

Response to Reviewers 1

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 O₃ variation across China and highlighted the importance of considering CO₂ 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 CO₂, 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 CO₂ played a critical role in the variability of O₃ through radiative forcing and isoprene emissions, particularly in southern China, inducing an increase in O₃ 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 O₃ levels, with limited attention given to the role of CO₂ variations. However, due to the rapid socioeconomic growth in China and the subsequent surge in energy consumption, CO₂ 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 CO₂ on O₃, it is crucial to evaluate the influence of changes in CO₂ concentration on the maximum daily 8-hour average (MDA8) O₃ concentrations at the surface. Thus, a comprehensive assessment of the impact of anthropogenic emissions, meteorological factors, and CO₂ on surface O₃ is imperative.”

Section 3.4 (L386~393): “In some years, the impact of changed CO₂ can be as significant as or even surpass that of anthropogenic emissions and meteorology (Figure 10). For example, in 2013, CO₂ caused an increase of 0.95 ppb in MDA8 O₃

in the YRD region, which exceeded that of anthropogenic emissions (0.87 ppb). Similarly, in the PRD region in 2012, the effect of CO₂, anthropogenic emissions, and meteorology was 1.41, 1.77, and 1.95 ppb, respectively. Even in the NCP in 2010, the impact of CO₂ (0.75 ppb) was comparable to that of anthropogenic emissions (1.5 ppb). In summary, CO₂ has a significant impact on surface O₃ concentrations by influencing radiation and isoprene emissions, with more prominent effects in regions with abundant vegetation.”

References

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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.

References

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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 CO₂ and O₃ concentrations were assumed to be constant throughout the year. However, atmospheric CO₂ and O₃ 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 CO₂ and O₃ 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 CO₂ and O₃ concentrations into the radiation module. Therefore, we included the varying CO₂ and O₃ 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, SO₂ column, NO₂ 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 O₃ (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.”

References

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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 O₃ and CO₂. 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, O₃, and CO₂ from 2015 to 2018.

Changes in manuscript (L238~261):

“Given that the monitoring of near-surface O₃ 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, O₃, and CO₂ 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 CO₂ and O₃ 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 O₃ in major urban areas such as the NCP, the YRD, the PRD, the SCB, and the FWP, while also successfully reproducing the gradient in CO₂ concentrations between eastern and western China. However, the model slightly underpredicted MDA8 O₃ concentrations (-4.02 to -3.21 ppb) and overestimated CO₂ 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 CO₂ and O₃ concentrations were deemed acceptable.”

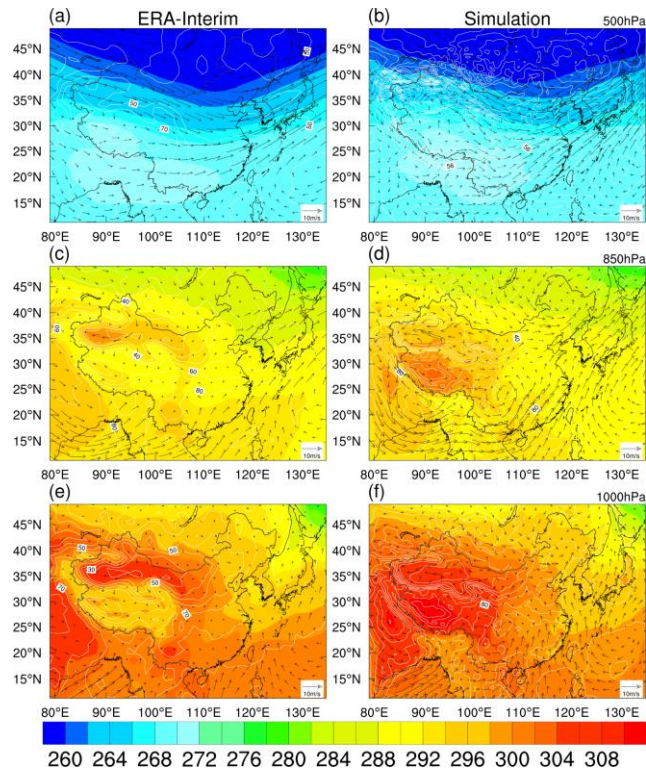


Figure S1. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) in 2015.

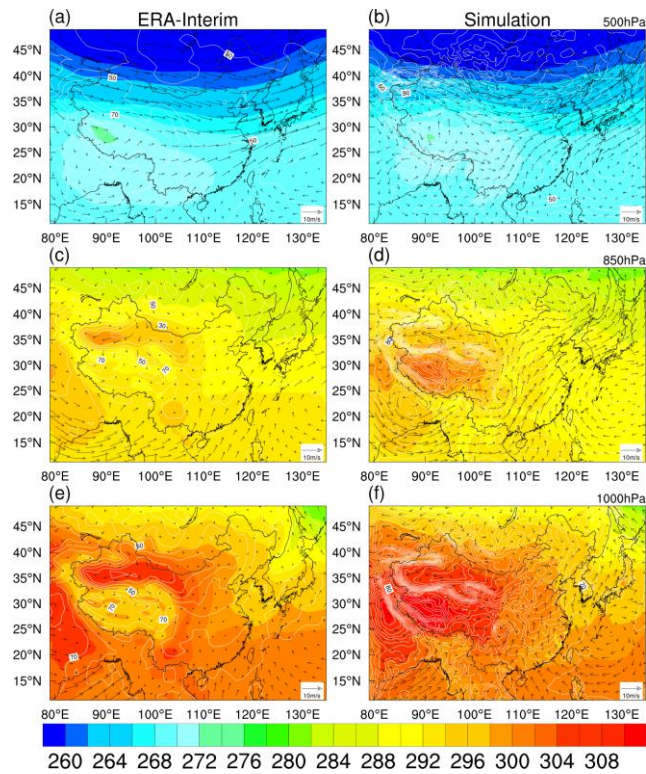


Figure S2. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) in 2016.

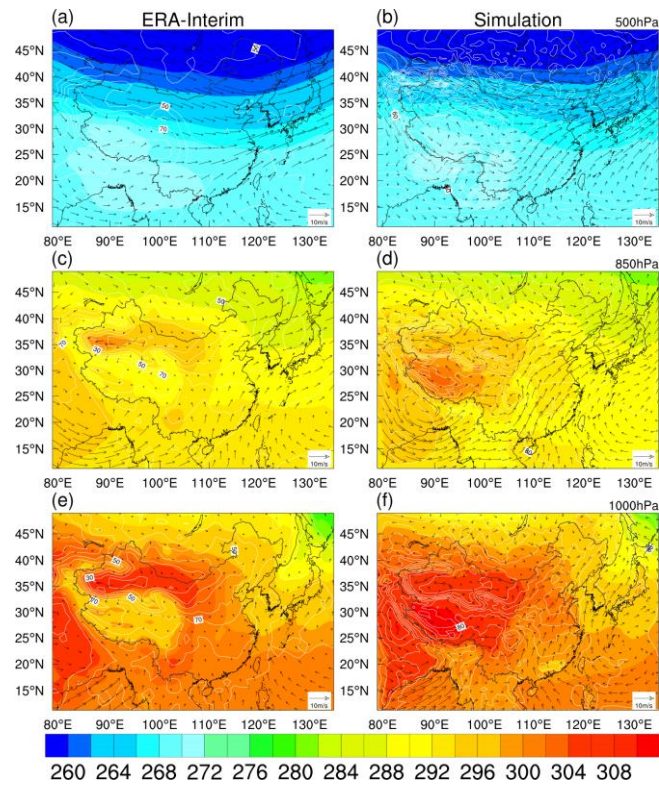


Figure S3. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) in 2017.

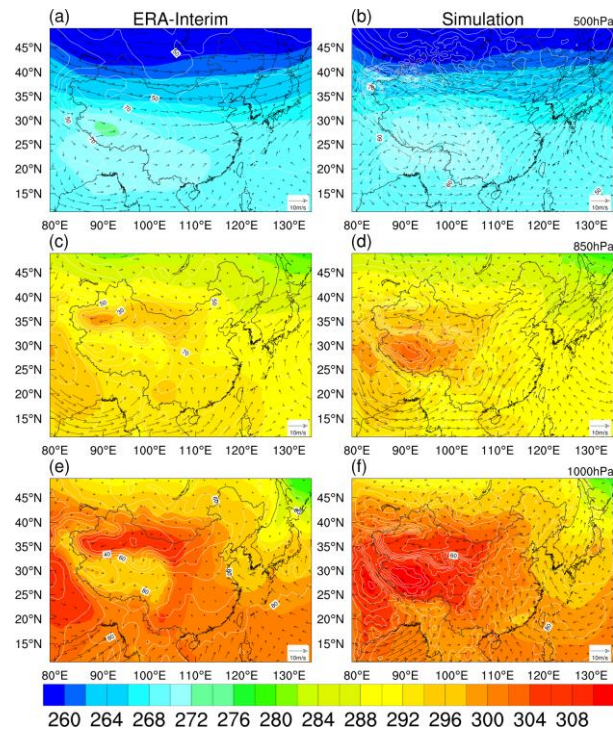


Figure S4. Comparisons between the simulated (a, c) and reanalysis (b, d) mean temperature (shading, units: K), wind (vectors, units: m/s), and relative humidity (contours, units: %) at 500 hPa (a, b), 850 hPa (c, d) and 1000 hPa (e, f) in 2018.

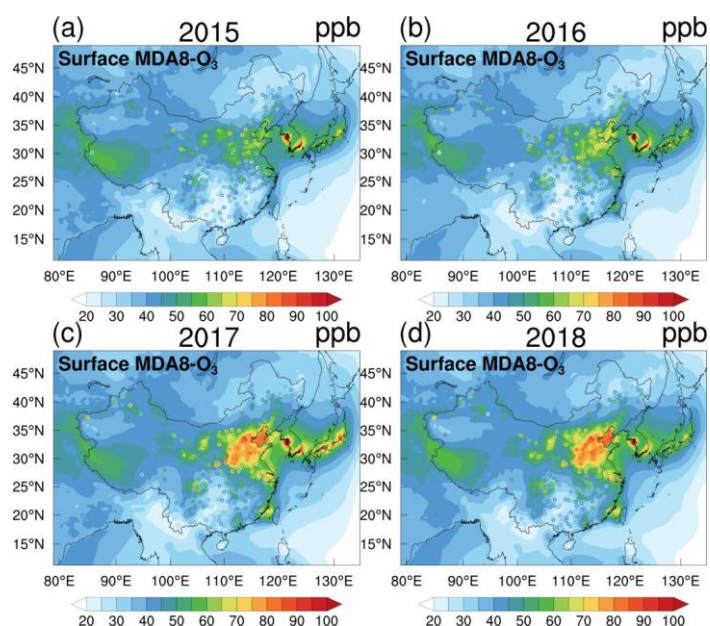


Figure S5. Comparisons between the simulated and observed surface MDA8 O₃ concentrations (units: ppb) during the summer monsoon period in (a)2015, (b)2016, (c)2017, (d)2018. Colored circles represent the observations.

