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 #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 CO_2 (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 CO_2 as a tracer, which is subject to regulation by sources, sinks, and atmospheric transport processes. The model represents the four sources and sinks of CO_2 as surface fluxes, including emissions from fossil fuels and biomass burning, air-sea CO_2 exchange, and terrestrial biosphere CO_2 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 (Khayatianyazdi 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 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."

Emissions and Experiment settings (L185~186): "CO_2 emissions and boundary conditions were derived from the NOAA CarbonTracker CT2019 dataset.”

References

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2. The use of a single year (2008) as a reference year for pregovernance 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 CO_2 emissions. Meteorological conditions (CO_2 emissions) were kept constant at 2008 levels over ten years, referred to as the $SIM_{MET=2008}$ ($SIM_{CO2=2008}$) experiment.
The changes in O_3 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 O_3 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 CO_2 emissions was similarly estimated (Eq. (3)). Finally, the influence of anthropogenic emissions was calculated by excluding the impact of meteorological factors and CO_2 from the changes in O_3 concentrations (Eq. (4)). Table 1 shows the results of the numerical experiments."

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

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

$$\Delta C_i = SIM_i - SIM_{i,CO2=2008} \quad (3)$$

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

ΔO_i : The changes in O_3 concentrations in the year i relative to 2008.
$Base_i$: The O_3 concentrations in the Base experiment in the year i.
ΔM_i : The changes in O_3 concentrations in the year i due to meteorological factors variations.
$SIMi,_{MET=2008}$: The O_3 concentrations in the $SIM_{MET=2008}$ experiment in the year i.

ΔC_i : The changes in O_3 concentrations in the year i due to CO_2 variations.
$SIM_{i,CO2=2008}$: The O_3 concentrations in the $SIM_{CO2=2008}$ experiment in the year i.
ΔE_i : The changes in O_3 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
$SIM_{CO2=2008}$		Varying	2008	Varying

3. I suggest re-ordering of sections to describe effect of emissions; effects of meteorology; and CO_2 effect in the order of importance. A 4 section could be added describing the addivity.

Response: Thanks. As described in the previous question (2), we first obtained the contributions of meteorological and CO_2 changes to O_3 concentrations. Then, the influence of anthropogenic emissions was calculated by excluding the impact of meteorological factors and CO_2 from the changes in O_3 concentrations. Therefore, we conducted a two-step analysis that first evaluated the effects of meteorology and CO_2 on O_3 changes, followed by an analysis of the effects of anthropogenic emissions on O_3 concentrations. In addition, we have added a new section 3.6 to conduct an attribution analysis of changes in O_3 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 O_3 concentration from 2008 to 2018. The total variation in O_3 concentration can be attributed to the combined effects of meteorological changes, changes in CO_2 concentration, and anthropogenic emissions (Figure 10).

The primary driver of the O_3 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 O_3 increase. Even in the PostG period, with the development of urbanization and industrialization, the impact of changed anthropogenic emissions on O_3 has gradually become more prominent than changed meteorology and CO_2. The contribution of changed meteorology to O_3 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 CO_2

concentration affected O_3 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 CO_2 concentration changes on O_3 concentration should be considered in regions with high vegetation coverage."

