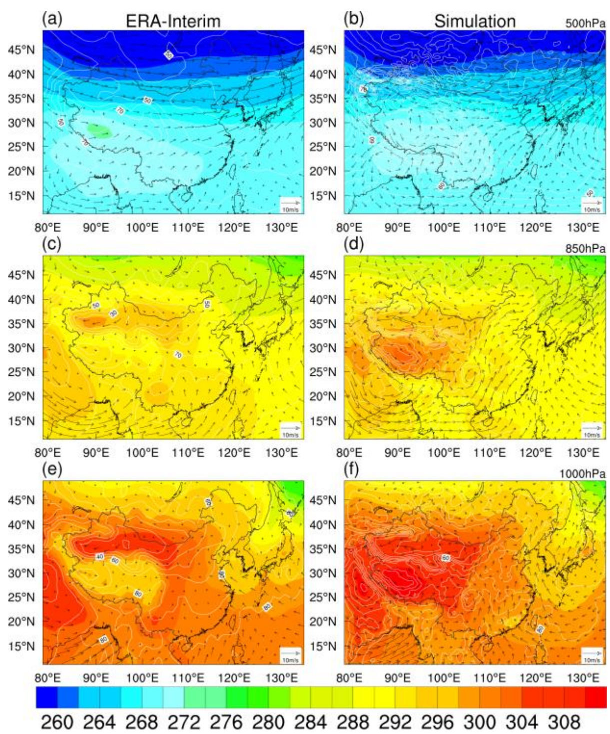

Figure S4. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) in 2018.

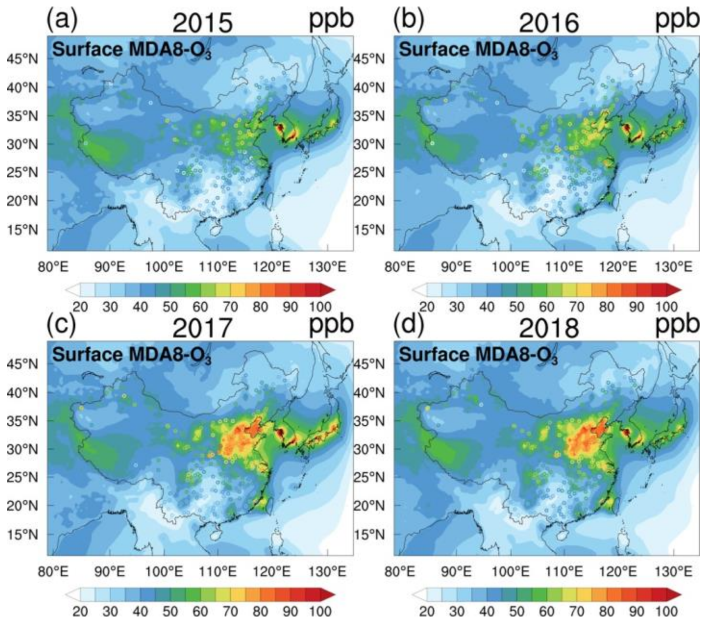

Figure S5. Comparisons between the simulated and observed surface MDA8 O_3 concentrations (units: ppb) during the summer monsoon period in (a)2015, (b)2016, (c)2017, (d)2018. Colored circles represent the observations.