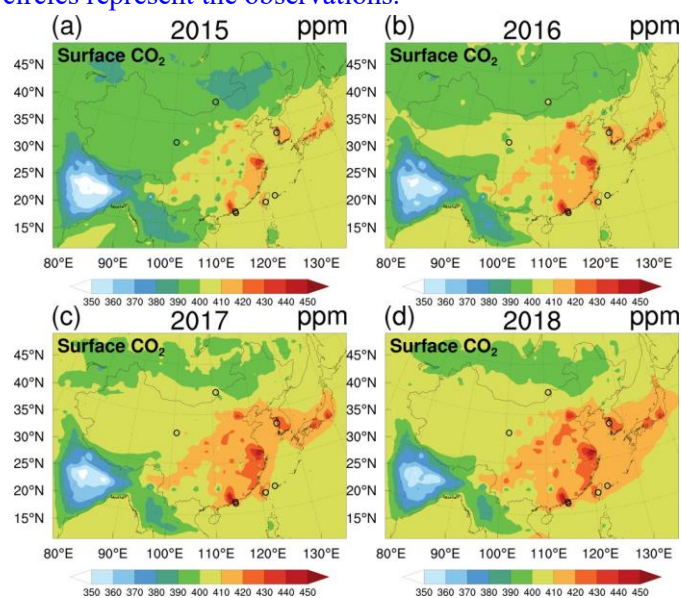


Figure S6. Comparisons between the simulated and observed surface CO₂ concentrations (units: ppm) during the summer monsoon period in (a)2015, (b)2016, (c)2017, (d)2018.

Table 2. Evaluations of the surface CO₂ (units: ppm) and MDA8 O₃ (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 O₃: the maximum daily 8-hour average O₃.

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 O₃ 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 O₃ 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 O₃ 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 O₃, 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 O₃ (Gorai et al., 2015).”

References

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(O₃, 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:

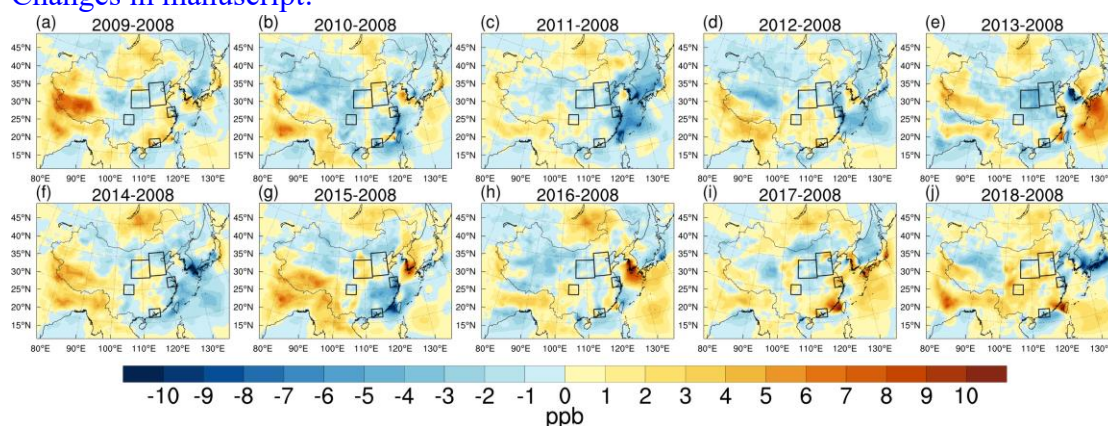


Figure 3. The responds of the surface MDA8 O₃ mixing ratios (units: ppb) to variations in meteorological conditions during the summer monsoon period in 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

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 O₃ concentrations. In Section 3.4, we analyzed the impact of CO₂ on O₃ 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 CO₂ on O₃ concentrations. We have improved the statements in this section.

Changes in manuscript (L373~385):

“CO₂ is a significant driver of climate change and alterations in biogenic emissions. As shown in Figures 6 b and c, the impact of CO₂ on O₃ 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. CO₂ affects O₃ concentration by influencing both precipitation and isoprene emissions. In western and central China, CO₂ primarily affects O₃ concentration through its impact on precipitation (Table 5). Elevated CO₂ concentrations lead to increased precipitation (0.06~0.64 mm/day) in the FWP and SCB regions, resulting in a decrease in surface O₃ (up to -0.51 ppb). In eastern and southern coastal China, where vegetation is abundant, CO₂ has a greater impact on isoprene emissions. In the YRD region, decreased isoprene (-0.58 to -0.32 μg/m³) and increased precipitations (0.09~0.13 mm/day) reduced MDA8 O₃ levels (0.09~0.14 ppb). In PRD, increased

isoprene levels ($0.31\sim 0.92\ \mu\text{g}/\text{m}^3$) and decreased precipitations ($-1.02\sim -0.33\ \text{mm}/\text{day}$) led to the enhancement of MDA8 O_3 ($0.28\sim 0.46\ \text{ppb}$).”

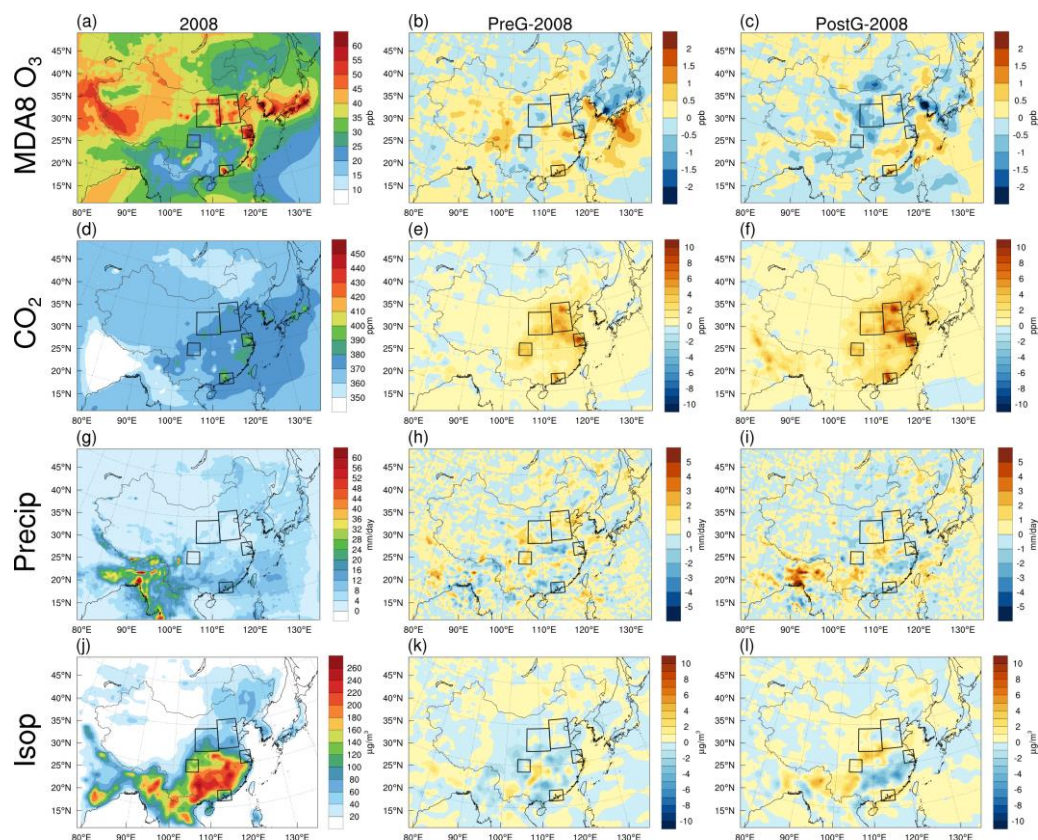


Figure 6. The simulated averaged MDA8 O_3 (a~c, units: ppb), CO_2 (d~f, units: ppm), precipitation (g~i, units: mm/day), and isoprene mixing ratios (j~l, units: $\mu\text{g}/\text{m}^3$) in 2008 from the base simulations (the left column) and their changes due to variations in CO_2 emissions in PreG (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 ($\mu\text{g}/\text{m}^3$)
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
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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:

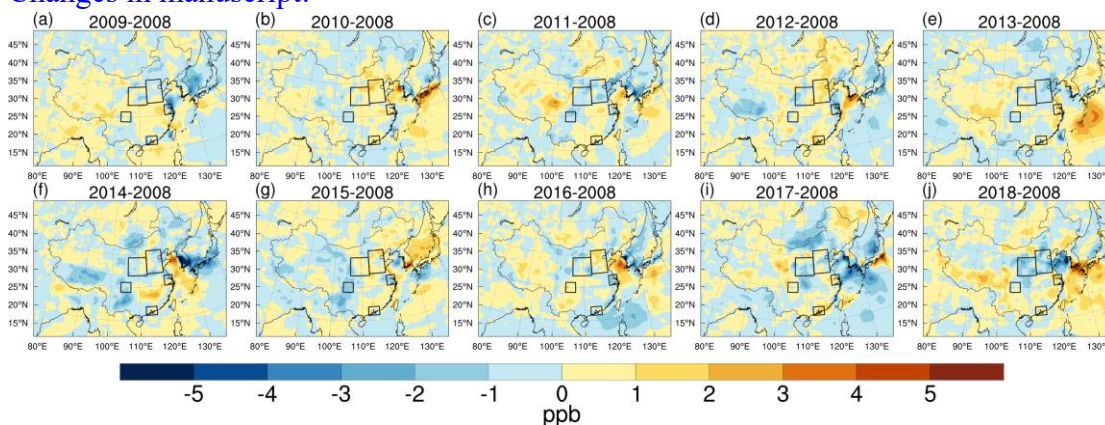


Figure 5. Simulated responses of surface MDA8 O₃ mixing ratios (units: ppb) to the variations in CO₂ emissions during the summer monsoon period in 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