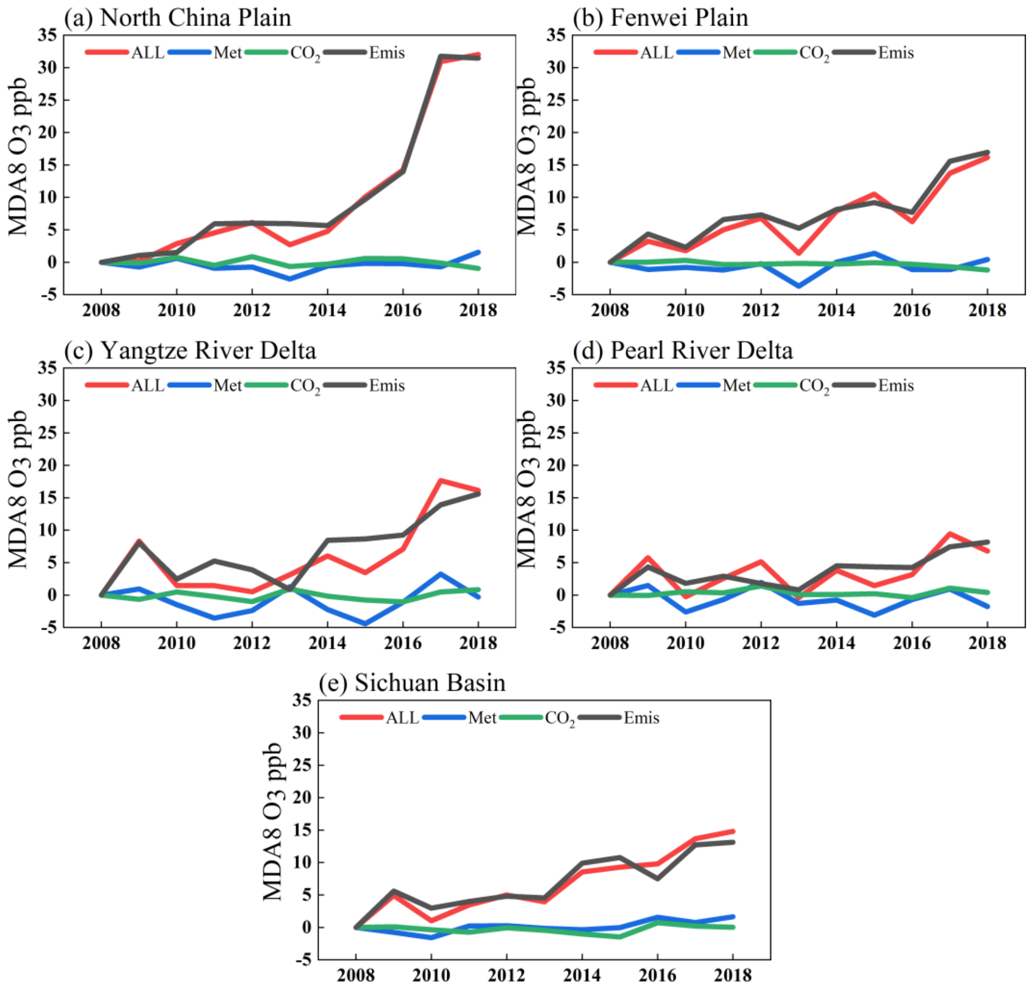

Figure 10. Interannual variations of the surface MDA8 O_3 mixing ratios (units: ppb) in the summer monsoon period (ALL) and the responses of variations in anthropogenic emissions (Emis), meteorological conditions (Met), and CO_2 emissions (CO_2) in (a) North China Plain, (b) Fenwei Plain, (c) Yangtze River Delta, (d) Pearl River Delta, and (e) Sichuan Basin in 2008~2018 relative to 2008.

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 CO_2- through affecting the radiative balance, but also through isoprene emissions. However, it is not clear to me how exactly CO_2 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 realisticly 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. CO_2 was added in the model as a tracer. CO_2 boundary conditions were derived from the NOAA CarbonTracker CT2019 dataset. The CCM3 radiation scheme was applied in the model to calculate the radiation of CO_2. The effect of CO_2 on plant isoprene emissions can be simulated in the YIB model.

We added the evaluations of meteorological fields, O_3, and CO_2 from 2015 to 2018.