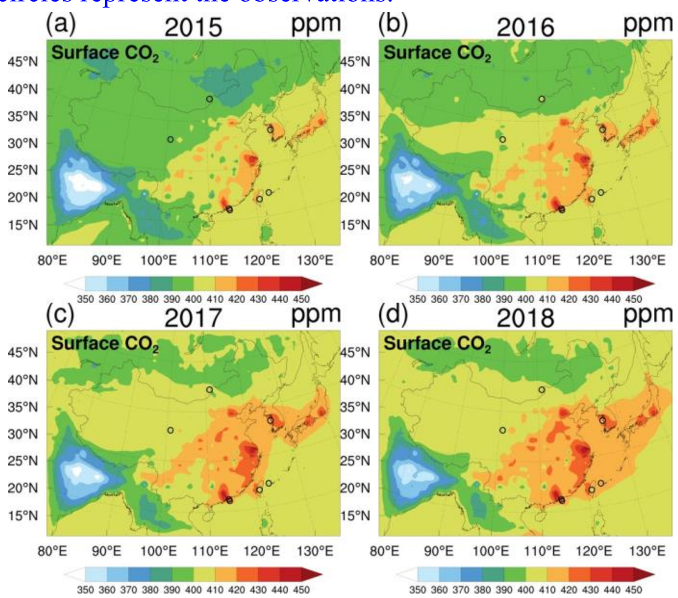

Figure S6. Comparisons between the simulated and observed surface CO_2 concentrations (units: ppm) during the summer monsoon period in (a)2015, (b)2016, (c)2017, (d)2018.

Table 2. Evaluations of the surface CO_2 (units: ppm) and MDA8 O_3 (units: ppb) during the summer monsoon period in East Asia.

Species	Year	OBS	SIM	MB	RMSE	R
CO_2 (ppm)	2015	402.82	406.98	4.16	9.37	0.44
	2016	407.12	410.44	3.32	8.22	0.69
	2017	408.35	413.62	5.27	11	0.39
	2018	409.61	416.68	7.07	11.32	0.41
MDA8 O_3 (ppb)	2015	48.77	44.75	-4.02	29.39	0.57
	2016	50.16	46.95	-3.21	27.56	0.60

	2017	55.43	51.87	-3.56	21.55	0.74
	2018	55.53	52.08	-3.42	24.78	0.73

OBS: observation; SIM: simulation; MB: bias; NMB: normalized mean bias; RMSE: root mean square error; R: correlation coefficient. MDA8 O_3: the maximum daily 8-hour average O_3.

References

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6. Line 243: This expression is ambiguous. Meteorological factors are favorable for ozone formation in summer.

Response: Thanks. We have revised the ambiguous expression.

Changes in manuscript (L300~301):

"Overall, the meteorological variations from 2008 to 2018 were unfavorable for the O_3 increase during the EASM period, as illustrated in Figure 3."

7. Line 260-270: There are some contradictions in this part. You attributed the decrease in ozone concentration to increased cloud fraction, decreased SWF, increased precipitation, and enhanced wind speed. But how the warmer surface and higher PBL can accompany these conditions?

Response: Thanks. We have added some discussions on this aspect.

Changes in manuscript (L319~332):

"As we know, the formation of surface O_3 is promoted by rising temperatures (Steiner et al., 2010). However, increased surface temperatures can also intensify turbulence within the planetary boundary layer (PBL), increasing PBL height (Guo et al., 2016). This increase in PBL height, coupled with the enhanced upward motion, can transport near-surface pollutants to the upper atmosphere, reducing their concentration in the lower atmosphere (Gao et al., 2016). Additionally, the upward motion can also facilitate cloud formation and precipitation, resulting in a reduction of near-surface atmospheric pollutants via precipitation washout (Yoo et al., 2014).

We have improved the accuracy of O_3 photodissociation rate calculations by including varying AOD and SSA in the TUV module, as described in Section 2.3.2. As a result, the increase in cloud cover reduced the shortwave radiation flux and photochemical formation rates of near-surface O_3, leading to decreased formation. Thus, the increase in near-surface temperature is often accompanied by an elevation in PBL height, enhanced cloud cover, precipitation, and reduced shortwave radiation. Moreover, higher wind speeds can enhance the dispersion of O_3 (Gorai et al., 2015)."

References

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(O-3, CO, NO2, SO2, and PM10) by summertime rain, Atmospheric Environment, 82, 226-237,https://doi.org/10.1016/j.atmosenv.2013.10.022, 2014.

8. Figure 5: Please modify the value range of the color bar.

Response: Thanks for pointing that out. We have modified the value range of the color bar in Fig.5.