11. Line 342-343: What did you mean by “due to the slight increase in air O₃ 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 O₃ 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 SO₂, NO_x, VOCs, NH₃, CO, PM₁₀, 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}, PM₁₀, SO₂, 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 (NO_x) exhibited an upward trend before 2013, but since then, the emissions have shown a linear decrease, with each subsequent year exhibiting lower levels of NO_x emissions. In comparison to other species, the emissions of ammonia (NH₃) 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 NO_x emissions (Figure S8 b, c) facilitated the rise of O₃ levels. Following the implementation of the Clean Air Action Plan in 2013, the emissions of VOCs and NO_x 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 O₃ (Bian et al., 2007). In addition, the reduced NO emission weakened the titration effect (Figure S8 b), thus increasing surface O₃ (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

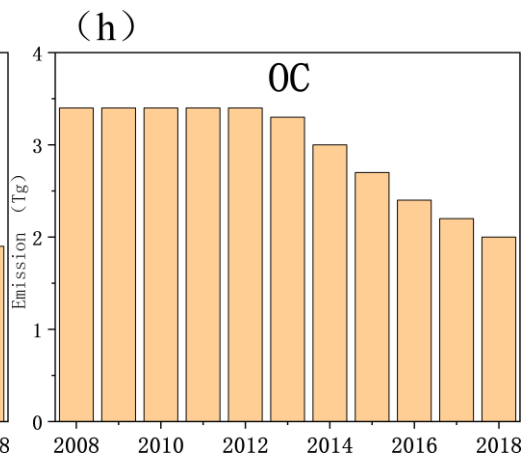
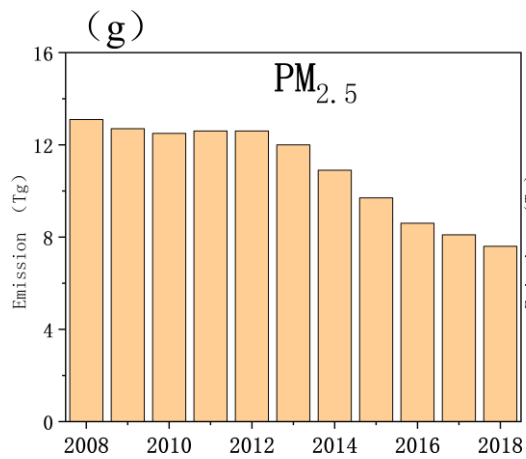
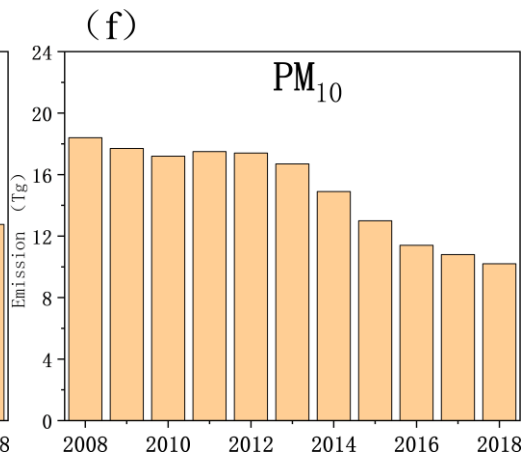
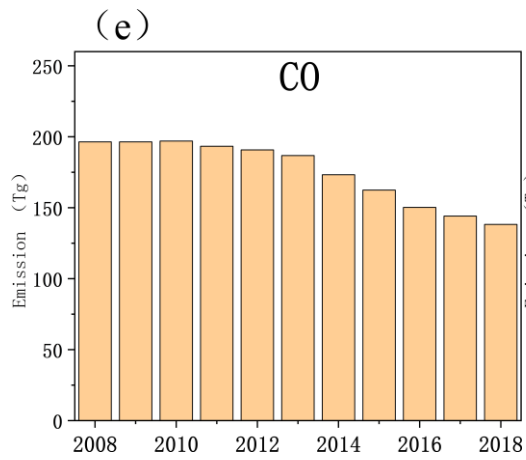
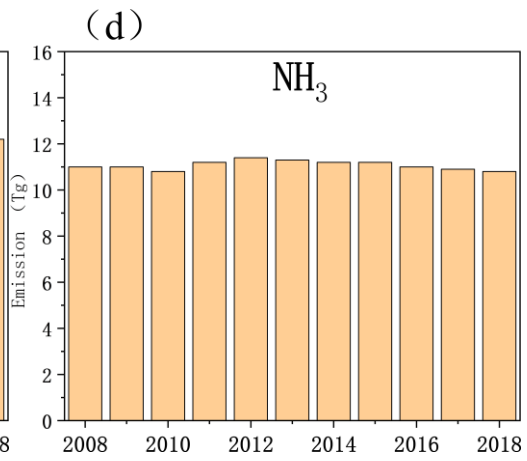
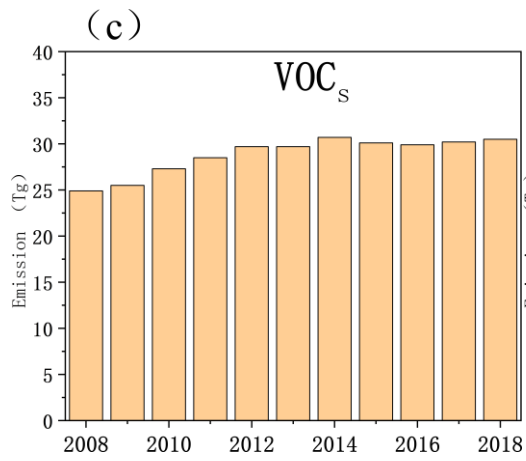
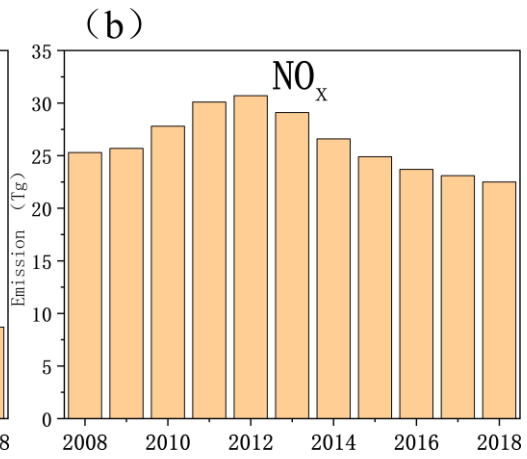
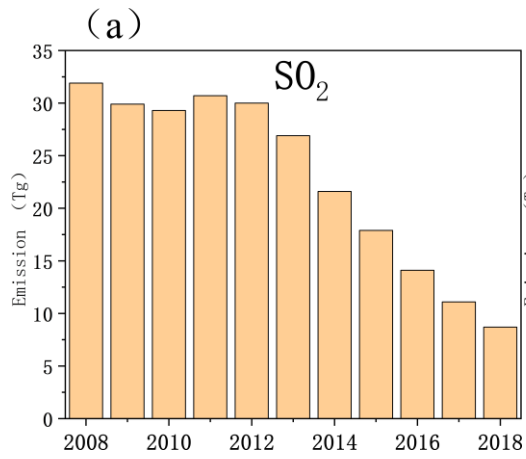


Figure S8. Changes in the anthropogenic emissions (Tg) from 2008 to 2018. The species include (a)SO₂, (b)NO_x, (c)VOCs, (d)NH₃, (e)CO, (f)PM₁₀, (g)PM_{2.5}, (h)OC.

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Response to Reviewers 2

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 #2:

Major comments:

This is an interesting study looking at the effects of emission changes, meteorological variability, and -for the first time- also impacts of CO₂ (via radiative forcing, but also through isoprene emission).

The manuscript needs to be improved on several aspects.

Response: We thank Referee #2 for his/her valuable comments, which have greatly improved our manuscript. We have attempted to make a revision addressing each of the points mentioned in his/her review. Please note that the line numbers given below refer to the clean version of the manuscript.

1. model descriptions, boundary conditions and a host of other assumptions made need to be improved and completed, to be able to understand what has been done in the study. Units need to be included in captions of tables and figures.

Response: Thanks. We have refined the statements of the model descriptions, model improvements, and emissions and Experiment settings in the revised manuscript. We added units in captions of tables and figures.

Changes in manuscript:

Model description (L121~134) : “The RegCM-Chem-YIBs is a regional climate-chemistry-ecology model developed from the RegCM model. RegCM is a regional climate model initially developed by the International Center for Theoretical Physics (ICTP) (Giorgi et al., 2012). Shalaby et al. (2012) integrated the Chem chemistry model into the RegCM model and incorporated the condensed version of the Carbon Bond Mechanism (CBM-Z) to enhance the model's capabilities. To further improve the model's performance, Yin et al. (2015) added a Volatility Basis Set (VBS) scheme to simulate Secondary Organic Aerosols (SOA). Xie et al. (2020) further modified the model by incorporating CO₂ as a tracer, which is subject to regulation by sources, sinks, and atmospheric transport processes. The model represents the four sources and sinks of CO₂ as surface fluxes, including emissions from fossil fuels and biomass burning, air-sea CO₂ exchange, and terrestrial biosphere CO₂ fluxes. Additionally, the model incorporated the Yale Interactive Terrestrial Biosphere (YIBs), a land carbon cycle model that enables the simulation of ecological processes, including carbon assimilation, allocation, and autotrophic and heterotrophic

respiration (Yue and Unger, 2015).”

(L146~153) : “The RegCM model offers a variety of physical and chemical parameterization options. Here, the climatological chemical boundary conditions were driven by the Model of Ozone and Related Chemical Tracers (MOZART). The gas-phase chemistry employed the CBM-Z scheme (Zaveri and Peters, 1999). For the boundary layer scheme, the Holtslag PBL approach was utilized (Khayatanyazdi et al., 2021). The Grell cumulus convection scheme was employed to simulate convective processes (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).”