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:

2.2 Model description (L127~134): "Xie et al. (2020) further modified the model by incorporating CO_2 as a tracer, which is subject to regulation by sources, sinks, and atmospheric transport processes. The model represents the four sources and sinks of CO_2 as surface fluxes, including emissions from fossil fuels and biomass burning, air-sea CO_2 exchange, and terrestrial biosphere CO_2 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): "CO_2 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 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 (Wang et al., 2014; Hong et al., 2017; Zhang et al., 2014). Overall, the simulated meteorological factors and surface

CO_2 and O_3 concentrations were deemed acceptable."

Section 3.4 (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)."

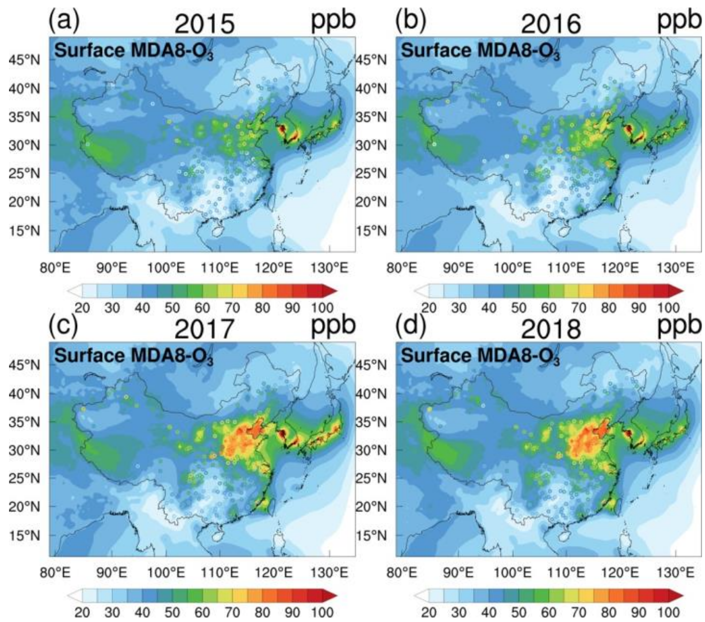

Figure S5. Comparisons between the simulated and observed surface MDA8 O_3 concentrations (units: ppb) during the summer monsoon period in (a)2015, (b)2016, (c)2017, (d)2018. Colored circles represent the observations.

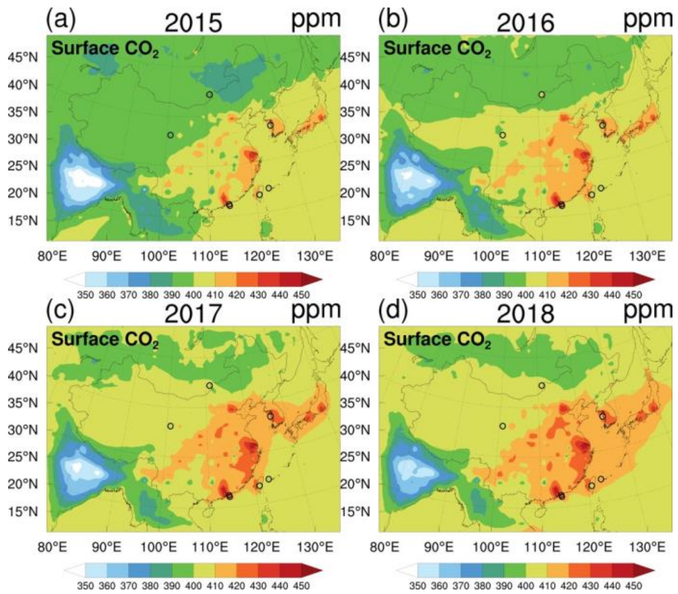

Figure S6. Comparisons between the simulated and observed surface CO_2 concentrations (units: ppm) during the summer monsoon period in (a)2015, (b)2016, (c)2017, (d)2018.

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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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 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)."