Changes in manuscript:

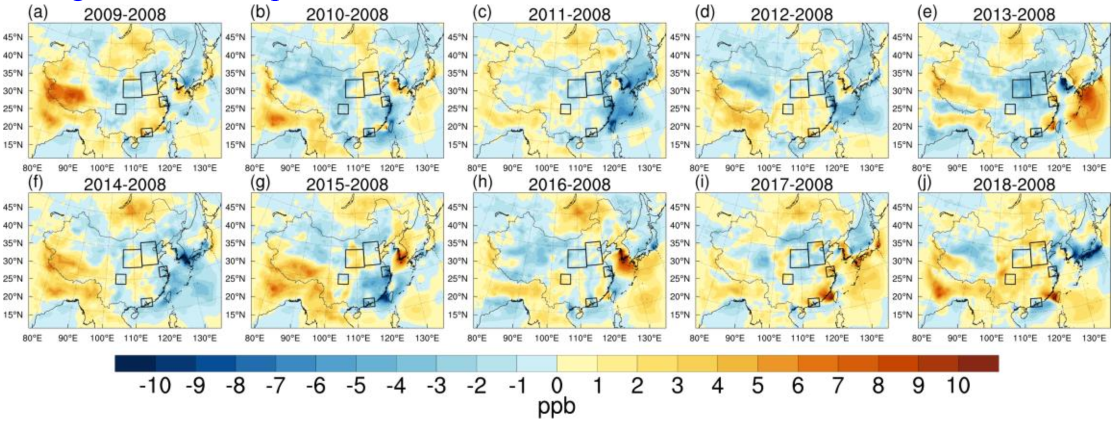


Figure 3. The responds of the surface MDA8 O_3 mixing ratios (units: ppb) to variations in meteorological conditions during the summer monsoon period in 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

9. Section 3.4: How did you quantify the contributions of isoprene and precipitation to ozone concentration?

Response: Thanks. We did not quantitatively differentiate the impacts of precipitation and isoprene on O_3 concentrations. In Section 3.4, we analyzed the impact of CO_2 on O_3 and provided explanations from two perspectives: isoprene emissions and precipitation changes. This approach facilitated a more comprehensive comprehension of the mechanisms that underlie the impact of CO_2 on O_3 concentrations. We have improved the statements in this section.

Changes in manuscript (L373~385):

"CO_2 is a significant driver of climate change and alterations in biogenic emissions. As shown in Figures 6 b and c, the impact of CO_2 on O_3 levels varies across locations, with a positive effect of 0.5~2 ppb along the southeastern coast of China but a negative influence of -0.5 to -2 ppb in the southwest and central China. CO_2 affects O_3 concentration by influencing both precipitation and isoprene emissions. In western and central China, CO_2 primarily affects O_3 concentration through its impact on precipitation (Table 5). Elevated CO_2 concentrations lead to increased precipitation (0.06~0.64 mm/day) in the FWP and SCB regions, resulting in a decrease in surface O_3 (up to -0.51 ppb). In eastern and southern coastal China, where vegetation is abundant, CO_2 has a greater impact on isoprene emissions. In the YRD region, decreased isoprene (-0.58 to -0.32 μg/m^3) and increased precipitations (0.09~0.13 mm/day) reduced MDA8 O_3 levels (0.09~0.14 ppb). In PRD, increased isoprene levels (0.31~0.92 μg/m^3) and decreased precipitations (-1.02~-0.33 mm/day) led to the enhancement of MDA8 O_3 (0.28~0.46 ppb)."