Model improvements:

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 CO₂ and O₃ concentrations were assumed to be constant throughout the year. However, atmospheric CO₂ and O₃ 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 CO₂ and O₃ 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 CO₂ and O₃ concentrations into the radiation module. Therefore, we included the varying CO₂ and O₃ 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, SO₂ column, NO₂ 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 O₃ (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.”

Emissions and Experiment settings (L185~186): “CO₂ emissions and boundary

conditions were derived from the NOAA CarbonTracker CT2019 dataset.”

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2. The use of a single year (2008) as a reference year for pre-governance 2009-2013 and post-governance (2014-2018) periods is problematic. Often what we are looking at is some specific feature of 2008 that is showing up as difference.

A more classic attribution approach using e.g. constant 2008 emissions with variable meteo; or constant year with varying emissions would be more appropriate. Alternatively, I could suggest using the period 2008-2013 as reference period (pre-governance) and just show the changes compared to this period. Make sure that the units are correct and comparable.

Response: Thanks. We did indeed do it this way. In the revised version, we have further clarified and enhanced this description and updated the expression in Table 1. We made an error in our unit conversion and should use ppb instead of ppb per year. We have corrected the units used in the manuscript.

Changes in manuscript:

(L195~205): “In the Base experiment, we incorporated interannual variations in anthropogenic emissions, meteorological fields, and CO₂ emissions. Meteorological conditions (CO₂ emissions) were kept constant at 2008 levels over ten years, referred to as the SIM_{MET=2008} (SIM_{CO₂=2008}) experiment.

The changes in O₃ concentrations relative to 2008 between 2009 and 2018 were determined by comparing simulations of different years with 2008 in the Base experiment (Eq. (1)). The impact of changed meteorological conditions on O₃ concentrations relative to 2008 was assessed by comparing results between SIM_{MET=2008} and the Base experiment in the same year (Eq. (2)). The contribution of changed CO₂ emissions was similarly estimated (Eq. (3)). Finally, the influence of anthropogenic emissions was calculated by excluding the impact of meteorological factors and CO₂ from the changes in O₃ concentrations (Eq. (4)). Table 1 shows the results of the numerical experiments.”

$$\Delta O_i = Base_i - Base_{2008} \quad (1)$$

$$\Delta M_i = SIM_i - SIM_{i,MET=2008} \quad (2)$$

$$\Delta C_i = SIM_i - SIM_{i,CO_2=2008} \quad (3)$$

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

ΔO_i : The changes in O₃ concentrations in the year i relative to 2008.

$Base_i$: The O₃ concentrations in the Base experiment in the year i.

ΔM_i : The changes in O₃ concentrations in the year i due to meteorological factors variations.

$SIM_{i,MET=2008}$: The O₃ concentrations in the SIM_{MET=2008} experiment in the year i.

ΔC_i : The changes in O₃ concentrations in the year i due to CO₂ variations.

$SIM_{i,CO_2=2008}$: The O₃ concentrations in the $SIM_{CO_2=2008}$ experiment in the year i.

ΔE_i : The changes in O₃ concentrations in the year i due to anthropogenic emissions variations.

Table 1. The Numerical experimental in this study.

Experiment	Time	Meteorological fields	CO ₂ emissions	Anthropogenic emissions
Base	2008-2018	Varying	Varying	Varying
$SIM_{MET=2008}$	2009-2018	2008	Varying	Varying
$SIM_{CO_2=2008}$		Varying	2008	Varying

3. I suggest re-ordering of sections to describe effect of emissions; effects of meteorology; and CO₂ effect in the order of importance. A 4 section could be added describing the addiviity.

Response: Thanks. As described in the previous question (2), we first obtained the contributions of meteorological and CO₂ changes to O₃ concentrations. Then, the influence of anthropogenic emissions was calculated by excluding the impact of meteorological factors and CO₂ from the changes in O₃ concentrations. Therefore, we conducted a two-step analysis that first evaluated the effects of meteorology and CO₂ on O₃ changes, followed by an analysis of the effects of anthropogenic emissions on O₃ concentrations. In addition, we have added a new section 3.6 to conduct an attribution analysis of changes in O₃ concentrations.

Changes in manuscript:

3.6 Attribution analysis of ozone changes in 2008~2018

(L472~488): “Finally, we presented an attribution diagram depicting the changes in O₃ concentration from 2008 to 2018. The total variation in O₃ concentration can be attributed to the combined effects of meteorological changes, changes in CO₂ concentration, and anthropogenic emissions (Figure 10).

The primary driver of the O₃ concentration variation from 2008 to 2018 was the changes in anthropogenic emissions, particularly in regions with high emissions, such as the NCP and FWP. Although the Clean Air Action Plan was implemented in 2013, it did not reduce the contribution of anthropogenic emissions to the O₃ increase. Even in the PostG period, with the development of urbanization and industrialization, the impact of changed anthropogenic emissions on O₃ has gradually become more prominent than changed meteorology and CO₂. The contribution of changed meteorology to O₃ was generally negative in the five regions, with a more significant impact in the YRD and PRD regions. This may be attributed to their proximity to the ocean and susceptibility to the summer monsoon influence. Changes in CO₂

concentration affected O₃ concentration by altering radiation and isoprene emissions, with a more significant impact in the YRD and PRD regions where vegetation was abundant. In some years, it even surpassed the effects of anthropogenic emissions. Therefore, we suggest that the influence of CO₂ concentration changes on O₃ concentration should be considered in regions with high vegetation coverage.”

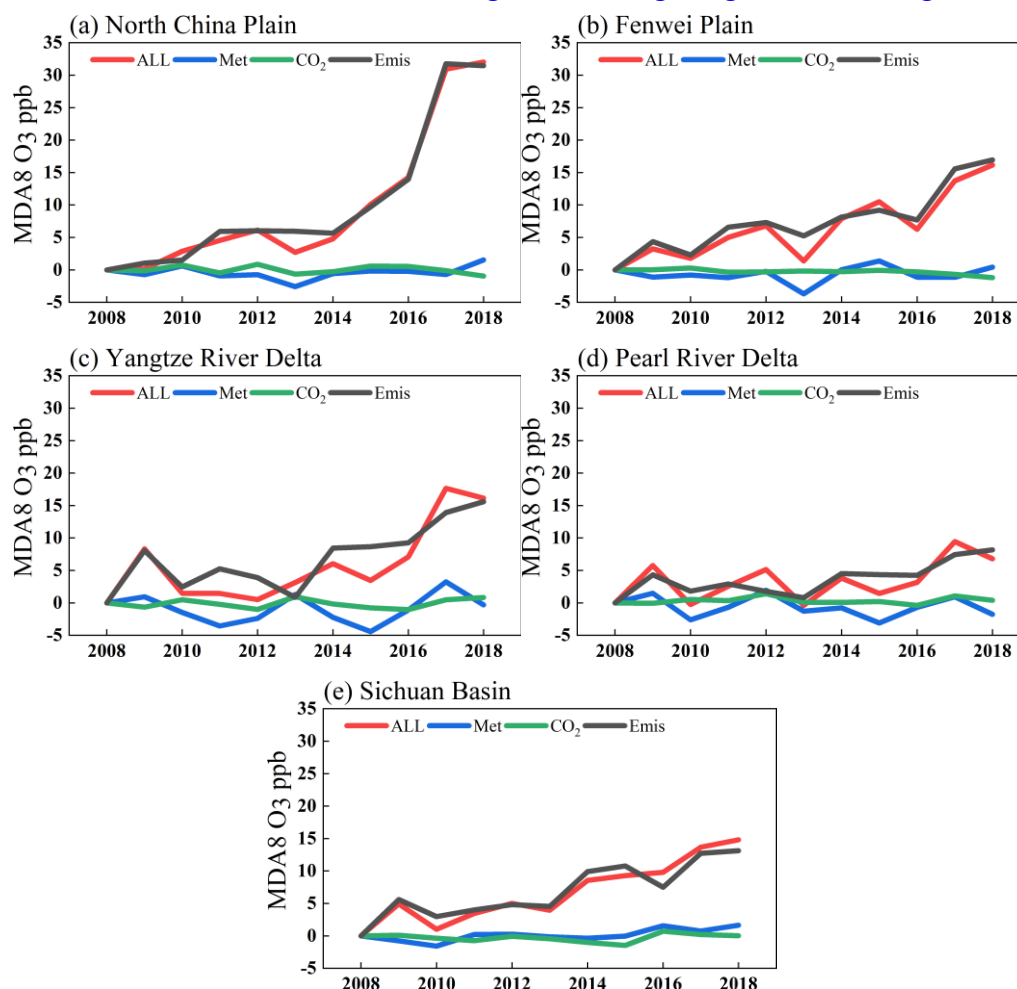


Figure 10. Interannual variations of the surface MDA8 O₃ mixing ratios (units: ppb) in the summer monsoon period (ALL) and the responses of variations in anthropogenic emissions (Emis), meteorological conditions (Met), and CO₂ emissions (CO₂) in (a) North China Plain, (b) Fenwei Plain, (c) Yangtze River Delta, (d) Pearl River Delta, and (e) Sichuan Basin in 2008~2018 relative to 2008.

4. The authors should pay attention to the language- which is often confusing and inaccurate. I have indicated a number of places where this is relevant.

Response: Thanks. We have carefully revised the places you have indicated. The manuscript has been edited by a native English speaker.

5. Figure/tables should be numbered in order of appearance.

Response: Thanks. We have reviewed all figures/tables and confirmed that they are arranged in order of appearance.

6. The new aspect of this paper is the inclusion of effects of CO₂- through affecting the radiative balance, but also through isoprene emissions. However, it is not clear to me how exactly CO₂ has been included in the calculations; what about boundary conditions?

Although the model bias is quite large (1 year offset), it is probably more important that the trends are realistically included and therefore the changes are reflected in the results. One other aspect is that two things are coming together: changes in (regional) radiative forcing, and isoprene emissions, but there is no discussion at all of the magnitude of these effect in the model.

Response: Thanks. CO₂ was added in the model as a tracer. CO₂ boundary conditions were derived from the NOAA CarbonTracker CT2019 dataset. The CCM3 radiation scheme was applied in the model to calculate the radiation of CO₂. The effect of CO₂ on plant isoprene emissions can be simulated in the YIB model.