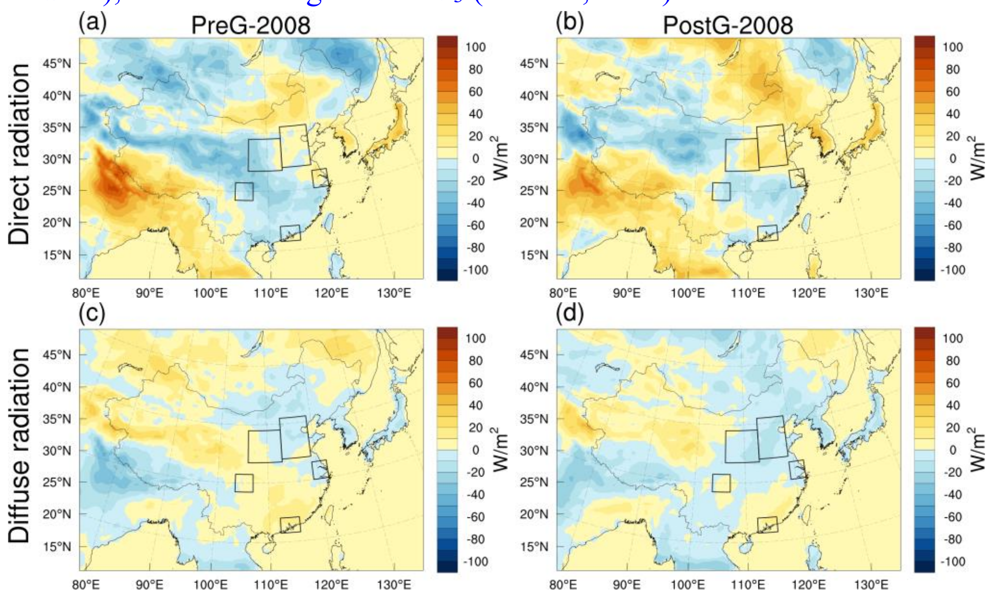

Figure 9. The variations of the surface direct radiation (a,b, units: W/m^2) and diffuse radiation