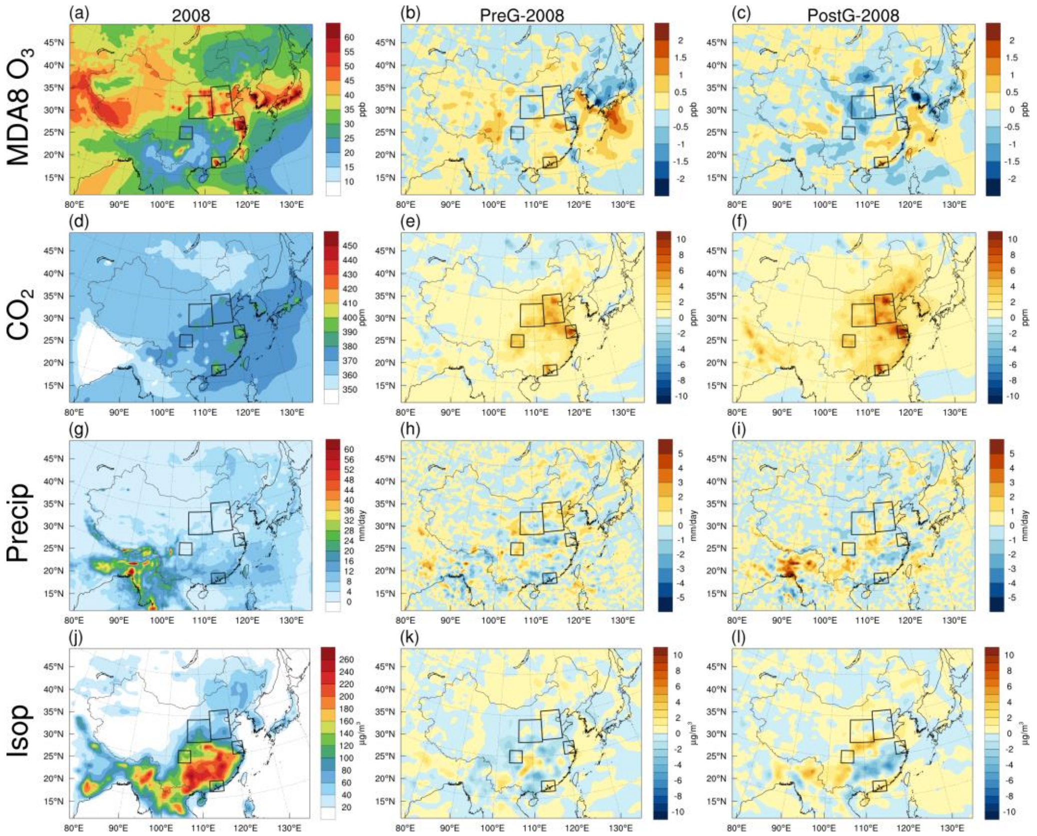


Figure 6. The simulated averaged MDA8 O_3 (a~c, units: ppb), CO_2 (d~f, units: ppm), precipitation (g~i, units: mm/day), and isoprene mixing ratios (j~l, units: μg/m^3) in 2008 from the base simulations (the left column) and their changes due to variations in CO_2 emissions in PreG (2009~2013, the central column) and PostG (2014~2018, the right column) relative to 2008.

Table 5. Simulated responses of MDA8 O_3 mixing ratios (units: ppb), CO_2 mixing ratios (units: ppm), precipitations (units: mm/day), 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 (2009~2013) and PostG (2014~2018) relative to 2008.

Regions	Period	MDA8 O_3 (ppb)	CO_2 (ppm)	Precipitation (mm/day)	Isoprene (μg/m^3)
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

10. Figure 7: Please modify the value range of the color bar.

Response: Thanks. We have modified the color bar in Fig.7.

Changes in manuscript:

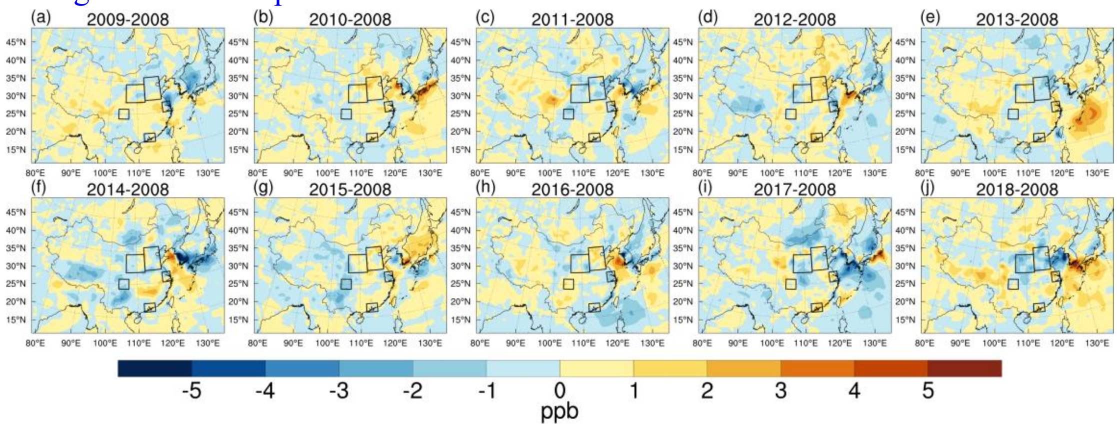


Figure 5. Simulated responses of surface MDA8 O_3 mixing ratios (units: ppb) to the variations in CO_2 emissions during the summer monsoon period in 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 I, 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