We added the evaluations of meteorological fields, O₃, and CO₂ from 2015 to 2018.

We did not quantitatively differentiate the impacts of precipitation and isoprene on O₃ concentrations. In Section 3.4, we analyzed the impact of CO₂ on O₃ 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 CO₂ on O₃ concentrations. We have improved the statements in this section.

Changes in manuscript:

2.2 Model description (L127~134): “Xie et al. (2020) further modified the model by incorporating CO₂ as a tracer, which is subject to regulation by sources, sinks, and atmospheric transport processes. The model represents the four sources and sinks of CO₂ as surface fluxes, including emissions from fossil fuels and biomass burning, air-sea CO₂ exchange, and terrestrial biosphere CO₂ fluxes. Additionally, the model incorporated the Yale Interactive Terrestrial Biosphere (YIBs), a land carbon cycle model that enables the simulation of ecological processes, including carbon assimilation, allocation, and autotrophic and heterotrophic respiration (Yue and Unger, 2015).”

2.4 Emissions and Experiment settings (L185~186): “CO₂ emissions and boundary conditions were derived from the NOAA CarbonTracker CT2019 dataset.”

3.1 Model evaluation (L253~261): “Figures S5 and S6 demonstrated that the model accurately reproduced the observed increase in surface CO₂ and O₃ 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 O₃ in major urban areas such as the NCP, the YRD, the PRD, the SCB, and the FWP, while also successfully reproducing the gradient in CO₂ concentrations between eastern and western China. However, the model slightly underpredicted MDA8 O₃ concentrations (-4.02 to -3.21 ppb) and overestimated CO₂ levels (3.32~7.07 ppm). These discrepancies are mainly attributed to uncertainties in the emissions inventory (Wang et al., 2014; Hong et al., 2017; Zhang et al., 2014). Overall, the simulated meteorological factors and surface

CO₂ and O₃ concentrations were deemed acceptable.”

Section 3.4 (L373~385):“CO₂ is a significant driver of climate change and alterations in biogenic emissions. As shown in Figures 6 b and c, the impact of CO₂ on O₃ 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. CO₂ affects O₃ concentration by influencing both precipitation and isoprene emissions. In western and central China, CO₂ primarily affects O₃ concentration through its impact on precipitation (Table 5). Elevated CO₂ concentrations lead to increased precipitation (0.06~0.64 mm/day) in the FWP and SCB regions, resulting in a decrease in surface O₃ (up to -0.51 ppb). In eastern and southern coastal China, where vegetation is abundant, CO₂ has a greater impact on isoprene emissions. In the YRD region, decreased isoprene (-0.58 to -0.32 μg/m³) and increased precipitations (0.09~0.13 mm/day) reduced MDA8 O₃ levels (0.09~0.14 ppb). In PRD, increased isoprene levels (0.31~0.92 μg/m³) and decreased precipitations (-1.02~-0.33 mm/day) led to the enhancement of MDA8 O₃ (0.28~0.46 ppb).”

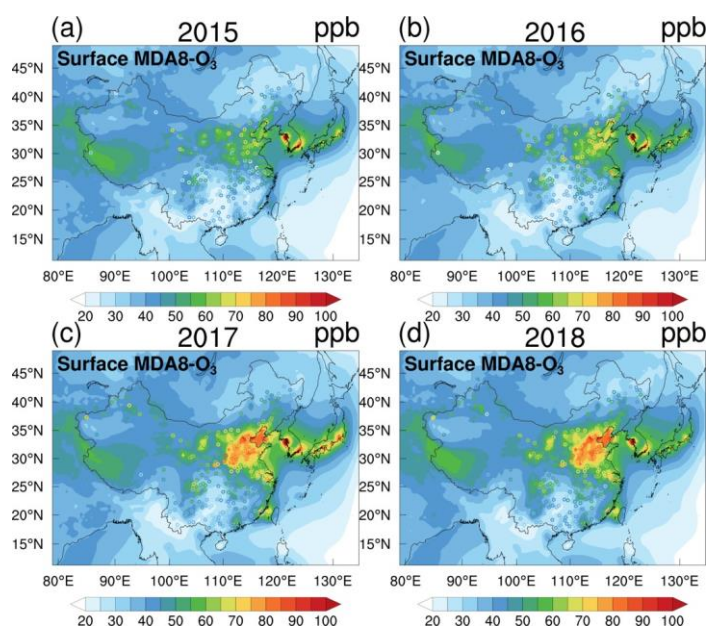


Figure S5. Comparisons between the simulated and observed surface MDA8 O₃ concentrations (units: ppb) during the summer monsoon period in (a)2015, (b)2016, (c)2017, (d)2018. Colored circles represent the observations.

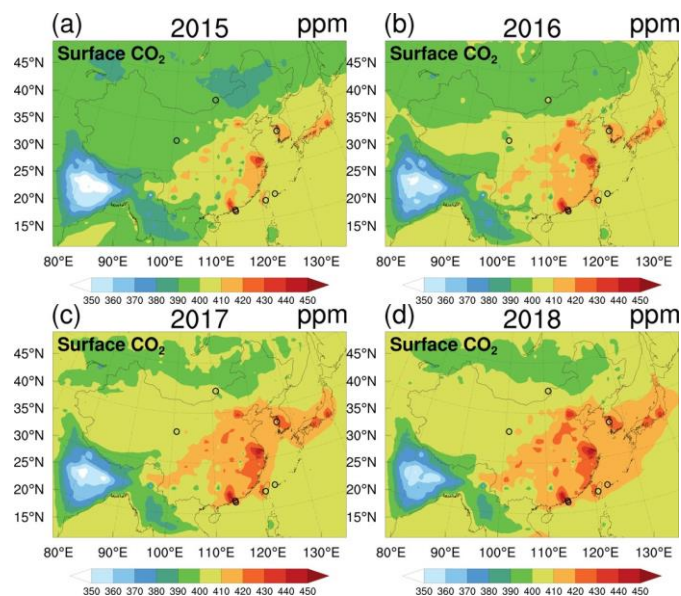


Figure S6. Comparisons between the simulated and observed surface CO₂ concentrations (units: ppm) during the summer monsoon period in (a)2015, (b)2016, (c)2017, (d)2018.

Table 2. Evaluations of the surface CO₂ (units: ppm) and MDA8 O₃ (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 O₃: the maximum daily 8-hour average O₃.

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Yue, X. and Unger, N.: The Yale Interactive terrestrial Biosphere model version 1.0: description, evaluation and implementation into NASA GISS ModelE2, Geoscientific Model Development, <https://doi.org/10.5194/gmd-8-2399-2015>, 2015.

Zhang, H. F., Chen, B. Z., van der Laan-Luijkx, I. T., Chen, J., Xu, G., Yan, J. W., Zhou, L. X., Fukuyama, Y., Tans, P. P., and Peters, W.: Net terrestrial CO₂ exchange over China during 2001-2010 estimated with an ensemble data assimilation system for atmospheric CO₂, Journal of Geophysical Research-Atmospheres, 119, 3500-3515, <https://doi.org/10.1002/2013jd021297>, 2014.

7. The authors mention that one of the improvements is an improved photolysis scheme. One of the post governance effects would be a gradual clean of aerosol (precursor) emissions, and consequently less aerosol scattering and diffusive radiation.

Could the authors elaborate (also in the manuscript) to what extent this included in the manuscript and how it affects the results.

Response: Thanks. We added some descriptions on this issue.

Changes in manuscript:

(L442~448): “Before 2013, the continuous increase in VOCs and NO_x emissions (Figure S8 b, c) facilitated the rise of O₃ levels. Following the implementation of the Clean Air Action Plan in 2013, the emissions of VOCs and NO_x 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 O₃ (Bian et al., 2007). In addition, the reduced NO emission weakened the titration effect (Figure S8 b), thus increasing surface O₃ (Li et al., 2022).”

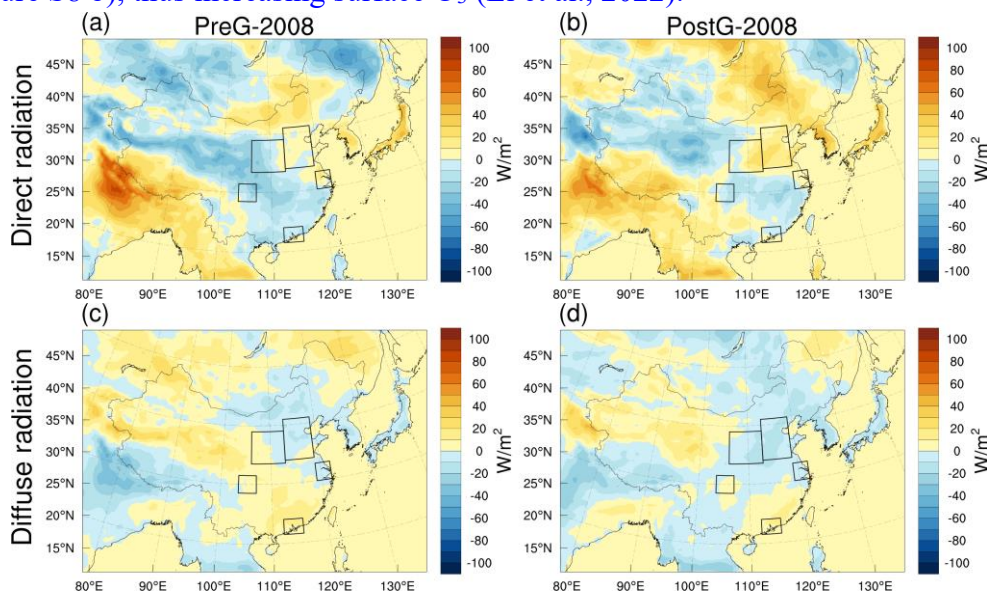


Figure 9. The variations of the surface direct radiation (a,b, units: W/m²) and diffuse radiation

(c,d, units: W/m^2) in the PreG (2009~2013, a,c) and PostG (2014~2018, b,d) period relative to 2008.