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

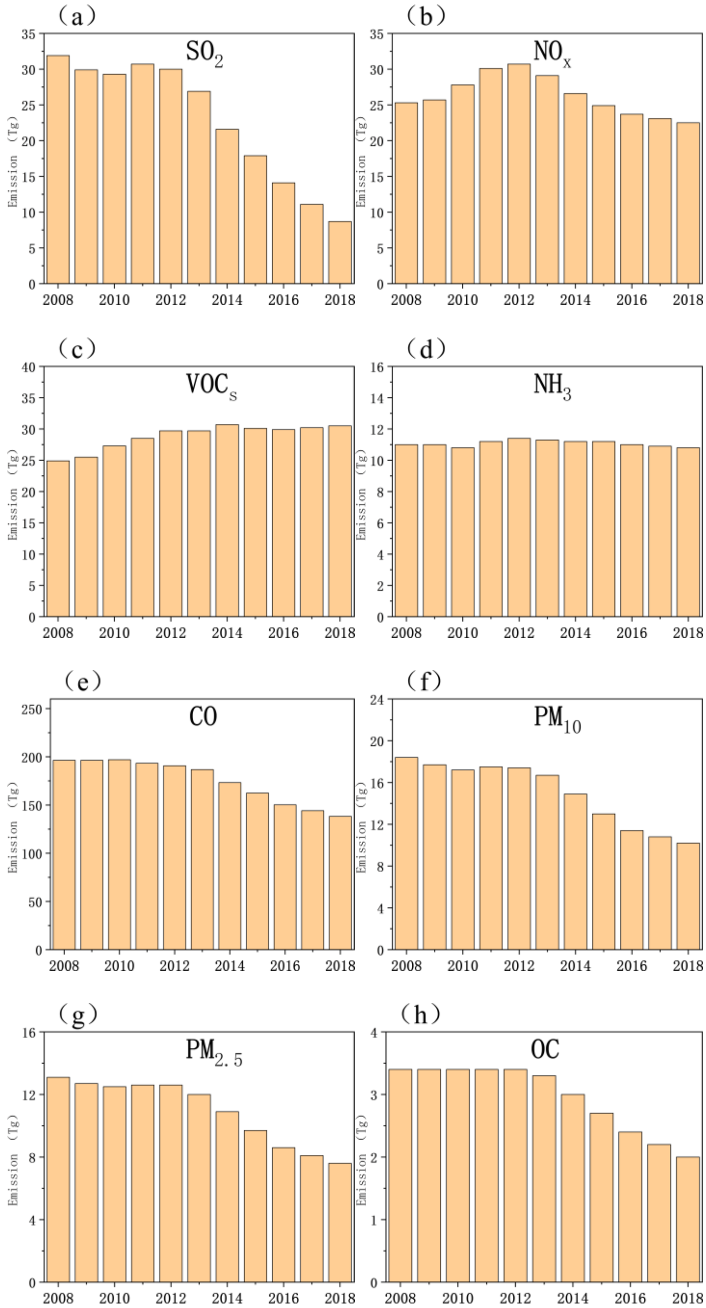

Figure S8. Changes in the anthropogenic emissions (Tg) from 2008 to 2018. The species include (a)SO_2, (b)NOx, (c)VOCs, (d)NH_3, (e)CO, (f)PM_{10}, (g)$PM_{2.5}$, (h)OC.

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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 O_3 and the radiative impacts of CO_2 and O_3."

23-24 Examples are not really for an abstract.
Response: Thanks for pointing that out. We have deleted the examples in the abstract.

25 CO_2 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. CO_2 alters O_3 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 CO_2 in the revised manuscript.

Changes in manuscript:

In 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)."

In 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."

26-29 not sure if the inclusion of CO_2 variations is so important.
Response: Thanks. As shown in Table 5, the impact of CO_2 on O_3 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 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

32 'the ecosystem' is a bit general. Rather suggest vegetation growth.
Response: Thanks. We have revised.

Changes in manuscript (L32~33): "O_3 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 O_3 precursors have been on the rise, leading to an annual increase in O_3 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 O_3 precursors and $PM_{2.5}$ has significantly decreased (Zhai et al., 2019). However, surface O_3 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): "CO_2 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 CO_2 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 CO_2 is how concentrations have been increasing?

Response: Thanks. In the preceding sections, we emphasized the impact of changes in CO_2 concentration on O_3 concentration and noted that most studies in recent years have focused solely on the effects of anthropogenic emissions and meteorological conditions on O_3 concentration. In this paragraph, we highlight the continuous increase in CO_2 concentration in China and thus underscore the importance of analyzing the impact of anthropogenic emissions, meteorological factors, and CO_2 on surface O_3 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 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."

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Ren, S. G., Yuan, B. L., Ma, X., and Chen, X. H.: International trade, FDI (foreign direct investment) and embodied CO2 emissions: A case study of Chinas industrial sectors, China Economic Review, 28, 123-134,https://doi.org/10.1016/j.chieco.2014.01.003, 2014.

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 ($PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, 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 O_3 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 CO_2 as a tracer, which is subject to regulation by sources, sinks, and atmospheric transport processes. The model represents the four sources and sinks of CO_2 as surface fluxes, including emissions from fossil fuels and biomass burning, air-sea CO_2 exchange, and terrestrial biosphere CO_2 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 (Khayatianyazdi 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): "CO_2 emissions and boundary conditions were derived from the NOAA CarbonTracker CT2019 dataset."