11. Line 342-343: What did you mean by "due to the slight increase in air O_3 concentration"?

Response: Thanks. Sorry for the mistake. We have revised the erroneous expression.

Changes in manuscript (L432~433):

"In the PRD region, anthropogenic emissions led to a slight enhancement of O_3 by 2.33~5.74 ppb."

12. Section 3.5: I suggest adding a figure or table showing emission trends of main air pollutants and precursors to support the explanation.

Response: Thanks for the suggestion. We have added Fig. S1 and Table S4 to illustrate the emission trend of SO_2, NOx, VOCs, NH_3, CO, PM_{10}, $PM_{2.5}$, and OC from the MEIC inventory.

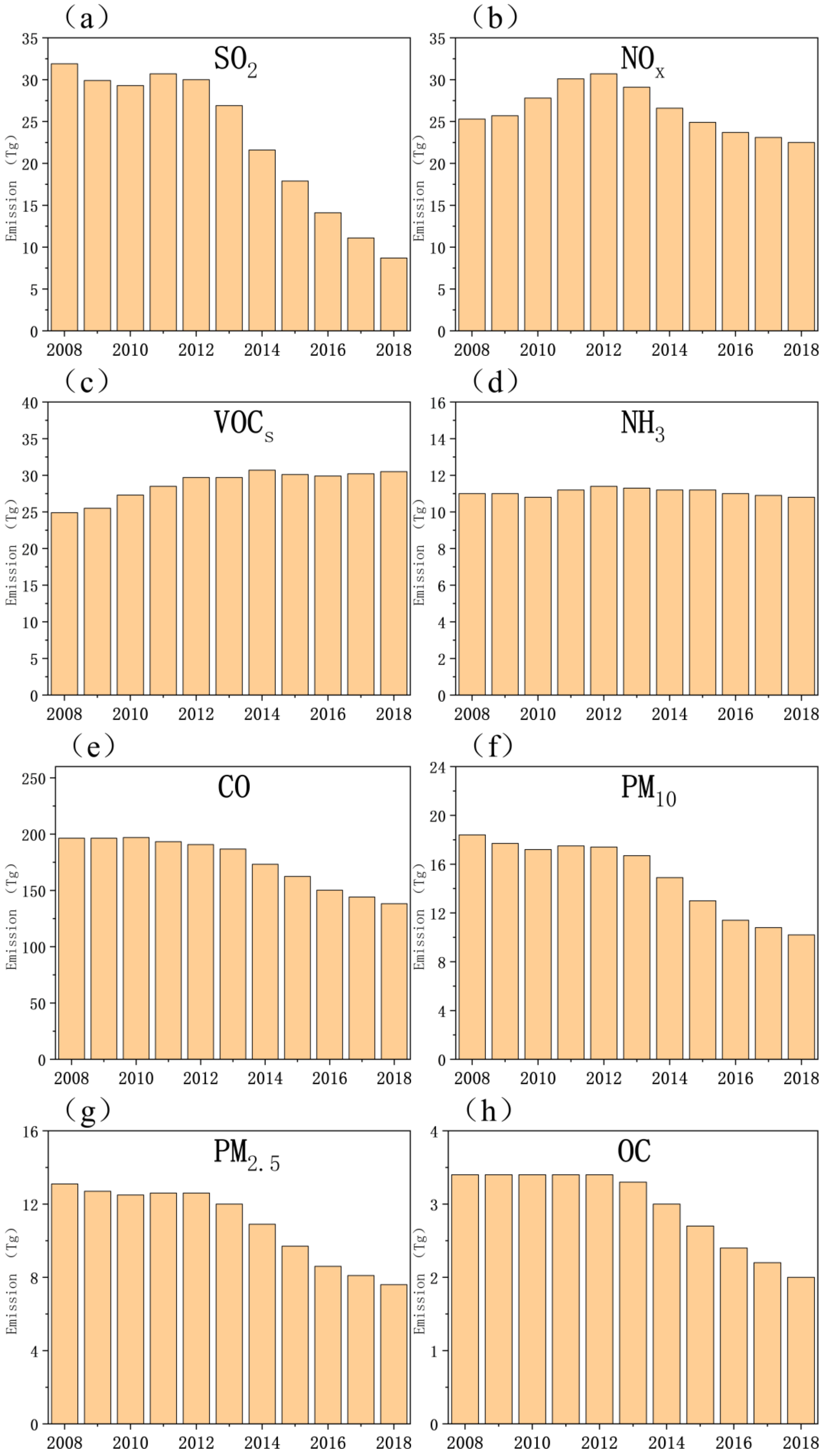
Changes in manuscript:

(L413~422):" Figure S8 and Table S1 illustrate that the levels of $PM_{2.5}$, PM_{10}, SO_2, CO, and OC emissions remained consistently high during the PreG period. However, a linear decrease in emissions was observed after the implementation of the Clean Air Action Plan in 2013. Prior to 2013, the emission of VOCs increased steadily but subsequently stabilized. Similarly, the emission of nitrogen oxides (NOx) exhibited an upward trend before 2013, but since then, the emissions have shown a linear decrease, with each subsequent year exhibiting lower levels of NOx emissions. In comparison to other species, the emissions of ammonia (NH_3) remained relatively stable from 2008 to 2018. Our analysis results of the emissions of different species align with those of Zheng et al. (2018), who computed the changes of each species in the MEIC inventory from 2010 to 2017."

(L442~448):"Before 2013, the continuous increase in VOCs and NOx emissions (Figure S8 b, c) facilitated the rise of O_3 levels. Following the implementation of the Clean Air Action Plan in 2013, the emissions of VOCs and NOx were regulated. However, with the decrease in $PM_{2.5}$ levels, direct radiation increased, and scattered radiation decreased (Figure 9), thereby promoting the photochemical formation of O_3 (Bian et al., 2007). In addition, the reduced NO emission weakened the titration effect (Figure S8 b), thus increasing surface O_3 (Li et al., 2022)."

Table S1. Changes in the model domain's anthropogenic emissions (Tg) from 2008 to 2018

Year	SO_2	NO_X	VOCs	NH_3	CO	PM_{10}	$PM_{2.5}$	OC
2008	31.9	25.3	24.9	11.0	196.4	18.4	13.1	3.4
2009	29.9	25.7	25.5	11.0	196.4	17.7	12.7	3.4
2010	29.3	27.8	27.3	10.8	197.0	17.2	12.5	3.4
2011	30.7	30.1	28.5	11.2	193.3	17.5	12.6	3.4
2012	30.0	30.7	29.7	11.4	190.7	17.4	12.6	3.4
2013	26.9	29.1	29.7	11.3	186.8	16.7	12.0	3.3
2014	21.6	26.6	30.7	11.2	173.2	14.9	10.9	3.0
2015	17.9	24.9	30.1	11.2	162.4	13.0	9.7	2.7
2016	14.1	23.7	29.9	11.0	150.2	11.4	8.6	2.4
2017	11.1	23.1	30.2	10.9	144.1	10.8	8.1	2.2
2018	8.7	22.5	30.5	10.8	138.2	10.2	7.6	2.0



(a)
SO2
(b)
NOx
(c)
VOCs
(d)
NH3
(e)
CO
(f)
PM10
(g)
PM2.5
(h)
OC
Emission (Tg)
2008
2010
2012
2014
2016
2018

Figure S8. Changes in the anthropogenic emissions (Tg) from 2008 to 2018. The species include (a)SO_2, (b)NOx, (c)VOCs, (d)NH_3, (e)CO, (f)PM_{10}, (g)PM2.5, (h)OC.

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