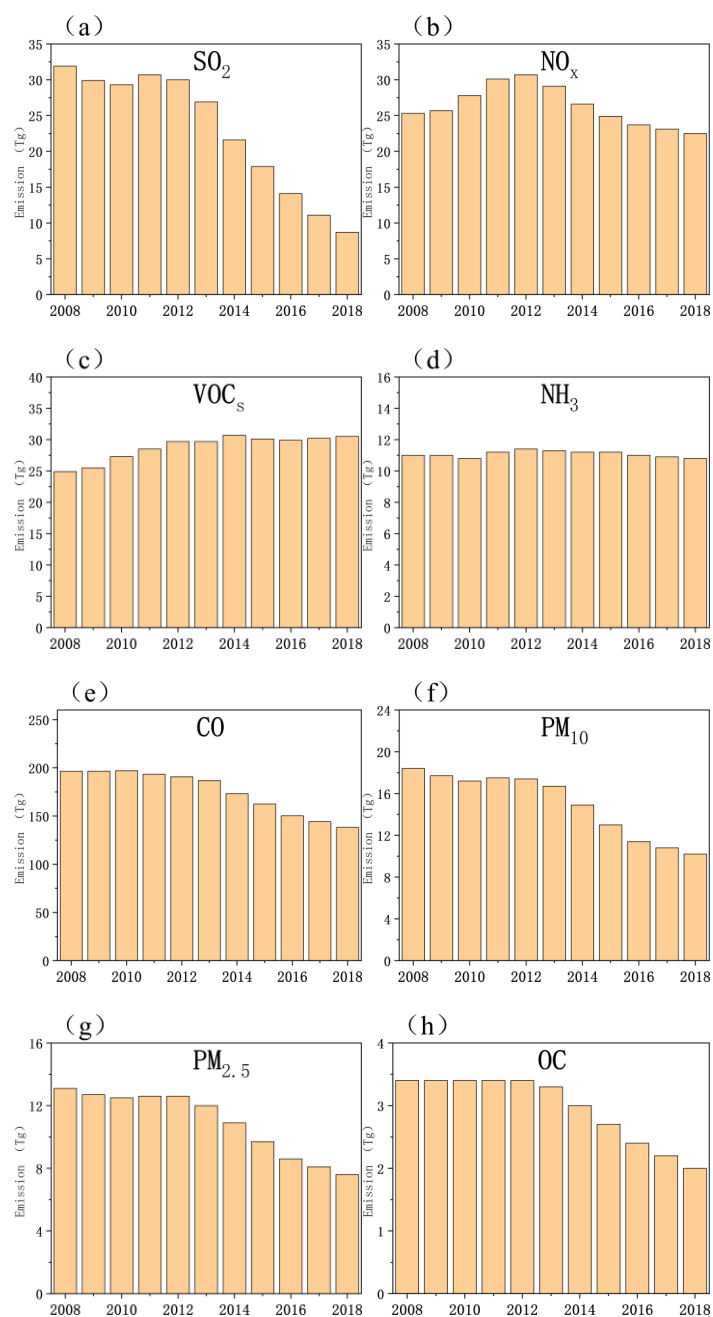


Figure S8. Changes in the anthropogenic emissions (Tg) from 2008 to 2018. The species include (a)SO₂, (b)NO_x, (c)VOCs, (d)NH₃, (e)CO, (f)PM₁₀, (g)PM_{2.5}, (h)OC.

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All taken together I suggest major revisions before this manuscript can be accepted for ACP.

Detailed comments.

20 improved compared to a previous version?

Response: Thanks. We have revised the unclear statement.

Changes in manuscript (L20~21): “Compared to its predecessor, the model has been enhanced concerning the photolysis of O₃ and the radiative impacts of CO₂ and O₃.”

23-24 Examples are not really for an abstract.

Response: Thanks for pointing that out. We have deleted the examples in the abstract.

25 CO₂ includes only radiative effects (in which case it is somewhat like a meteorological). Or also ecophysiological? The effects rather suggest secondary rather than critical.

Response: Thanks. CO₂ alters O₃ concentrations through the combined effects of radiative forcing and emissions of isoprene. We have revised the imprecise statement and elaborated more on the significance of CO₂ in the revised manuscript.

Changes in manuscript:

In Abstract (L25~28) :“Changed CO₂ played a critical role in the variability of O₃ through radiative forcing and isoprene emissions, particularly in southern China, inducing an increase in O₃ 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).”

In Section 3.4 (L386~393): “In some years, the impact of changed CO₂ can be as significant as or even surpass that of anthropogenic emissions and meteorology (Figure 10). For example, in 2013, CO₂ caused an increase of 0.95 ppb in MDA8 O₃ in the YRD region, which exceeded that of anthropogenic emissions (0.87 ppb). Similarly, in the PRD region in 2012, the effect of CO₂, anthropogenic emissions, and meteorology was 1.41, 1.77, and 1.95 ppb, respectively. Even in the NCP in 2010, the impact of CO₂ (0.75 ppb) was comparable to that of anthropogenic emissions (1.5 ppb). In summary, CO₂ has a significant impact on surface O₃ concentrations by influencing radiation and isoprene emissions, with more prominent effects in regions with abundant vegetation.”

26-29 not sure if the inclusion of CO₂ variations is so important.

Response: Thanks. As shown in Table 5, the impact of CO₂ on O₃ levels varies across locations, with a positive effect of 0.28~0.46 ppb along the southeastern coast of

China (PRD) but a negative influence of -0.51 to -0.11 ppb in the southwest and central China (FWP).

Table 5. Simulated responses of MDA8 O₃ mixing ratios (units: ppb), CO₂ mixing ratios (units: ppm), precipitations (units: mm/day), and isoprene mixing ratios to the changes in CO₂ 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

32 'the ecosystem' is a bit general. Rather suggest vegetation growth.

Response: Thanks. We have revised.

Changes in manuscript (L32~33): “O₃ is a strong oxidant detrimental to human health and vegetation growth”.

33 specie=>species or compound. Influencing the earth's radiative balance.

Response: Thanks. We have revised.

Changes in manuscript (L33~34): “it is a crucial active compound influencing the earth's radiative balance”.

40 performed=>issued; initialised.

Response: Thanks. We have revised.

Changes in manuscript (L44~45): “Although the Chinese government initialized the Clean Air Action Plan in 2013 to control air pollution”.

46- clarify that the previous paragraph was about emission.

Response: Thanks. We added some statements about emissions to clarify the previous paragraph was about emissions.

Changes in manuscript:

(L38~39): “With the rapid development in China, emissions of O₃ precursors

have been on the rise, leading to an annual increase in O₃ concentrations since the beginning of the 20th century.”

(L44~47) :“Although the Chinese government initialized the Clean Air Action Plan in 2013 to control air pollution, the concentration of O₃ precursors and PM_{2.5} has significantly decreased (Zhai et al., 2019). However, surface O₃ concentrations continue to increase in major urban areas.”

References

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47 force=>anthropogenic radiative forcer of the climate system.

Response: Thanks. We have revised.

Changes in manuscript (L57): “CO₂ is the primary anthropogenic radiative force of the climate system.”

57-62 Suggest to integrated these sentences in the next paragraph as the connection to CO₂ is not immediately clear.

Response: Thanks. We have revised.

79 should not be negligible=> can be substantial.

Response: Thanks. We have revised.

Changes in manuscript (L80~81): “Meanwhile, the effects of meteorological parameters can be substantial.”

88 sure that emissions have increasing- but here what counts for CO₂ is how concentrations have been increasing?

Response: Thanks. In the preceding sections, we emphasized the impact of changes in CO₂ concentration on O₃ concentration and noted that most studies in recent years have focused solely on the effects of anthropogenic emissions and meteorological conditions on O₃ concentration. In this paragraph, we highlight the continuous increase in CO₂ concentration in China and thus underscore the importance of analyzing the impact of anthropogenic emissions, meteorological factors, and CO₂ on surface O₃ is imperative. We have refined the statements.

Changes in manuscript (L88~96): “Previous studies have mainly focused on the impact of anthropogenic emissions and meteorological factors on the rise of O₃ levels, with limited attention given to the role of CO₂ variations. However, due to the rapid socioeconomic growth in China and the subsequent surge in energy consumption,

CO₂ 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 CO₂ on O₃, it is crucial to evaluate the influence of changes in CO₂ concentration on the maximum daily 8-hour average (MDA8) O₃ concentrations at the surface. Thus, a comprehensive assessment of the impact of anthropogenic emissions, meteorological factors, and CO₂ on surface O₃ is imperative.”

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98-101 ERA-interim is not really observed- it of course assimilates observations, and can be a useful approximation if observations are absent or difficult to retrieve. Explain better the rationale of using ERA-interim. And why in contrast stations were used for ozone. (I guess they were not available to the ECMWF/CAMS global ozone assimilation).

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.

In 2018, the China National Environmental Monitoring Center (CNEMC) operated over 1400 environmental monitoring stations, tracking six pollutants (PM_{2.5}, PM₁₀, SO₂, NO₂, O₃, and CO) since its establishment in 2013. The observational data from CNEMC has been widely employed for model validation of major pollutants in the Chinese region (Wang et al., 2018; Dai et al., 2021; Cheng et al., 2019). Therefore, we utilized data collected at these stations to evaluate O₃ levels.

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119 what is an authentic atmosphere?

Response: Thanks. Sorry for the mistake. We have revised the incorrect statements.

Changes in manuscript (L135~137): “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 actual atmosphere.”

112-127 The model description is rather empty. A short description of the what the two models are doing would be useful (e.g. what chemistry and physics schemes in the RegCM-Chem), what boundary conditions were used? What are the critical weaknesses for this study- that will be addressed in more detail? What are the characteristics of YIB?

Response: Thanks. We added some discussions on model descriptions and boundary conditions. The chemistry and physics schemes, as well as the boundary conditions

used in the model, were presented in Section 2.4: Emissions and Experiment settings. We have moved these statements to Section 2.2: Model Description. The critical weakness in this study was we ignore the influence of boundary conditions on ozone.