Weakness (L222~232): "In this work, both meteorological and CO_2 boundary conditions were kept consistent in base and sensitivity studies. We did not consider the impact of boundary conditions on O_3 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 O_3 studies in China (Liu and Wang, 2020a, b; Wang et al., 2019b). Moreover, regional emissions are the primary source of surface O_3 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 approriate 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 CO_2 as a tracer in the regional model? If so, where do you get the boundary conditions from?
Response: Thanks. CO_2 was added in the model as a tracer. CO_2 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 CO_2 as a tracer, which is subject to regulation by sources, sinks, and

atmospheric transport processes. The model represents the four sources and sinks of CO_2 as surface fluxes, including emissions from fossil fuels and biomass burning, air-sea CO_2 exchange, and terrestrial biosphere CO_2 fluxes."

Emissions and Experiment settings (L185~186): "CO_2 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 reigonal 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 gird 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 CO_2 between the simulations and the OBS are really highand 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 CO_2 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 O_3 and CO_2 from 2015 to 2018. The CO_2 trends were well simulated by the model. Please refer to Major comments 6 for a detailed response.

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.

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

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

Changes in manuscript (L269~271): "High O_3 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)".

References

Gao, M., Gao, J., Zhu, B., Kumar, R., Lu, X., Song, S., Zhang, Y., Jia, B., Wang, P., Beig, G., Hu, J., Ying, Q., Zhang, H., Sherman, P., and McElroy, M. B.: Ozone pollution over China and India: seasonality and sources, Atmospheric Chemistry and Physics, 20, 4399-4414,https://doi.org/10.5194/acp-20-4399-2020, 2020.

Mousavinezhad, S., Choi, Y., Pouyaei, A., Ghahremanloo, M., and Nelson, D. L.: A comprehensive investigation of surface ozone pollution in China, 2015-2019: Separating the contributions from meteorology and precursor emissions, Atmospheric Research, 257,https://doi.org/10.1016/j.atmosres.2021.105599, 2021.

Wei, J., Li, Z. Q., Li, K., Dickerson, R. R., Pinker, R. T., Wang, J., Liu, X., Sun, L., Xue, W. H., and Cribb, M.: Full-coverage mapping and spatiotemporal variations of ground-level ozone (O3) pollution from 2013 to 2020 across China, Remote Sensing of Environment, 270,https://doi.org/10.1016/j.rse.2021.112775, 2022.

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:

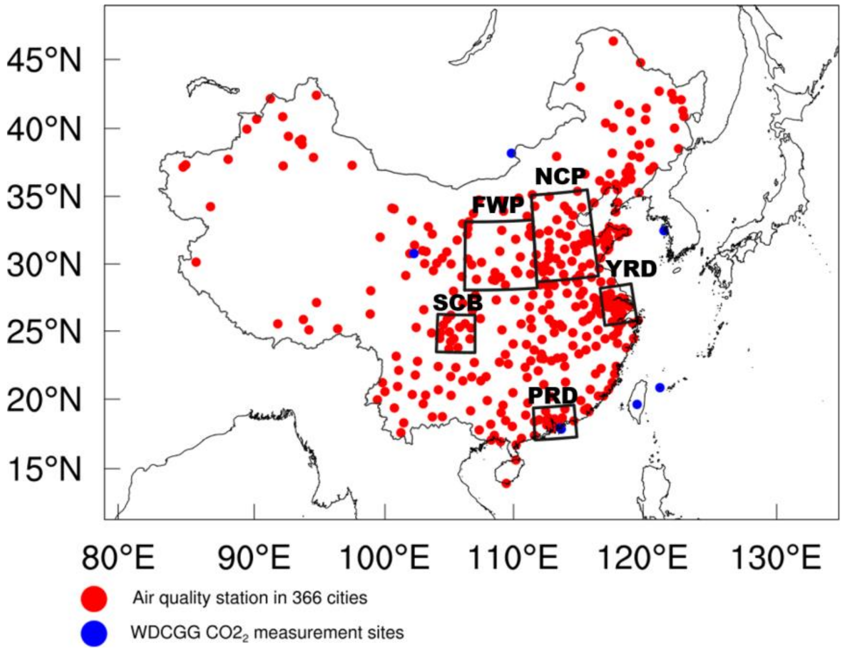

Figure 1. Model domains for the RegCM-Chem-YIBs model. The regions with black boundaries are the North China Plain (34~41°N, 113~119°E), the Yangtze River Delta (30~33°N, 119~122°E), the Pearl River Delta (21.5~24°N, 112~115.5°E), the Sichuan Basin (28.5~31.5°N, 103.5~107°E), and the Fenwei Plain (33.5~39°N,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