Changes in manuscript:

Model descriptions (L121~134): “The RegCM-Chem-YIBs is a regional climate-chemistry-ecology model developed from the RegCM model. RegCM is a regional climate model initially developed by the International Center for Theoretical Physics (ICTP) (Giorgi et al., 2012). Shalaby et al. (2012) integrated the Chem chemistry model into the RegCM model and incorporated the condensed version of the Carbon Bond Mechanism (CBM-Z) to enhance the model's capabilities. To further improve the model's performance, Yin et al. (2015) added a Volatility Basis Set (VBS) scheme to simulate Secondary Organic Aerosols (SOA). Xie et al. (2020) further modified the model by incorporating CO₂ as a tracer, which is subject to regulation by sources, sinks, and atmospheric transport processes. The model represents the four sources and sinks of CO₂ as surface fluxes, including emissions from fossil fuels and biomass burning, air-sea CO₂ exchange, and terrestrial biosphere CO₂ fluxes. Additionally, the model incorporated the Yale Interactive Terrestrial Biosphere (YIBs), a land carbon cycle model that enables the simulation of ecological processes, including carbon assimilation, allocation, and autotrophic and heterotrophic respiration (Yue and Unger, 2015).”

Model schemes (L146~153): “The RegCM model offers a variety of physical and chemical parameterization options. Here, the climatological chemical boundary conditions were driven by the Model of Ozone and Related Chemical Tracers (MOZART). The gas-phase chemistry employed the CBM-Z scheme (Zaveri and Peters, 1999). For the boundary layer scheme, the Holtslag PBL approach was utilized (Khayatanyazdi et al., 2021). The Grell cumulus convection scheme was employed to simulate convective processes (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).”

Emissions and Experiment settings (L185~186): “CO₂ emissions and boundary conditions were derived from the NOAA CarbonTracker CT2019 dataset.”

Weakness (L222~232): “In this work, both meteorological and CO₂ boundary conditions were kept consistent in base and sensitivity studies. We did not consider the impact of boundary conditions on O₃ 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, the 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. Finally, fixed boundary conditions were widely used in some O₃ studies in China (Liu and Wang, 2020a, b; Wang et al., 2019b). Moreover, regional emissions are the primary source of surface O₃ in China, with contributions accounting for 80% from May to August (Lu et al., 2019). Therefore, the impact of fixed boundary conditions can be ignored in the current stage.”

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129 Recommend to use clear version numbers rather than "previous version of xxx".

Response: Thanks. The RegCM-Chem-YIBs model is a collaborative effort developed by our research group in conjunction with the International Center for Theoretical Physics (ICTP) in Italy. No specific version number was assigned to the model, as ongoing improvements are continuously being made.

131 I guess it would be appropriate to give a short description of the radiative transfer scheme; and also of the photolysis scheme? Are these two unified, or separate routines? Are they consistent?

Response: Thanks for pointing that out. We added some descriptions of the model improvement. Please refer to Major comments 1 for a detailed response.

143: not clear Are you including CO₂ as a tracer in the regional model? If so, where do you get the boundary conditions from?

Response: Thanks. CO₂ was added in the model as a tracer. CO₂ boundary conditions were derived from the NOAA CarbonTracker CT2019 dataset. We added some descriptions on this issue.

Changes in manuscript:

2.2 Model description (L127~131): "Xie et al. (2020) further modified the model by incorporating CO₂ as a tracer, which is subject to regulation by sources, sinks, and

atmospheric transport processes. The model represents the four sources and sinks of CO₂ as surface fluxes, including emissions from fossil fuels and biomass burning, air-sea CO₂ exchange, and terrestrial biosphere CO₂ fluxes.”

Emissions and Experiment settings (L185~186): “CO₂ emissions and boundary conditions were derived from the NOAA CarbonTracker CT2019 dataset.”

Figure 2: EAR=>ERA. I assume that the Boundary conditions were taken from ERA interim, but what about the RH? I notice also the much finer resolution of ERA-interim, but it is not clear what the resolution of the regional model was. Both should be given.

Response: Thanks. Sorry for the mistake. “EAR” is replaced by “ERA” in table 2. The initial meteorological boundary data are obtained from the ERA-Interim reanalysis dataset, including temperature, relative humidity, wind etc.

The simulation domain was illustrated in Figure 1, with the target region centered at 36°N and 107°E, and a grid resolution of 60 km by 60 km. The model used 18 vertical levels, ranging from the surface to 50 hPa (L192~194).

The reasons for the 60 km by 60 km grids applied for this investigation are multifaceted. First, the modeling domain was extensive and covered all East Asia regions, which was 6720 km × 4800 km. Second, The regional climate-chemical model (RegCM-Chem) and ecological model (YIBs) were fully coupled to consider the interactions between atmospheric composition and terrestrial ecosystem. The meteorological factors and the concentrations of air pollutants output by RegCM-Chem are input into YIBs to simulate the physiological processes of vegetation and calculate land surface parameters such as carbon dioxide flux, BVOC emissions, and stomatal conductance of the terrestrial ecosystem. Conversely, the simulations of the YIBs model are taken into the RegCM-Chem model to adjust the air qualities, temperature, humidity, circulation, and other meteorological fields (Xie et al., 2018; Xie et al., 2019). Therefore, extensive computing resources are required for the model. In addition, the 60 by 60 km grids were widely used in the RegCM-Chem-YIBs model, which has been proven to capture East Asia's climate and air pollution features (Pu et al., 2017; Xie et al., 2019; Xu et al., 2022; Zhuang et al., 2018). Therefore, the grid independence test was not performed in this study.

Changes in manuscript (L186~189): “The initial meteorological boundary data, such as temperature, relative humidity, and wind, are derived from the ERA-Interim reanalysis dataset with a horizontal resolution of 0.125°, a temporal resolution of 6 hours, and 37 vertical levels.”

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197 and table 2: Explain what is MDA8? What is the evaluation period (summer 2018?). I notice that the difference of CO₂ between the simulations and the OBS are really high and could be much better if the boundary were taken into account appropriately into account. Perhaps it is not a super-critical issue as long as the CO₂ trends are appropriately taken into account- but this is to proven.

Response: Thanks. We added the explanation of MDA8 and the evaluation period in Table 2.”

We also added the evaluations of O₃ and CO₂ from 2015 to 2018. The CO₂ trends were well simulated by the model. Please refer to Major comments 6 for a detailed response.

Table 2. Evaluations of the surface CO₂ (units: ppm) and MDA8 O₃ (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 O₃: the maximum daily 8-hour average O₃.

215 also high temperatures and humidities are conducive to ozone production.

Response: Thanks. We added the statement and references on temperatures and humidities.

Changes in manuscript (L269~271): “High O₃ concentrations appeared in eastern China, which can be attributed to increased emissions, high temperatures, humidities, and intense radiation in the region (Gao et al., 2020; Mousavinezhad et al., 2021; Wei et al., 2022)”.

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218 Can these regions be graphically shown, together with some qualitative information? I have no clue what/where is: NCP, the YRD, the PRD, the SCB, and the FWP

Response: Thanks. The locations of the NCP, the YRD, the PRD, the SCB, and the FWP regions are illustrated in Figure 1. We have added black boundaries in all Figures to delineate the boundaries of the respective regions.

Changes in manuscript:

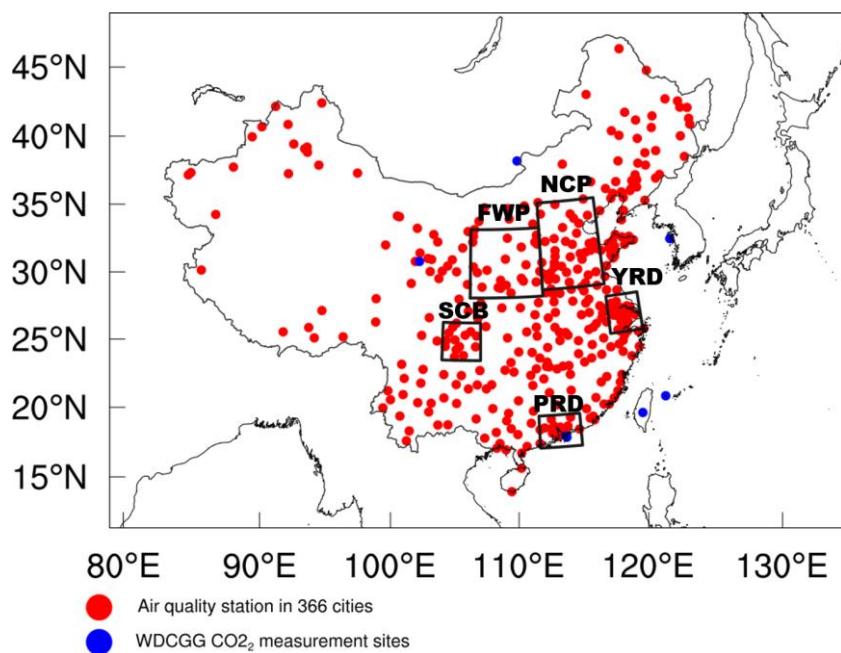


Figure 1. Model domains for the RegCM-Chem-YIBs model. The regions with black boundaries are the North China Plain (34~41°N, 113~119°E), the Yangtze River Delta (30~33°N, 119~122°E), the Pearl River Delta (21.5~24°N, 112~115.5°E), the Sichuan Basin (28.5~31.5°N, 103.5~107°E), and the Fenwei Plain (33.5~39°N,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

Taking Figure 2 as an example:

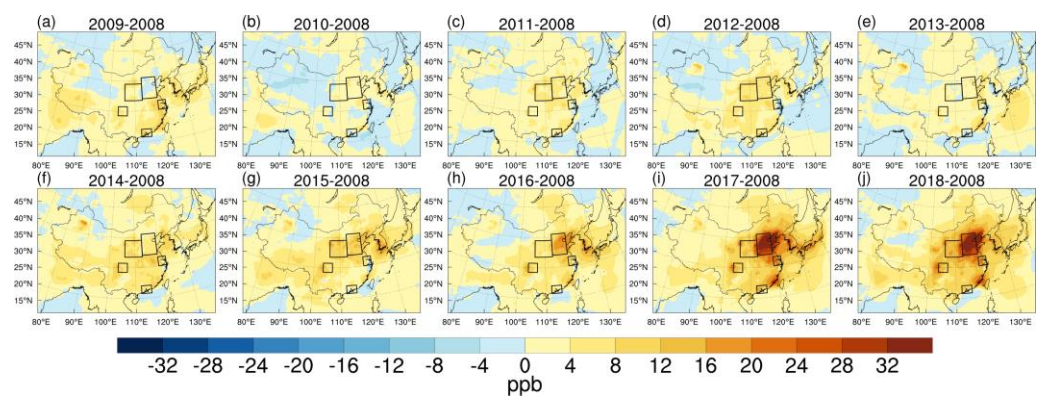


Figure 2. Changes in the surface MDA8 O₃ concentrations (units: ppb) during the summer monsoon period from 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

217 The ozone seems to increase mostly in 2017 and 2018. While the 10 plots are useful, I could suggest to use the target analysis regions (tables), and show also average concentration change as a function of year (lines).