Taking Figure 2 as an example:

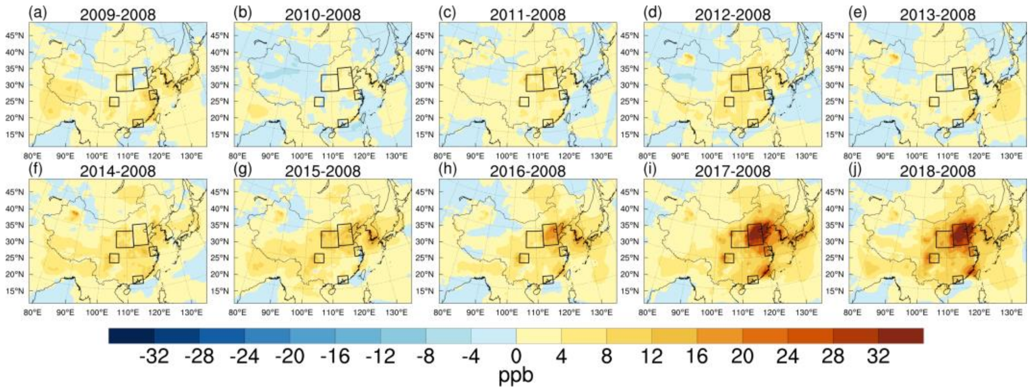

Figure 2. Changes in the surface MDA8 O_3 concentrations (units: ppb) during the summer monsoon period from 2009 (a), 2010 (b), 2011 (c), 2012 (d), 2013 (e), 2014 (f), 2015 (g), 2016 (h), 2017 (i) and 2018 (j) relative to 2008.

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 fucntion of year (lines).

Response: Thanks. We have added Table 3 to display the changes in MDA8 O_3 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.