Response: Thanks. We have added Table 3 to display the changes in MDA8 O₃ from 2009 to 2018 relative to 2008. The average concentration change as a function from 2008 to 2018 has presented in Figure 10 (lines). We added some descriptions for Table 3.

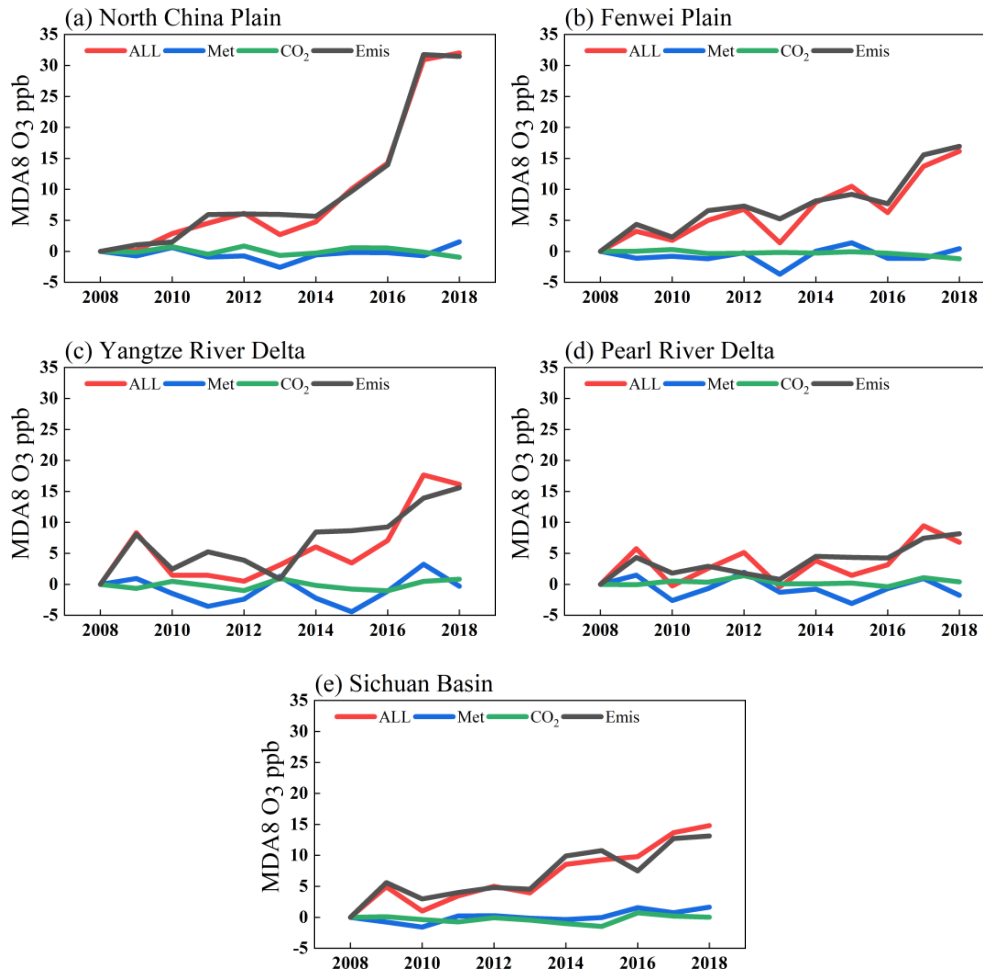


Figure 10. Interannual variations of the surface MDA8 O₃ mixing ratios (units: ppb) in the summer monsoon period (ALL) and the responses of variations in anthropogenic emissions (Emis), meteorological conditions (Met), and CO₂ emissions (CO₂) in (a) North China Plain, (b) Fenwei Plain, (c) Yangtze River Delta, (d) Pearl River Delta, and (e) Sichuan Basin in 2008~2018 relative to 2008.

Changes in manuscript (L283~291):

Figure 2 and Table 3 illustrate the changes in surface MDA8 O₃ concentrations from 2009 to 2018 relative to 2008. The surface MDA8 O₃ concentrations in China increased drastically over the past decade, particularly in 2017 and 2018 (6.79~32.03 ppb). We divided the period from 2009 to 2018 into two phases based on the Clean Air Action Plan implemented in 2013: the pre-governance period (PreG, 2009~2013) and the post-governance period (PostG, 2014~2018). The surface MDA8 O₃ concentration increased significantly in NCP (18.42 ppb), followed by SCB (11.21 ppb), FWP (10.9 ppb), and the YRD (10.07 ppb), while increased slightly in PRD (4.94 ppb), in PosG relative to 2008. Our results were consistent with previous studies by Lu et al. (2020), Ma et al. (2016), and Mousavinezhad et al. (2021).

Table 3. The changes of MDA8 O₃ over North China Plain, Fenwei Plain, Yangtze River Delta, Pearl River Delta, and Sichuan Basin during the summer monsoon period from 2009 to

2018 relative to 2008.

Regions	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	PreG	PostG
NCP	0.14	2.85	4.53	6.13	2.7	4.78	10.1	14.25	30.92	32.03	3.27	18.42
FWP	3.23	1.78	5.01	6.78	1.37	7.9	10.5	6.24	13.71	16.17	3.63	10.90
YRD	8.33	1.47	1.46	0.5	3.12	6.04	3.46	7.09	17.64	16.12	2.98	10.07
PRD	5.76	-0.26	2.56	5.13	-0.4	3.82	1.46	3.16	9.45	6.79	2.56	4.94
SCB	4.92	1.03	3.46	5	3.94	8.54	9.27	9.78	13.67	14.8	3.67	11.21

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232 If table 5 is discussed first, it should be placed before. Tables (and figures) appear in order of discussion. Also I guess these are reponses in ppb not ppb per year? Table is not clear about units.

Response: Thanks. According to our model, we derived the impacts of meteorological factors and CO₂ variations on the changes in O₃ concentration from 2008 to 2018. Subsequently, we calculated the contribution of anthropogenic emissions to the changes in O₃ by subtracting the effects of meteorological factors and CO₂ variations from the total changes in O₃ concentration. Therefore, Table 5 appeared at the end. Here, we only referenced some of the partial results from Table 5.

Thanks for pointing out that, we made an error in our unit conversion. We should use ppb instead of ppb per year. We have added units to all tables and corrected the wrong units used in the manuscript.

243 What is meant with: "The meteorological factors were generally unfavorable to O₃ formation during the study period". I think they are very favourable. But I guess the authors intend to say that in general changes in meteo conditions, led to overall (small) changes in O₃. Of course using one reference year for meteorology is somewhat misleading (figure 4). Also Figure 6 shows that some of the changes are quite determined by the year 2008 (especially for precipitation).

Response: Thanks. Sorry for the mistake. We have revised the ambiguous expression. The reasons for using 2008 as a reference year have been addressed in the response to Major comments 2.

Changes in manuscript (L300~301):

“Overall, the meteorological variations from 2008 to 2018 were unfavorable for the O₃ increase during the EASM period, as illustrated in Figure 3.”

245-253: which table are these results?

Response: Thanks. We have added information on the origins of the numerical data.

Changes in manuscript (L302): “Based on Figure 3 and Table 4”.

256 changes in meteo factors are unfavorable (not the meteo itself).

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

Changes in manuscript (L313~316): “Chen et al. (2019) and Liu and Wang (2020a) also suggested that changed meteorological conditions had a negative impact on O₃ formation in the NCP and FWP regions, and that the influence of meteorology on surface-level O₃ decreased in PostG.”

288 the distinction of preG and postG needs to be revised to tell variability from signal.

Response: Thanks. We added the descriptions of PreG and PostG in all Tables.

Changes in manuscript:

Taking Table 3 as an example:

Table 4. Response of the MDA8 O₃ mixing ratios (units: ppb), precipitations (units: mm/day), clouds fraction (units: %), shortwave flux (units: W/m²), wind speed (units: m/s), temperature (units: K), and planetary boundary layer height (units: m) to the changes in meteorological conditions over North China Plain, Fenwei Plain, Yangtze River Delta, Pearl River Delta, and Sichuan Basin during the summer monsoon period in PreG (2009~2013) and PostG (2014~2018) relative to 2008.

Regions	Period	MDA8 O ₃ (ppb)	Precip (mm/day)	Clouds (%)	SWF (W/m ²)	Wind Speed (m/s)	Temp (K)	PBL (m)
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

293 driver of climate *change*.

Response: Thanks for pointing that out. We have revised.

Changes in manuscript (L373): “CO₂ is a significant driver of climate change and alterations in biogenic emissions.”

321 give units also in figure caption (hard to read).

Response: Thanks. We have added units in all figure/Table captions.

325 units?

Response: Thanks. We have added units to all tables/figures and their captions.

351 multiplied=>multiple

Thanks. We have revised.

Changes in manuscript (L442): “The reasons for this characteristic are multiple.”

366 give units.

Response: Thanks. We have added units to all tables/figures and their captions.

373 Suggest to have a separate section that discusses Figure 11, and also gives an attribution. (is the red line fully explainable by the blue, green and black line).

Response: Thanks. We added Section 3.6: Attribution analysis of ozone changes in 2008~2018. Please refer to Major comments 3 for a detailed response.

377 this is not an uncertainty analysis, rather a discussion of why boundary conditions were kept the same in the sensitivity studies. (I guess CO₂ was however not kept constant). This information should be given much earlier.

A uncertainty analysis could try to explain how this would affect the results. And there are other factors as well that need to be discussed.

Response: Thanks. We have moved this section to Section 2.4: Emissions and Experiment settings. Both meteorological and CO₂ boundary conditions were kept consistent in base and sensitivity studies. We added some discussions on this issue.

Changes in manuscript (L222~223): “In this work, both meteorological and CO₂ boundary conditions were kept consistent in base and sensitivity studies.”