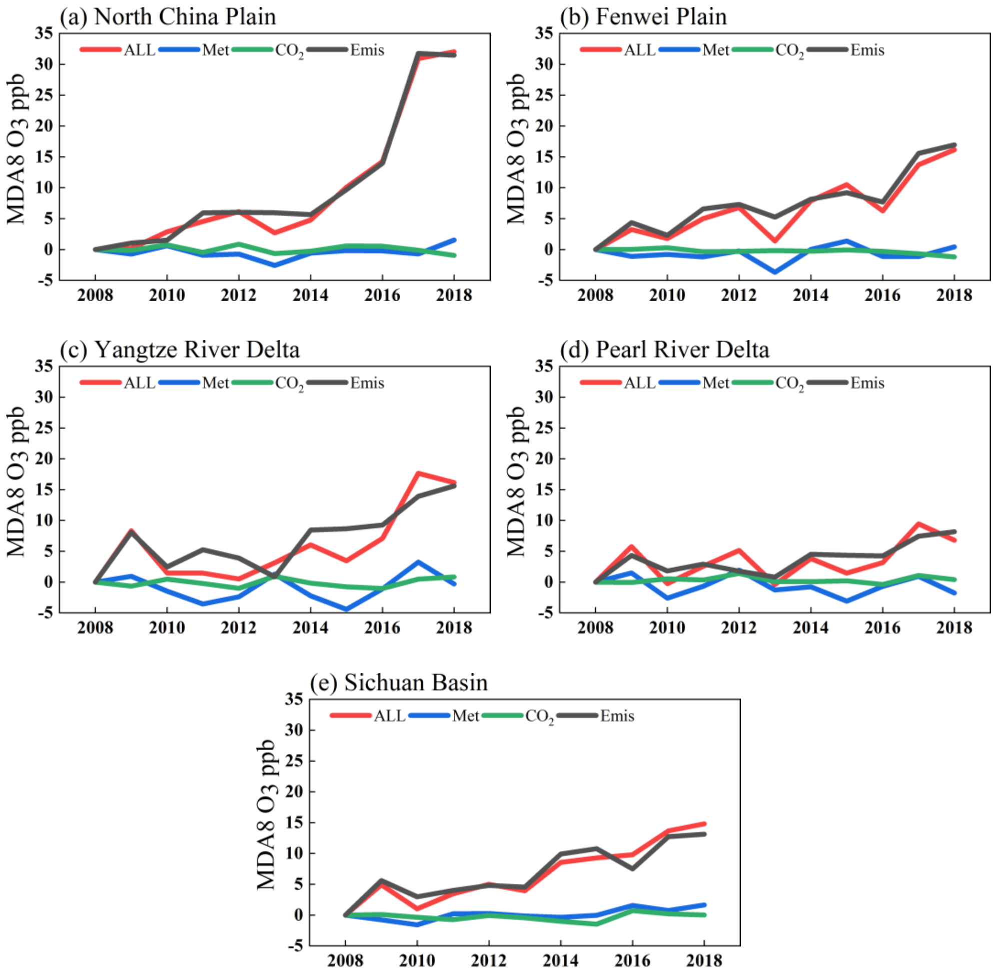

Figure 10. Interannual variations of the surface MDA8 O_3 mixing ratios (units: ppb) in the summer monsoon period (ALL) and the responses of variations in anthropogenic emissions (Emis), meteorological conditions (Met), and CO_2 emissions (CO_2) in (a) North China Plain, (b) Fenwei Plain, (c) Yangtze River Delta, (d) Pearl River Delta, and (e) Sichuan Basin in 2008~2018 relative to 2008.

Changes in manuscript (L283~291):

Figure 2 and Table 3 illustrate the changes in surface MDA8 O_3 concentrations from 2009 to 2018 relative to 2008. The surface MDA8 O_3 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 O_3 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 O_3 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

References

Lu, X., Zhang, L., Wang, X. L., Gao, M., Li, K., Zhang, Y. Z., Yue, X., and Zhang, Y. H.: Rapid Increases in Warm-Season Surface Ozone and Resulting Health Impact in China Since 2013, Environmental Science & Technology Letters, 7, 240-247,https://doi.org/10.1021/acs.estlett.0c00171, 2020.

Ma, Z., Xu, J., Quan, W., Zhang, Z., Lin, W., and Xu, X.: Significant increase of surface ozone at a rural site, north of eastern China, Atmospheric Chemistry and Physics, 16, 3969-3977,https://doi.org/10.5194/acp-16-3969-2016, 2016.

Mousavinezhad, S., Choi, Y., Pouyaei, A., Ghahremanloo, M., and Nelson, D. L.: A comprehensive investigation of surface ozone pollution in China, 2015-2019: Separating the contributions from meteorology and precursor emissions, Atmospheric Research, 257,https://doi.org/10.1016/j.atmosres.2021.105599, 2021.

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 CO_2 variations on the changes in O_3 concentration from 2008 to 2018. Subsequently, we calculated the contribution of anthropogenic emissions to the changes in O_3 by subtracting the effects of meteorological factors and CO_2 variations from the total changes in O_3 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 O_3 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 O_3. 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 O_3 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 O_3 formation in the NCP and FWP regions, and that the influence of meteorology on surface-level O_3 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 O_3 mixing ratios (units: ppb), precipitations (units: mm/day), clouds fraction (units: %), shortwave flux (units: W/m^2), 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): "CO_2 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 seperate 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 CO_2 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 CO_2 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 CO_2 boundary conditions were kept consistent in base and sensitivity studies."